

**Proceedings of the  
THIRD INTERNATIONAL CONGRESS  
of the  
INTERNATIONAL RADIATION  
PROTECTION ASSOCIATION**

**September 9-14, 1973  
Washington, D.C.**



**INTERNATIONAL RADIATION PROTECTION ASSOCIATION**

**U. S. ATOMIC ENERGY COMMISSION  
Office of Information Services  
Technical Information Center  
Oak Ridge, Tennessee**

CONF 730907 P1

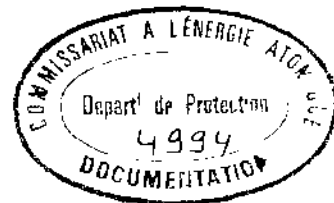
Distribution Category UC 41

THIRD INTERNATIONAL CONGRESS OF THE  
INTERNATIONAL RADIATION PROTECTION ASSOCIATION

Held at the Washington Hilton Hotel  
Washington D C September 9-14 1973

Editor

W S Snyder



February 1974

## PREFACE

The Third International Congress of the International Radiation Protection Association (IRPA) was held in Washington, D.C., September 9-14, 1973, and was hosted by the Health Physics Society. Two previous Congresses were held in Rome, Italy, in September 1966 and Brighton, England, in May 1970.

The scientific program, consisting of 237 papers, was truly international in scope. Scientists and administrators of 33 countries from all corners of the world actively participated in the Congress. In addition to the scientific program, refresher courses were held as part of a continuing professional development program. Scientific tours, an exhibition of radiation protection equipment and services, and a lively social program completed the Congress activities. A modified rapporteur system of presentation was used, and a relatively few papers were given orally by individual authors. This system, not in great use in the United States, received some criticism. However, it does provide an efficient system of presenting in a limited time a large number of papers, with only the pertinent facts being offered by the rapporteur, thus saving the audience listening to details.

The Scientific Program Committee under chairmanship of Dr. Bo Lindell had a most difficult task in developing the technical program. Perhaps I am somewhat biased; however, I believe the committee did an outstanding job. At its first meeting in Budapest, Hungary, in May of 1972, the committee made the decision that all projection slides presented at the Congress would receive severe screening and inspection in order to prevent the presentation of poor slides and incomprehensible data. Some authors resented such an inspection procedure; however, I am certain that the audience appreciated the fact that the committee instituted this. Such a procedure should be an integral part of all scientific meetings.

The IRPA Executive Council agreed to allow publication of the Congress Proceedings after receiving a most welcome invitation from the U. S. Atomic Energy Commission to print the papers in two volumes at no cost to IRPA. The task of editing and working with the USAEC, of course, fell to the IRPA Publications Director, Dr. Walter S. Snyder, and his staff at the Oak Ridge National Laboratory. The publication of such an extensive body of scientific papers requires a tremendous amount of effort on the part of Dr. Snyder and his staff, and the IRPA Executive Council sincerely appreciates this effort. These Proceedings will provide a valuable record of the Congress and serve as a significant reference in the future.

The papers in these Proceedings are arranged by the sessions as specified by the Scientific Program Committee and as presented at the Congress. Unfortunately, it was impossible to include the rapporteur papers in this publication. Nevertheless, the Program Committee and the International Meeting Commission gratefully acknowledge the effort that went into the preparation and presentation of their reports. Only a few papers for various reasons could not be obtained for publication. I am sorry for those authors who will not have their papers published, after going to the effort of presenting their information at the Congress. The Editor and I both acknowledge with gratitude and express our thanks for the cooperation of the authors of the papers presented here.

At the IRPA General Assembly, we observed the passing of the Presidential Bell from Dr. W. G. Marley to the capable hands of Dr. Carlo Polvani. We all express our most grateful thanks to Gregg Marley for a job well done; to Carlo Polvani, we wish great success in leading IRPA these next three years. It was indeed gratifying to welcome to the fold of IRPA our most recent affiliated society--the Radiation Hygiene Section of the Scientific Society of Hygienists of the USSR. We also observed that IRPA now has a reasonable bank account and will wisely use its funds to support international and regional meetings and appropriate international organizations concerned with radiation protection. The Assembly also accepted the invitation of the Czechoslovak Society of Nuclear Medicine and Radiation Hygiene to hold the Fourth International Congress in 1976. We will be pleased to assist Dr. Vladislav Klener, the new Vice President for Congress Affairs, in preparing for that Congress.

Another outstanding event of the Congress was the first presentation of the Sievert Award to Dr. Bo Lindell of Sweden. Rolf M. Sievert was a giant in the field of radiation physics and protection and a gentleman. Bo Lindell is of the same mold and a most worthy recipient of the award. His lecture on the assessment of radiation risk was an outstanding contribution to the Congress.

The Congress would not have been a success without the financial contributions and assistance of many national and international groups and agencies. These are acknowledged on page of this volume. Also our deep appreciation is given to the scientific and technical exhibitors who gave us an opportunity to observe their equipment and learn of their services to our fields of endeavor. Due thanks is also given to the exhibitors for contributing to the financial success of our Congress.

It is an impossible task to thank personally each individual who worked hard and long, behind the scenes and at the forefront, to make the Congress the success that it was. To each of you, it was a pleasure for me to share in the effort and to work with so many fine people from so many lands.

Claire C. Palmiter  
Congress President

## IRPA EXECUTIVE COUNCIL

W. G. Marley, President-----United Kingdom  
C. Polvani, Vice President-----Italy  
C. C. Palmiter, Vice President for  
Congress Affairs----- USA  
J. R. Horan, Executive Officer-----USA  
P. Courvoisier, Treasurer----- Switzerland  
W. S. Snyder, Publications Director-----USA  
L. Bozoky----- Hungary  
A. R. Gopal-Ayengar----- India  
H. P. Jammet----- France  
B. Lindell----- Sweden  
Y. Nishiwaki----- Japan (IAEA)  
Y. Feige----- Israel  
K. Z. Morgan, President Emeritus-----USA

## INTERNATIONAL MEETING COMMISSION

Claire C. Palmiter, Chairman  
Robert J. Catlin, Secretary General  
H. Davis Bruner, Deputy Secretary General  
Lawrence A. Brauch, Treasurer  
John C. Villforth, Secretary, Scientific Program Committee  
Lester A. Slaback, Jr., Executive Secretary  
James C. Malara, Congress Manager  
Raphael S. Daniels, Banquet  
Cecil R. Buchanan, Exhibition  
Arthur A. Levin, State Department Coordinator  
Joseph M. Brown, Housing  
Harold W. Gaut, Social Members' Program  
John G. Bailey, Public Information  
Glenn W. Zimmer, Congress Social Program  
Abraham Schwebel, Scientific Tours & Refresher Courses  
Robert E. Alexander, Transportation  
Samuel Sperling, Final Program & Brochure  
Francis A. Leone, Acting Secretary General (on departure of Catlin)

## SCIENTIFIC PROGRAM COMMITTEE

B. Lindell, Sweden, Chairman  
J. C. Villforth, USA, Secretary  
H. Brunner, Switzerland  
M. Gras, France  
M. Izawa, Japan  
J. Liniecki, Poland  
B. Lister, Scotland  
A. Marko, Canada  
Z. Nooteboom-Beekman,  
Netherlands  
D. Sowby, ICRP

## ACKNOWLEDGMENTS

The IRPA Executive Council and the International Meeting Commission gratefully acknowledge the assistance of the Health Physics Society for hosting the Third International Congress and the grants and contributions from the following national and international groups:

U. S. Atomic Energy Commission	Defense Apparel Nu-Con Products
U. S. Department of Commerce	Edison Electric Institute
U. S. Environmental Protection Agency	Gulf Foundation
U. S. Food and Drug Administration (HEW), Bureau of Radiological Health	Harshaw Chemical Company
Atomic Energy of Canada, Ltd.	Kerr-McGee Corporation
Commission on European Communities	R. S. Landauer Jr. & Company
International Atomic Energy	Nuclear Associates, Inc.
Abbott Laboratories, Radiopharmaceutical Products Division	Nuclear Engineering Company, Inc.
Bechtel Power Corporation	E. R. Squibb & Sons, Inc.
Boston Edison Company	Teledyne Isotopes
Consolidated Edison Company of New York, Inc.	Victoreen Instrument, Division of VLN
	Westinghouse Environmental Systems

## FOREWORD BY THE EDITOR

The Editor hopes these Proceedings will be a useful summary of the Third International Congress. Every effort was made to hold authors to their allotted number of pages, and the compromises finally reached were related to the total number of pages for each category of papers. In a few instances, authors did arrange for use of supplementary pages originally assigned to another author from the same institution. No retyping has been done to achieve a uniform style, and thus the presentation of the material is due entirely to the authors. Some papers were withdrawn prior to the Congress, and some authors were unable or unwilling to deliver papers in time for inclusion in the Proceedings. The Table of Contents consists of the complete program as presented for the Congress. The reader may wish to contact individual authors for missing papers or for further details on the work presented here. The Editor wishes to acknowledge the work of James Hickey and Sally Stockstill who helped in the editorial work for these Proceedings.

Walter S. Snyder  
Publications Director  
IRPA

Oak Ridge, Tennessee  
February 1974

# CONTENTS

Preface	ii
Foreword by the Editor	vi

## PART 1

### RADIATION PERSPECTIVE IN THE UNITED STATES OF AMERICA

The U. S. Congress and Radiation Protection . . . . .	1
Melvin Price	
Radiation Perspective in the United States of America . . . . .	8
Ralph Nader	
Perspectives on Radiation Risks . . . . .	14
Ralph E. Lapp	

### RADIATION AND MAN

Presentation of U.S.A. National Academy of Sciences Report on the Effects on Populations of Exposure to Low Levels of Ionizing Radiation (BEIR Report) . . . . .	28
General Review and Implications . . . . .	28
C. L. Comar	
Genetic Effects . . . . .	37
J. F. Crow	
Somatic Effects . . . . .	43
A. C. Upton	

### NON-IONIZING RADIATION

Biological Effects and Exposure Standards for Non-Ionizing Electromagnetic Energies . . . . .	51
S. M. Michaelson	
A Programme on Non-Ionizing Radiation Protection of the World Health Organization Regional Office for Europe . . . . .	64
M. J. Suess	
Pitfalls in the Assessment of Microwave Radiation as a Hazard . . . . .	76
R. L. Carpenter, E. S. Ferri and G. J. Hagan	
Control and Use of Lasers in N.Y.S. Industry . . . . .	79
F. J. Bradley and S. N. Roberto	



Combined Effect of Ionizing Radiation and a Superhigh-Frequency Field on the Body . . . . .	85
A. N. Liberman, I. E. Bronstein, V. V. Orabei, M. S. Sakavskaja, A. P. Tchesnokova, V. M. Shubik and R. I. Bikkulov	
Public Health and Control of Non-Ionizing Radiation in Japan . . . . .	91
H. Iida and T. Koshijima	
Progress in the Reduction of Microwave Exposure from Microwave Ovens Used in Commercial Food Vending Operations . . . . .	96
M. C. Wukasch and J. F. Thiel	
Ultraviolet Radiation Dosimetry Utilizing Thermoluminescence . . . . .	100
A. Dhar, L. A. DeWerd and T. G. Stoebe	
A Calibration Facility for Microwave Monitors--Design and Operating Experience . . . . .	106
I. S. Leith	
Computer-Assisted Laser Hazard Calculations and a Critical Analysis of the Prediction Equations . . . . .	112
W. A. Cornelius and I. S. Leith	

#### RADIATION EFFECTS ON ANIMALS

Materials on the Toxicology of Californium-252 . . . . .	117
G. A. Zalikin, Yu. I. Moskalev, A. I. Semenov, Ye. S. Zharava, V. N. Strel'tsova, I. K. Petrovich and V. I. Trifonov	
Effect of Fluorination of Drinking Water and a Calcium-Augmented Diet on the Radiotolerance of Animals during External Gamma Irradiation . . . . .	129
V. A. Knizhnikov and V. A. Grozovskaya	
Joint Effect of Strontium-90 and Chemicals Used in Agriculture on the Rat Organism . . . . .	136
I. Ye. Mukhin, N. M. Borovikova, and L. I. Nagovitsyna	
Effect of Incorporated Radionuclides on Immunity . . . . .	141
M. A. Nevstruyeva, V. A. Kolotvin, R. Ye. Livshits, and V. M. Shubik	
Comparison of the Biological Effect of Strontium-90, Cesium-137, Iodine-131 and External Irradiation . . . . .	147
N. A. Zapol'skaya, V. V. Borisova, L. Ya. Zhorna, L. N. Lavrent'ev, E. D. Pavlitskaya, A. V. Fedorova and N. G. Yakovleva	

Remote After Effects following External Beta Irradiation . . . . .	153
Yu. I. Moskalev, V. I. Shelesnava and A. I. Milovidava	
Change in Mean Lifetime of Rats in Dependence an Frequency of Development of Osteosarcomas Induced by Strontium-90 . . . . .	159
V. L. Shvedov	
Remote After Effects Associated with Damage by Transuranium Elements . . . . .	167
Yu. I. Moskalev, E. I. Rudnitskaya, L. A. Buldakov, A. P. Nifatav and L. G. Filippava	
Biological Effects Observed with Combined Exposure to Radiations . . . . .	172
V. S. Kalistratava, I. S. Katsapav and V. I. Trifonov	
Characteristics of the Combined Effect of Radioactive Substances (Iodine-131, Promethium- 147) and External Radiation on the Body . . . . .	178
L. N. Burykina, R. L. Orlyanskaya, V. L. Ponomareva, L. A. Vasil'yeva, K. A. Veselovskaya, Sh. Zhanadilav and N. N. Mel'nikova	
Quantification of Post-Irradiation Leukocyte Data Relative to Estimating Effective Exposure . . . . .	184
G. M. Angleton, C. D. Rabinette, P. B. Carter and L. F. Wailly	
Study of Puric and Pyrimidic Compounds in Urine of Rabbits and Rats after Irradiation . . . . .	190
Ch. Valle, G. Pepin, F. Duprey and L. Breuil	
Structural Aberrations in Bone Marrow Cells after Tritiated Water Administration in Rats . . . . .	196
I. Andreuta and N. T. Racoveanu	
Modification of Some Biological Indicators Induced by Single and Repeated Administration of Tritiated Water . . . . .	201
V. Stanculescu and A. Ciubotaru-Bordeianu	
Comparative Toxicity of Inhaled Beta-Emitting Radionuclides in Beagle Dogs . . . . .	208
R. O. McClellan, B. B. Boecker, F. F. Hahn, C. H. Hobbs, R. K. Jones and M. B. Snipes	
Benefits and Risks to Nuclear Power in the United States of the "As Low As Practicable" Philosophy . . . . .	214
M. I. Goldman	

## RADIATION QUANTITIES AND UNITS

The Dose Equivalent Equation . . . . .	225
H. O. Wyckoff	
The "Index Quantities" of the ICRU . . . . .	229
H. H. Rossi	
SI Units in Ionizing Radiation . . . . .	232
A. Allisy	
An Experimental and a Computational Program for Obtaining a Relationship between Quality Factor, Q, and Linear Energy Transfer, L (Not available for these Proceedings)	
J. Neufeld	

## RADIOECOLOGY

Distribution of Environmental Plutonium in the Trinity Site Ecosystem after 17 Years . . . . .	242
T. E. Hakonson and L. J. Johnson	
The Distribution of Plutonium in Liquid Waste Disposal Areas at Los Alamos . . . . .	248
T. E. Hakonson, L. J. Johnson, and W. D. Purtymun	
Radiation-Hygienic Evaluation of Geochemical Provinces with Increased Cesium-137 Migration . . . . .	254
A. N. Marey, R. M. Barkhudarov and N. Ya. Novikova	
Migration of Radioactive Nuclides under Different Hydrogeological Conditions . . . . .	260
A. S. Belitskiy, Ye. I. Orlova, Yu. A. Zhakov, R. A. Chelysheva and V. A. Smirennaya	
Analysis of Geographical Differences in the Rate of Purification of the Chain Lichens-Reindeer- Man from Cesium-137 . . . . .	266
M. N. Troitskaya, M. S. Ibatullin, B. Ya. Litver, A. A. Moiseyev, A. I. Nizhnikov, P. V. Ramzayev and L. A. Teplykh	
Levels of Environmental Radioactivity in Bikini Atoll (Not available for these Proceedings)	
T. F. McCrow	

High Radioactivity in Drinking Water and Ground Water in South Texas . . . . .	272
M. C. Wukasz and L. M. Cook	
$^{137}\text{Cs}$ and $^{134}\text{Cs}$ Distribution in Sediment, Water, and Biota of the Lower Hudson River and Their Dosimetric Implications for Man . . . . .	279
M. E. Wrenn, S. M. Jinks, N. Cohen and L. M. Hairr	
An Investigation into the Reasons for a Lack of a Trophic Level Effect for $^{137}\text{Cs}$ in <u>Fundulus Heteroclitus</u> and Its Food in the Hudson River Estuary . . . . .	285
J. Mauro and M. E. Wrenn	

REACTOR EXPERIENCES, WASTE MANAGEMENT  
AND ENVIRONMENTAL MONITORING

Measurements of the Dispersion of Gaseous Radioactive Effluents from an Operating Boiling Water Reactor . . . . .	291
W. M. Carey, L. Battist and W. E. Keene	
Atomic Electric Power Stations as Sources of Carbon-14 Discharge . . . . .	296
V. P. Rublevskiy, A. S. Zykova and A. D. Turkin	
The Safe Disposal of Liquid Radioactive Wastes from the United Kingdom Nuclear Power Programme: A Current View of the Environmental Situation . . . . .	301
N. T. Mitchell	
Climatological and Meteorological Considerations for Siting of Nuclear Installations in Hilly or Mountainous Environment . . . . .	308
E. Nagel	
A Model for the Evaluation of the Deep Ocean Disposal of Radioactive Waste . . . . .	313
G. A. M. Webb and F. Morley	
Impact of Ten Years Liquid Waste Management Practices on Environmental Safety at Bradwell Power Station, U.K. . . . .	319
M. M. Wasson and N. T. Mitchell	
Environmental Radiation from Advanced Gas-Cooled Reactors in Normal Operation . . . . .	326
F. H. Passant	
Research to Keep Nuclear Power Safe: The Experience of a Major Industry . . . . .	332
B. M. Wheatley	

Radiological Problems on Magnox Reactors with Integral Boilers, with Particular Reference to Personnel Entry into the Pressure Vessel . . . . .	338
H. C. Tresise	
Long Range Transport and Diffusion: An Approach to the Problem from the Health Protection Point of View . . . . .	344
P. Cagnetti and M. Pagliari	
Estimation of Gaseous Radioactivity Release Rates from an Operating Boiling Water Reactor . . . . .	350
W. E. Keene, William M. Carey and Lewis Battist	
An Investigation of Radioactive Contamination Imposed upon a Big Nuclear Power Installation in the Periodical Examination (Not available for these Proceedings)	
M. Suzuki, M. Fojtik, S. Watanabe, S. Hongo, Y. Inoue, S. Ohna, Y. Haruyama, S. Yoshino and H. Okabayashi	
Siting Evaluation of a Nuclear Power Plant in Southern Israel . . . . .	356
A. Donagi, Y. Feige, Y. Mamane and Y. Argaman	
A Review of the First Seven Years of Operational Health Physics at Dungeness 'A' (Magnox) Nuclear Power Station . . . . .	364
E. P. Goldfinch	
Trapping and Desorption of Iodine in Graphite Gas Reactors . . . . .	370
J. Barbier, G. Benezech, B. Cadet, J. Miribel and P. Sigli	
Adsorption of Fission Noble Gases on Charcoal . . . . .	376
A. S. Galdin, D. W. Underhill and H. A. Trindade	
New Low Level Environmental Radiation Monitoring around Nuclear Facilities Discriminating from Natural Radiation . . . . .	382
S. Moriuchi, K. Imai and I. Miyanaga	
Monitoring Technique for Radioiodine in Sodium Aerosol . . . . .	388
M. Naritomi, M. Murata and Y. Yoshida	

Long-Term Use of Various Solid-State Dosimeters for Environmental Monitoring of Nuclear Plants— Experience and Results . . . . .	394
B. Burgkhardt, E. Piesch and M. Winter	
Investigation of the Tritium Level in the Environment of the Karlsruhe Nuclear Research Center . . . . .	400
L. A. König and M. Winter	
Analysis of an Analytical Technique for Distributing Air Sampling Locations around Nuclear Facilities . . . . .	406
D. A. Waite	
Atmospheric Contamination Observed at the Chooz (Ardennes) Pressurized Water Plant . . . . .	411
L. Bertron, P. Beau and J. J. Martin	

#### LATE RADIATION EFFECTS

Malignancy Risk to Humans from Total Body $\gamma$ -Ray Irradiation . . . . .	417
C. W. Moys, R. D. Lloyd and J. H. Marshall	
Concept of the Acceptable Risk as the Basis for Standardizing Radiation Exposures (Not available for these Proceedings)	
A. V. Terman and A. A. Moiseyev	
The Risk of New Cancer in Women RADIATED for Breast Cancer . . . . .	429
L. A. Sagan and G. Linden	
Radiosensitivity and Dosimetry of the Tissues of Bone — Implications for Setting Maximum Permissible Levels of Beta Emitters . . . . .	439
F. W. Spiers	
A Theory of Radiation Risk Based on Microdosimetry . . . . .	448
R. D. Caldwell	
A Common Molecular Mechanism in Radiobiology— Its Implications in Radiological Protection . . . . .	457
K. H. Chadwick and H. P. Leenhouts	
Recent Trends towards the Dose Hazard Reference in Standards for Different Nuclear Activities . . . . .	469
L. Failla	

Experience in Using Clinical Data for Establishing the Maximum Admissible Levels of Occupational Irradiation . . . . .	475
A. K. Gus'kova, Ye. A. Denisova, E. N. L'vovskaya, G. I. Kirsanova and I. A. Gribova	
Early Haematological Detection of the Effects of Low Doses of Ionizing Radiation in Professionally or Medicinally Exposed Persons . . . . .	481
T. Predmerszky, O. Geszti, I. Elöd and I. Loványi	
Physiological Investigations of the Central Nervous and Cardiovascular Systems in Persons Subjected to Long-Term Occupational Irradiation over a Period of Years . . . . .	485
V. K. Sel'tser, V. V. Gamper, G. N. Katsnel'son, N. V. Loseva and A. I. Stepanov	
Trial Evaluation of the Long-Term Survey of Death Causes in the Population Living Near Radiation Rich Hot Spring . . . . .	491
K. Maeda, S. Abe, Y. Kurokawa and Y. Minagawa	
X-Ray Effects on the Embryo and Fetus: A Review of Experimental Findings . . . . .	495
R. Rugh and W. Leach	
Study of the Adrenal Function in Subjects with Long-Term Low Level Occupational Exposure to Ionizing Radiations . . . . .	501
H. I. Popescu and I. Lancranjan	
PERSONNEL DOSIMETRY INCLUDING TLD	
Occupational Exposure to X-Rays in Poland—Probabilistic Extrapolation to Life-Time Doses . . . . .	506
J. Jankowski, J. Liniecki and B. Krych	
Progress in Personnel Neutron Dosimetry . . . . .	515
D. E. Hankins	
Ionizing Radiation Records for Individuals . . . . .	528
C. S. Cook	
The Rapid Determination of the Transuranium Elements by Extraction Chromatography in Urines Containing DTPA . . . . .	532
A. Delle Site, G. Santori and C. Testa	

Excretion of Tritium in Workers Preparing Labelled Compounds . . . . .	538
M. Houskova, A. Cajkova, E. Kunz and L. Namestek	
Occupational Exposure to Ionizing Radiation in Iraq . . . . .	544
M. S. Abbas	
A New Technique for Improved Track Recognition in Nuclear Emulsion Film Dosimetry Using Soft X-Rays . . . . .	550
V. Balasubrahmanyam and L. Szegedi	
A Thermoluminescent Personal Dosimeter Compatible with Automatic Processing and the Central Recording of Dose Histories . . . . .	556
K. B. Shaw, T. O. Marshall and P. N. Casbolt	
A Universal Automatic TLD-Reader for Large-Scale Radiation Dosimetry . . . . .	563
H. W. Julius, C. W. Verhoef and F. Busscher	
Annealing TLD's Exposed to High Levels of Neutron and Gamma Radiation . . . . .	568
G. W. R. Endres, F. N. Eichner and L. L. Nichols	
A TLD Personnel Monitoring System with Automatic Processing . . . . .	572
L. Botter-Jensen, P. Christensen and B. Majborn	
Some Practical Applications of Sintered BeO as TL Dosimeter . . . . .	578
G. Benincasa, L. Ceravola and G. Scarpa	
Compounds of TLDs and High-Melting Organics for Fast Neutron Personnel Dosimetry . . . . .	584
K. Becker, T. D. Tham and F. F. Haywood	

#### THE SIEVERT LECTURE

Rolf M. Sievert: The Pioneer in the Field of Radiation Protection . . . . .	589
K. Z. Morgan	
Radiation and Man . . . . .	591
B. Lindell	



## DOSE CALCULATIONS

Dosimetric Aspects of a Forthcoming Report of ICRP on Internal Emitters . . . . .	603
W. S. Snyder	
Use of the Transport Operators Method for Analytical Computation of Tissue-Dosimetric Characteristics when Radioactive Isotopes Enter the Body by Inhalation (Not available for these Proceedings)	
V. P. Shamov and V. M. Malykhin	
Statistical Approach in Developing Mathematical Models for Evaluating Internal Irradiation (Not available for these Proceedings) . . . . .	610
V. I. Bad'in, A. P. Yermilov, U. Ya. Margulis and V. T. Khrushch	
A Tabulation of Specific Effective Energies for Dose Calculations . . . . .	619
M. R. Ford and W. S. Snyder	
Estimates of Dose Rate to Gonads of Infants and Children from a Photon Emitter in Various Organs of the Body . . . . .	625
W. S. Snyder	
Dose from Photon Emitters Distributed Uniformly in the Total Body as a Function of Age . . . . .	631
M. J. Hilyer, G. S. Hill and G. G. Warner	
Dosimetric Study of Residual Brain Contamination after the Injection of a Solution of Ytterbium-169- DTPA in the Cerebrospinal Fluid . . . . .	637
J. C. Zerbib, L. Imbert, M. Bonleu, H. Joffre, G. De Tovar, F. Beaufils, M. Collonge and R. Trial	
A Technique for Estimating Thyroid Doses from the Intake of Iodine-129 . . . . .	643
E. E. Oscarson	
Study of Spatial Distribution of Tissue Doses with the Aid of a Phantom-Mannequin . . . . .	648
L. N. Smirenniy, E. G. Litvinova A. V. Khortsev	

The Need for an Operational Radiation Dose Index System . . . . .	654
S. J. Harris	
Dose Calculations in External Exposure to Inert Gases . . . . .	659
R. Schaeffer	
Correlation between Fast Neutron Dose Equivalents and In Vivo Activation Products . . . . .	671
T. Schlesinger and Y. Klopfer	
Determination of Absorbed Energy for Various Types of Medical Irradiations . . . . .	677
L. Bozoky and I. Feher	
Beta Dose from Cutaneous Contamination . . . . .	683
K. Unnikrishnan, R. K. Hukkoo, S. Bhati and S. Somasundaram	
Calculation of Surface Dose and Internal Organ Dose for Human Exposure to Weapon Neutrons . . . . .	688
R. A. Facey	
Dose Due to Practical Neutron Energy Distributions Incident on Concrete Shielding Walls . . . . .	694
J. M. Wyckoff and A. B. Chilton	
Evaluation of Personnel Exposures in a High-Energy, High-Intensity Beta-Gamma Field . . . . .	700
B. L. O'Neal	
Dose, Dose Equivalent, Effective Dose and Cell Survival from Negative Plans . . . . .	707
H. A. Wright, R. N. Hamm and J. E. Turner	

METABOLISM OF URANIUM AND  
TRANSURANIUM ELEMENTS

Biomedical Follow-Up of the Manhattan Project Plutonium Workers . . . . .	713
L. H. Hempelmann, W. H. Langham, G. L. Voelz and C. R. Richmond	
Solubility Changes of <sup>238</sup> Pu Oxide in Water Suspension and Effect on Biological Behavior after Inhalation by Beagle Dogs . . . . .	719
J. F. Park, D. L. Catt, K. D. Craig, R. J. Olson and V. H. Smith	

The Biological Disposition of $\text{Es}(\text{NO}_2)_3$ in Rats after Intravenous, Intramuscular and Subcutaneous Administration . . . . .	725
V. H. Smith	

Retention and Elimination of Berkelium-249- Californium-249 following Acute Accidental Inhalation . . . . .	731
J. Rundo and J. Sedlet	

Toxicology of Transportable Uranium: A Critical Contribution to Health Protection of Workers . . . . .	736
F. Breuer and E. Righi	

The Effect of Natural Physiological Stresses and Artificial Hormonal Stresses on the Retention of Americium and Plutonium by Rat Bone . . . . .	743
J. G. Hollins, M. C. Storr and A. Durakovic	

#### RADIATION ACCIDENTS

Industrial Radiation Incidents in the United Kingdom . . . . .	749
C. D. Burgess and E. G. Weatherley	

The Investigation and Reconstruction of a Severe Radiation Injury to an Industrial Radiographer in Scotland . . . . .	760
N. T. Harrison, P. C. Escott, G. W. Dolphin, R. J. Purrott, J. L. Alexander and K. E. Halnan	

Large Radiation Exposure . . . . .	769
V. Bhushan	

Clinical Course and Dosimetry of Acute Hand Injuries to Industrial Radiographers from Multicurie Sealed Gamma Sources . . . . .	773
E. L. Saenger, J. G. Kereiakes, N. Wald and G. E. Thoma	

Evaluation and Dosimetry of Accidental Exposures to an X-Ray Analysis Beam . . . . .	783
I. S. Leith and W. A. Cornelius	

---

Facts, Fables and Follies in Medical Radiology . . . . .	791
R. H. Chamberlain	

## PART 2

## OPERATIONAL HEALTH PHYSICS - 1

Operational Health Physics during Dismantling of the Elk River Reactor . . . . .	799
D. McConnon	
Special Problems in Radiation Safety and Sanitary-Dosimetric Monitoring when Working with Transplutonium Elements . . . . .	810
M. S. Yegorova, V. V. Kopayev, L. N. Korzhov and V. A. Sarychev	
Ensuring Radiation Safety when Using Large Isotopic Sources of Ionizing Radiation for Bringing about Radiation Processes . . . . .	814
E. D. Chistov, A. V. Larichev, A. V. Terman and O. M. Zaryaev	
Estimating the Effect of the Absorbed Phase of Contamination of Protective Coatings and Means for Individual Shielding for Radiation Conditions in Workrooms . . . . .	824
A. L. Kononovitch	
Ensuring Radiation Safety when Using Instruments with Radioisotopic Radiation Sources . . . . .	830
N. V. Vershinin, D. A. Bodrova, V. V. Volkovitskaya, P. P. Lyarskiy and P. I. Moiseyev	
Health Physics and Operational Experiences in a Treatment and Packaging Facility for Solid and Semi-Solid Wastes at E.I.R. . . . .	
J. Schwamberger and H. Brunner	
Some Preliminary Investigations on the Contribution of Muons to the Stray Radiation Level around the CERN 28 GeV Proton Synchrotron . . . . .	841
M. Hofert and J. Baarli	
Radiological Safety Experience in Handling and Fabrication of Plutonium Fuel Elements . . . . .	846
S. Janardhanan, T. N. Krishnamurthi and S. D. Soman	
Cellulose Nitrate Plastic Film - Its Preparation and Applications in Health Physics . . . . .	852
A. M. Bhagwat and S. D. Soman	
Retention of Iodine on Duct Surfaces . . . . .	857
W. E. Grummitt	

Nuclear Safety Organization in a University Research Facility . . . . .	864
P. Kissel and C. Renard	

EXPOSURES FROM RADIATION SOURCES  
OF NATURAL ORIGIN

Investigation and Standardization of the Radioactivity of Construction Materials . . . . .	870
E. M. Krisyuk, E. P. Lisachenko, S. I. Tarasov, V. P. Shamov and N. I. Shalak	
Eye Exposure from Thoriated Optical Glasses . . . . .	882
R. C. McMillan and S. A. Horne	
Dosimetry of Galactic and Salar Radiation On-Board the Supersonic Transport Concorde . . . . .	889
H. Francois, R. P. Defahaye, P. Simon, G. Portal, H. Kaiser and P. Durney	
The Approach to Radon Problems in Non-Uranium Mines in Sweden . . . . .	900
J. O. Snihs	
New Data Concerning the Safety of Miners in Uranium Mines . . . . .	912
J. Pradel, P. Zettwoog, G. Madelaine and Y. Francois	
Long-Term Measurements of Radon Daughter Activity in Mines . . . . .	920
B. Haider and W. Jacobi	
Evaluation of the Radiation Factor at Enterprises in the Rare and Ferrous Metals Industry . . . . .	926
P. P. Lyarskiy, L. T. Yelovskaya, Yu. T. Kapitanov, Yu. P. Likhachev, S. Ya. Satarina and S. A. Khukhlayeva	
A Radon Daughter Monitor for Use in Mines . . . . .	932
A. C. James and J. C. Strong	
Radon Emanation Studies in Jaduguda Uranium Mine . . . . .	939
A. H. Khan and M. Raghavayya	
Distribution of Airborne Activity in a Uranium Mill Using Cobwebs . . . . .	945
G. Jha and K. P. Eappen	

An Instant Working Level Meter with Automatic Individual Radon Daughter Readout for Uranium Mines . . . . .	950
P. G. Groer, D. J. Keefe, W. P. McDowell and R. F. Selman	

IN VIVO MEASUREMENTS

Monitoring of Low-Energy X-Ray Radionuclide Content in Human Body . . . . .	957
E. I. Dolguirev, G. N. Kajdanovsky N. V. Porozov and V. M. Shamov	
Optimization of Conditions for Gamma-Spectrometric Measurements in Radiation Shielding . . . . .	963
O. V. Lebedev, V. M. Malykhin and V. Ye. Pestrikov	
The Methods of Absolute Calibration of Equipment for Measurements of Pb-210, Pu-239 and Am-241 in Human Body . . . . .	969
E. Dolguirev, G. N. Kaidanovsky, N. V. Porozov and V. P. Shamov	
Sensitive Radiometric Method for in vivo Study of Incorporated Nuclides of the Thorium Series . . . . .	976
A. A. Petushkov, M. R. Zel'tser and P. P. Lyarskiy	
Assessment of Plutonium in Human Lungs with Thin NaI(Tl) Detector Systems . . . . .	979
R. C. Sharma, S. Somasundaram, T. Surendram, D. K. Kapur and S. P. Garg	
In Vivo Measurements following Exposure to $^{133}\text{Xe}$ and Associated Dose Assessment Procedures . . . . .	985
F. Fry and G. Tyler	
In-Vivo Monitoring of Nuclear Fuel Workers . . . . .	991
C. Pomroy, J. Gordon and E. G. Letourneau	
Application of Phoswich Detectors for Lung Counting Plutonium-238 . . . . .	999
F. K. Tomlinson, R. Brown, H. Anderson and B. Robinson	
Chestwall Tissue Measurements for Lung Counting Applications . . . . .	1005
F. K. Tomlinson, R. Brown, H. Anderson and B. Robinson	

In Vivo Measurement of Uranium Contamination in the Lung . . . . .	1011
E. Riklis and N. Degani	

#### EXPOSURE FROM NUCLEAR POWER

"As Low As Practicable" in Theory and Practice . . . . .	1017
G. H. Whipple	
Radiation Danger to the Population During Operation of Atomic Power Stations in the USSR . . . . .	1022
N. G. Gusev, V. A. Belyayev, Yu. A. Zhakov, A. S. Zykova, A. N. Marey, G. A. Kuznetsova and G. A. Gunyakov	
Average Effluent Releases from U. S. Nuclear Power Reactors, Compared with Those from Fossil- Fueled Plants, in Terms of Currently Applicable Environmental Standards . . . . .	1035
A. P. Hull	
Environmental Tritium and the Dose to Man . . . . .	1047
B. G. Bennett	
Individual Risk, Collective Damage, and Nuclear Power Plants . . . . .	1054
M. Delpla	
An Evaluation of Nuclear Gas Stimulation in Terms of Potential Radiation Exposure to the Public . . . . .	1060
P. S. Rohwer, C. J. Barton, R. E. Moore and S. V. Kaye	
Genetic Dose Limit for General Population, Dose Limit and Derived Concentration Guides for Members of the Public Compulsive Action Guides for Emergency Situations . . . . .	1066
R. M. de Nulman and C. A. de la Rosa	
VADOSCA: A Simple Code for the Evaluation of Population Exposure due to Radioactive Discharges . . . . .	1072
L. Bramati, T. Marzullo, I. Rosa and G. Zara	

#### MEDICAL EXPOSURE

Nationwide Evaluation of X-Ray Trends (NEXT) . . . . .	1078
J. O. Lubenau, C. M. Hardin, J. W. Shaver, I. P. Leggett, Jr., and L. Seabron	

Risk Criteria Associated with Use of  
Ionizing Radiations in Medicine (Not  
available for these Proceedings)

V. Ya. Golikov and I. P.  
Korenkov

The Genetically Significant Dose Due to Medical X-Ray Examinations in The Netherlands . . . . .	1085
J. A. Koen and J. Weber	
Evaluation of Diagnostic X-Ray Contribution to the Annual Genetically Significant Dose Equivalent of Taiwan Urban Population . . . . .	1091
P. S. Weng and C. Y. Huang	
The Role of the Health Physicist in Reducing Medical Radiation . . . . .	1096
R. E. Gross, M. E. Marks, M. F. Audet and J. S. Benson	
Dental Technique Normalization: Development and Pilot Test . . . . .	1103
K. L. Travis, J. R. Stanton, J. E. Hickey and B. M. Burnett	
Nashville Dental X-Ray Study . . . . .	1109
O. N. Johnson, C. L. Crabtree and S. J. Gibbs	
Absorbed Dose to Selected Internal Organs from Typical Diagnostic Exposures . . . . .	1115
J. W. Poston and G. G. Warner	
Handy Device for Protection of the Testicles in X-Ray Examinations . . . . .	1121
E. Lindholmer and O. Berg	
Dose Reduction by Electronic Radiography . . . . .	1127
D. Sashin, E. R. Heinz and E. J. Sternglass	
Medical Radiation Protection in the Eastern Mediterranean Region . . . . .	1133
G. Gomez-Crespo and D. R. E. Ernberg	
Sources of Unnecessary Irradiation During Fluoroscopy . . . . .	1139
N. T. Racoveanu, G. Modoran and S. Suma	



Medical Irradiation of the Population in Romania during 1970 . . . . .	1144
N. T. Rocoveanu, C. Diaconescu, G. Modoran, C. Vasilescu-Fulea, P. Plavosin and C. G. Milu	
Use of Medical X-Ray Diagnostic Units in Iraq . . . . .	1150
M. S. Abbas	
Radiobiological Significance of $^{125}\text{I}$ Microdosimetry . . . . .	1156
A. R. Reddy and K. K. Nagpal	
Dosimetry of Internal Emitters in Nuclear Medicine and Radiation Protection: At What Level . . . . .	1163
A. Nagarajnam and A. R. Reddy	
Contamination of Air and Surroundings by Patients Treated with Large Quantities of Iodine $^{131}\text{I}$ for Thyroid Carcinoma . . . . .	1169
C. H. Marshall, R. Chandra and M. Blum	
A Comprehensive Approach for the Evaluation of Comparative Dosimetry of Internally Administered Radiopharmaceuticals . . . . .	1175
P. Hosain and F. Hosain	
Reduction of Radiation Exposure to Nuclear Medicine Personnel by the Use of New "Instant" Techniques for Preparation of Technetium Radiopharmaceuticals . . . . .	1180
P. J. Robbins, B. M. Branson and R. J. Grant	
Reducing Personnel Exposure in Nuclear Medicine . . . . .	1185
B. M. Branson, R. G. Hoops and R. J. Grant	
Radioprotection in a Nuclear Medicine Unit . . . . .	1191
A. Favino and G. F. Besseghini	
Clinical-Dosimetric Relationships in Local Irradiation from Sources of Different Energies (Not available for these Proceedings)	
A. V. Barabanova, L. L. Sokolina and L. V. Novikova	
Radiation Protection in a Nuclear Pharmacy . . . . .	1197
R. D. Ice and K. R. Hetzel	

Radioactive Aerosols as a Factor in Internal Irradiation when Using Radioactive Substances . . . . .	1203
V. I. Bad'in, Z. G. Batova and G. M. Parkhomenko	
Validation of the Principal Radiobiological Characteristics of Thorium-232 and Its Decay Products and Hygienic Norms (MAAC and MAD) in the Atmosphere of Workrooms . . . . .	1208
P. P. Lyarskiy, N. A. Pavlovskaya, Yu. T. Kapitanov, L. T. Yelovskaya and L. G. Makeyeva	
Inhalation Study of a Submicron Size Lead-212 Aerosol . . . . .	1214
A. Bianco, F. R. Gibb and P. E. Morrow	
The Effect of Breath-Holding on the Deposition of Half-Micron Aerosol Particles in the Human Lung . . . . .	1220
M. C. Subba Ramu	
HASL Cyclone as an Instrument for Measuring Aerosol Parameters for New Lung Model . . . . .	1226
P. Kotrappa, S. K. Dua, D. P. Bhanti and P. P. Joshi	
Dose Commitment to the Lung from Inhalation of Radiocobalt in Power Reactor Operations . . . . .	1232
P. Kotrappa, S. D. Soman and B. Raghunath	
A Method for Determining the Dissolution Characteristics of Accidentally Released Radioactive Aerosols . . . . .	1237
G. M. Kanapilly, O. G. Raabe and H. A. Boyd	
Biological Modeling for Predicting Retention Patterns of Inhaled Contaminants . . . . .	1243
R. G. Cuddihy, W. C. Griffith and B. B. Boecker	
Deposition and Elimination of Iron Oxide Aerosol from the Lung of Rats: Comparison with ICRP Predictions for Man . . . . .	1249
P. J. Hewitt	
Inhalation Hazards: They Could be Worse . . . . .	1255
R. G. Thomas	

Protection Problems Involved with Non-Ionizing Radiation . . . . .	1261
H. Jammet	

PUBLIC INFORMATION, LEGAL ASPECTS,  
EDUCATION AND TRAINING

Radiation Protection Training Courses at E.I.R. Würenlingen, Switzerland . . . . .	1287
R. Spiess and H. Brunner	
Project Training for Graduate Students in Health Physics . . . . .	1298
J. R. A. Lakey and A. V. Yarke	
The Law and Low Level Radiation . . . . .	1307
C. F. Eason and N. Y. St. Denis	
Radiation Protection Information and Education for the Public: Mistakes and Lessons, Strategies and Tactics . . . . .	1318
H. Brunner	
Public Confidence in Nuclear Power . . . . .	1330
J. A. Bonnell and G. C. Dale	
Informing the Public about Nuclear Energy . . . . .	1337
H. Kiefer, W. Koelzer and G. Stäblein	

METABOLISM OF RADIONUCLIDES

A Study of Inhaled Sodium-22 . . . . .	1347
D. Bush	
Regularities in Strontium-90 Accumulation in Young Children (Not available for these Proceedings)	
B. K. Barisov	
Metabolism of Lead-210 and Polonium-210 in the Body . . . . .	1353
A. I. Nizhnikov, A. L. Zel'din, M. S. Ibatullin, B. Ya. Litver, P. V. Ramzayev and M. N. Traitskaya	
Use of a Specialized Analog Computer for the Model of the Metabolism of Some Radionuclides . . . . .	1358
I. A. Likhtarev, G. V. Arkhangejskaya, A. I. Dobroskok, I. A. Zvonova, N. F. Karelina, G. P. Krasnoshyekova, V. S. Repin and A. P. Ushakova	

Interpretation of Experimental Data on Polonium-210 Metabolism for Computing Admissible Levels . . . . .	1364
Yu. D. Parfenov	
Regularities in Metabolism of Radioactive Isotopes Upon Incidence on the Skin . . . . .	1371
L. A. Ilyin	
Determination of the Content and Kinetics of the Behaviour of Some Radionuclides in the Body of Man by the In Vivo Technique . . . . .	1377
A. D. Turkin, V. P. Stolyarov and Yu. Ja. Sokolov	
Retention and Distribution of Inorganic Mercury ( $^{197}\text{Hg}$ , $^{203}\text{Hg}$ ) in the Human Body after Single Inhalation . . . . .	1384
Y. Izumi, M. Fujita, A. Yabe and T. Hattori	
A Case Study of Human Contamination Due to Inhaled Thulium-170 Oxide . . . . .	1390
A. Yabe, H. Yamagishi, H. Fukuda, Y. Takahashi, S. Mizushita and T. Hattori	
Gastro-Intestinal Absorption of Cerium in Suckling Mice . . . . .	1396
Y. Feige, A. Naharin, E. Lubin and T. Sadeh	
A Human Metabolic Model for $^{14}\text{C}$ -Labelled Metabolites Useful in Dose Estimation . . . . .	1400
S. R. Bernard	
Metabolic Balances of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in Unexposed Men . . . . .	1406
R. B. Holtzman, H. Spencer, F. H. Ilcewicz and L. Kramer	
Long-Term Metabolism of $^{90}\text{Sr}$ in Rhesus Monkeys ( <i>Macaca mulatta</i> ) . . . . .	1412
P. W. Durbin, N. Jeung, M. H. Williams and M. W. Parratt	
A Study of the Translocation of Radiostrontium from Wounds and Therapy by Local Insolubilization . . . . .	1418
R. Ducousso, A. Causse and C. Pasquier	

OPERATIONAL HEALTH PHYSICS - II

Instruments and Methods for Day-to-Day Monitoring of Tritium and Carbon-14 in Production . . . . .	1422
M. I. Balonov, E. I. Dolguirev, A. S. Karasik and O. N. Prokofiev	
Sampling for Tritiated Water Vopour . . . . .	1428
R. V. Osborne	
A Ruggedized Ultrasensitive Field Air Sampler for Differentially Determining Tritium Oxide and Gas in Ambient Air Atmosphere . . . . .	1434
R. Brown, H. E. Meyer, B. Robinson and W. E. Sheehan	
Comparison of Albedo Dosimeters and Nuclear Track Detectors for Neutron Monitoring . . . . .	1440
E. Piesch and B. Burgkhardt	
Some Possibilities of Use of Silicon Detectors of Ionizing Radiations in Investigations of Radiation Shielding . . . . .	1446
A. A. Petushkov and V. A. Manchuk	
High Energy Photon Detection by Fission Track Registration . . . . .	1452
H. Iida and T. Koshijima	
An Inexpensive Lightweight Environmental Survey Instrument . . . . .	1458
I. M. G. Thompson and H. C. Orchard	
A New Technique in Environmental Neutron Spectroscopy . . . . .	1464
M. Awschalom and L. Coulson	
Conductimetric Properties of Some Solid Tract Detectors and Hypothesis on the Track Formation Mechanism in These Irradiated and Etched Detectors . . . . .	1470
Y. Martin-Borret, M. Bourdeaux, F. Roux and C. Briand	

---

Author Index

Index 1

## RADIATION PERSPECTIVE IN THE UNITED STATES OF AMERICA

### THE U.S. CONGRESS AND RADIATION PROTECTION

Congressman Melvin Price  
Chairman, Joint Committee on Atomic Energy

First of all, I would like to welcome all of you to the United States. I hope that your Third International Conference will be enjoyable and productive as well. We are pleased to be the host country for this important event.

I am particularly pleased to have the opportunity to address you in these opening remarks. I am acquainted in general terms with the structure and objectives of your organization. I believe that it is doing important work and that it will continue to be of importance.

It is clear to all that throughout the world man is utilizing radiation sources to a greater extent each year for a variety of purposes, all of which might be described as being under the umbrella of improving the quality of life. The prime examples under this heading are the utilization of radioisotopes and x-ray machines and other accelerators for diagnostic and therapeutic purposes in clinical practice and in research. More recently there has been public attention drawn to the matter of radiation protection associated with the utilization of atomic energy in the generation of electrical power.

It is perhaps trite to observe, at least to this group, that radiation knows no international borders. It recognizes no differences in man because of his national origin or ethnic group. For this very reason I congratulate you on your foresight in forming this International Association in which you can effectively interchange technical information and pursue your individual objectives with a commonality of purpose.

#### The Government Structure for Radiation Protection in the United States

In this country there are at least seven separate Federal agencies having jurisdiction in the area of providing protection against significant amounts of radiation exposure. In addition to the Atomic Energy Commission, these agencies include the Department of Defense; the Department of Transportation; the Environmental Protection Agency; the Department of Health, Education, and Welfare; and the Department of Labor. Moreover, most of our States have established their own radiation control programs in this area. In this regard, there has been some concern on the part of the States that the responsibility in the protection of the public and the worker from exposure to radiation has been significantly fragmented at the Federal level. In fact, the States have urged the Congress to consider this problem of fragmentation and to take the necessary legislative action to consolidate the various Federal responsibilities relating to radiation and protection of the public and the worker within a single agency.

The concern of the States over the apparent fragmentation of Federal jurisdiction in this area is understandable. The Joint Committee on Atomic Energy

would, of course, be one of several congressional committees which would have a responsibility in this area. Therefore, the Joint Committee is currently reviewing this matter to determine the best method of obtaining the additional information which would be necessary for any congressional consideration of the problem and of securing concerted action by the various congressional committees which would be involved. I mention this because it is possible that several of you may have the same situation in your own countries.

#### Role of the U. S. Congress

I would like to tell you a little bit about the role of the U. S. Congress with respect to the establishment and enforcement of radiation protection standards in this country. It is appropriate that the Congress have a role in this matter. The establishment of radiation protection standards involves certain considerations which are sociological in nature, and, therefore, political. In our form of government the public can express its views through its elected officials. Surely, the understanding of the biological effects of exposure of man to ionizing radiation requires background and technical expertise which cannot be obtained overnight. In fact, I feel certain that many members of the lay public would readily admit that as individuals they might never understand such a complex subject. Nevertheless, the views of the lay public concerning these matters are of interest to the Congress. It goes without saying that we also consider as valuable input data the views of trained scientists from outside those agencies charged with the setting of radiation protection standards.

There are a variety of disciplines which are essential to the setting of radiation protection standards. These include biology, genetics, physics, bioradiology, chemistry, and many others. The problem is, of course, complicated by the realization that ionizing radiation, even at very low levels, can change the chemical and physical nature of matter and, thereby, bring about what can be referred to as "damage." We know, of course, that man has always lived with radioactivity and, therefore, has been subjected to continuous exposure at some level. The question, therefore, is whether the additional increment allowable under the protection standard causes any significant change, or "damage" if you like, relative to the benefit anticipated as a consequence of the exposure.

The Congress in the United States does its work through its committees. This is necessary because the matters which come before the Congress are many and varied. By use of the committee system, it is possible for some Members of Congress to develop background in specialized areas and give greater attention to those areas than could be given by the full Congress. The committee then makes recommendations which are acted upon by the full Congress. A committee of Congress which clearly has exercised its jurisdiction with respect to radiation standards, and I believe properly so, is the Joint Committee on Atomic Energy. Other Committees have exercised roles with respect to establishing the organizational structures which are involved in standards setting.

#### Joint Committee on Atomic Energy

The special interest of the Joint Committee on Atomic Energy was drawn to the area of radiation protection by reason of radioactive fallout which occurred from nuclear weapons tests in the early 1950s. The Limited Nuclear Test Ban Treaty of 1963 brought about a considerable reduction in nuclear weapons testing. As you know, some world powers have not signed the treaty and continue to conduct atmospheric tests.

The Committee started detailed hearings on radiation protection criteria and standards in the fifties. Our hearing records for the past three decades provide a basic source of information in this field.

The subject of employee radiation hazards, including recordkeeping, for example, was given extensive examination by the Committee in 1959 and 1966. The special problems of exposure of uranium miners and problems associated with the accumulation of uranium mill tailings were treated in hearings held during 1959, 1967, 1969, and 1971.

On the subject of possible radiation exposure to the public as a consequence of routine operation of nuclear power generating stations, the Committee held extensive hearings in 1969 and 1970 under the title of "Environmental Effects of Producing Electric Power." Testimony was received on the environmental impact resulting from the operation of all kinds of electric generating stations.

The record of these hearings consists of over 3,000 pages of testimony and pertinent appended material. We have been unable to fill all of the requests for copies of these hearing records. It was brought out in these hearings that the record of operation of nuclear power plants clearly indicates that they can be operated so that routine releases of effluents can be held to quite low levels, in fact, substantially below levels allowable under the Federal exposure guides. Subsequently, the Atomic Energy Commission proposed design criteria for light water nuclear power plants which would have as their objective keeping effluents from these reactors to levels which are "as low as practicable."

The Commission is currently, through an appointed board, conducting a rule-making hearing on this subject. The Commission Staff has prepared and issued an environmental impact statement with respect to the proposed rule. The objective of this action and the concomitant engineering accomplishment of the nuclear industry have been such that there has been no substantive comment or objection to the proposed rule by environmentalists.

While on the subject of nuclear power plants, I would like to point out that the Joint Committee has scheduled public hearings beginning on September 25 on the subject of nuclear reactor safety. We anticipate that during these and subsequent hearings we will be getting into substantive matters concerning the risk of nuclear accidents, and we anticipate a rather complete examination of the Commission's Reactor Safety Research Program. This phase of our hearings will provide an opportunity to the Executive Branch of our Government to put on the public record a concise presentation of all matters related to reactor safety. During a later phase (probably within a month or two) other interested parties will be given an opportunity to present their views. This will include representatives of industry, representatives of environmental groups, members of the scientific community, and the public at large.

A point which I would like to make at this juncture is that the Joint Committee, in the case of each of the aforementioned Committee hearings, has published and made freely available to the public a hearing record which presents the views of witnesses, both pro and con, on these various matters relating to radiation protection. I am sure that many of you are familiar with these publications and that a number of you here today have actively participated in the many hearings which have been held.

#### History of the Federal Radiation Council

The Federal Radiation Council was established by Executive Order of the President in August of 1959. In September of that same year the Council was



made statutory by an amendment to the Atomic Energy Act. The Joint Committee felt that it was appropriate that this council, as an advisory body to the President, have a charter provided for by statute in order that the importance of its role be firmly established at the outset of its existence. The Council was made up of heads of the Federal agencies of the Government concerned with radiation protection standards. The detailed work of the Council has been performed by a working group drawn from the staffs of the Federal agencies having membership on the Council.

The Council was formed in recognition of the fact that previously there had been no governmental body responsible for the establishment of radiation protection guides. The guides established by a nongovernmental body, such as the National Council on Radiation Protection, were being utilized by the Government in the conduct of its business and other activities where Federal regulation was imposed upon the activities of others relating to radiation.

In the fall of 1970 an Environmental Protection Agency was established within the Executive Branch of the government and charged with the responsibility of advising the President with respect to radiation matters directly or indirectly affecting health, including guidance for all Federal agencies in the formulation of radiation standards and in the establishment and execution of programs of cooperation with the States. We in Congress have heard little from the Environmental Protection Agency concerning the manner in which it is carrying out the functions of the Federal Radiation Council which was absorbed under the 1970 reorganization. It may now well be appropriate for the Committee which I chair to examine the manner in which the prescribed functions of the Council are now being carried out by the new Agency.

#### National Council on Radiation Protection and Measurements

I feel that it is safe to assume that you are all well acquainted with the work of the National Council on Radiation Protection and Measurements and its forerunner, the National Committee on Radiation Protection and Measurements. The earlier committee began its work in 1929 and has collected, analyzed, developed, and disseminated information and recommendations on radiation protection and measurements throughout the years.

In 1964 the Congress granted the NCRP a Federal charter. My colleague, Congressman Chet Holifield, was probably the one man in the Congress most responsible for bringing about this legislation which provided for the continuing independence of the NCRP while, at the same time, gave an identity to the Council which it had not previously had.

It is certainly an understatement to observe that the work of the NCRP has been invaluable to the establishment of radiation protection guides in this country. We in the Congress are well acquainted with Dr. Lauriston Taylor, President of the NCRP, and many of his colleagues who serve on the Council. We on the Joint Committee have had the benefit of having his testimony and counsel on radiation matters for many years. I feel sure that those in the position of responsibility for establishing Federal radiation protection guides in this country appreciate the information and recommendations of the Council.

As most of you know, in the Fall of 1969 there was considerable questioning of the adequacy of the Federal Radiation Protection Guides. Some members of the scientific community believed that since the conduct of activities involving radiation was quite feasible at radiation exposure levels considerably below the guides then in being, that the guides should automatically be lowered by a factor of 10 or even 100 -- all of this, notwithstanding the fact that no biological data had been produced indicating any unfavorable health effects

resulting from the low levels of radiation exposure allowable under the guides. The NCRP, it turns out, was just completing a 10-year study directed toward reassessing the adequacy of the then current radiation protection guides. The conclusions of the Council were to the effect that on the basis of past and presently available scientific data there existed no reason to modify the guides in any substantive fashion.

It was interesting to me personally to note that in testimony received by the Joint Committee during the Spring of this year, Atomic Energy Commission witnesses pointed out that a scientific experiment to assess any possible biological consequences of exposure of humans to 1700 milliroentgens per year (10 times the amount provided for in the present population protection guide) would require the utilization of 8 billion mice. In other words, an almost impracticable number of experimental animals would have to be observed over several generations in order to bring out any possible somatic or genetic implications.

Even to a layman, the variation in background radiation exposure which exists throughout the world (factors of 10 or more in some places) would suggest that any unfavorable effects resulting from such low levels of radiation would have become evident in man himself. Thus, it would not appear necessary or practical to go to the extremes of examining billions of mice at very low exposure levels and then extrapolating whatever we learn to man. In fact, if there is an effect, we should be able to observe it in the human data which are available to us on longevity, the occurrence of cancer, and other possible adverse effects of radiation.

#### Proposed Reorganization with Respect to Energy

In June of this year the President sent an energy message to the Congress which had as a principal element a reorganization of the executive agencies of our Government in a manner intended to bring about more centralized control of research and development in the energy field as well as greater control and direction in the utilization of our natural resources. Later, a reorganization plan was submitted in the form of a legislative proposal which is now before the government operation committees of both houses of Congress. My esteemed colleague, Chet Holifield, a member of the Joint Committee on Atomic Energy since its origin in 1946, is the Chairman of the House Government Operations Committee and has already begun the conduct of public hearings on the proposal. In Chairman Holifield's words:

"We . . . will need time to study this proposal in detail. We will have to determine whether it is well-considered and deserving of acceptance. Undoubtedly there will be some modification. The Congress will want to be assured that this is a workable organization, one which will perform with realism and competence."

Briefly, the proposal would create a new Department of Energy and Natural Resources (DENR) based upon the present Department of the Interior. That portion of the Atomic Energy Commission concerned with the raw materials uranium and thorium would be moved to this new Department.

A new agency, the Energy Research and Development Administration (ERDA), would be established as the key government agency for research and development in all forms of energy. It would be founded upon the broad scientific talent and experience of the Atomic Energy Commission and its national laboratories. It would fund and carry out research and development in all forms of energy,

assigning the priorities in a manner which would hopefully achieve the proper balance to solve both our near-term and long-range energy needs.

The licensing of nuclear facilities and related activities of the present Commission would be moved to an independent Nuclear Energy Commission (NEC). This would eliminate the old bug-a-boo of a single agency (as in the present AEC) being responsible for both the development and regulation of nuclear reactors. There are pluses and minuses which can be enumerated for having both functions carried out within a single agency and likewise pluses and minuses with respect to separation of the functions. The Joint Committee has long been aware of the desirability of a separation at the proper time.

In summary, the depth and magnitude of the U. S. Congress's interest and concern in your special area of interest is obvious. I have tried to point out our efforts to obtain every available fact on radiation for our guidance in legislative actions. I think this is obvious from the extensive hearing record the Joint Committee has developed. One of the prime sources of information we have utilized and we must continue to have access to is the specialized talents of your organization. My plea is that you do everything possible to facilitate access of each of your legislative bodies to this information. Only in this way will we get the best laws on radiation protection.

I should not conclude without a few remarks concerning the confrontations and controversy which exist in many fields of activity at the present time. I think we must all bear in mind that this sort of thing should not be unexpected in these days of highly improved communication equipment and communication services. Members of the public, individual scientists, and others have every right to be heard on matters which concern them. If their views have merit, they should be seriously considered, but this does not itself assure them of any right to delay or obstruct development programs or construction projects without good cause.

A special word about critics is in order. Critics are important. Constructive criticism has long had a place in our society, and it should by all means continue to be an integral part of the decision-making process for it is the well-spring of improvement. Informed and responsible critics have made valuable contributions to our social, economic, and political structures, and I am confident that thoughtful and objective citizens will continue to suggest worthwhile concepts and changes. Responsible critics are those who get their facts straight. They insist on satisfactory and complete answers to their concerns. And when such satisfaction is provided, they react with integrity.

I am afraid, however, that some have confused the real thing with its opposite number and have spawned a school of thought which subscribes to the tenet of "criticism for the sake of criticism." In turn, this has enhanced an environment and sensationalism where innuendo and insinuation have then been substituted for reason and rational judgement. Frankly, I believe we can all profit from the adage, "Come, let us reason together."

One problem which is evident in a number of the current controversies is a feeling on the part of some that every determination that is made must be based upon all of the possible data which could be gathered pertinent to the determination. I believe that Judge Arraj put it very well in his decision of March 1970 concerning the Rulison case in the District Court in Colorado when he said:

"The field of radiation protection is constantly changing with the appearance of new scientific knowledge on the biological effects of ionizing radiation. Careful decisions must be made in the context of contemporaneous knowledge. Such decisions cannot be indefinitely postponed if the potentials of atomic energy are

to be fully realized. All that is required to establish reasonableness of the decision setting a standard under the statutory directive to protect the public health and safety is that it be made carefully in light of the best of available scientific knowledge. Absolute certainty is neither required nor possible."

This philosophical point of view is applicable to many human endeavors -- beyond the field of expertise which brings your group together for this conference.

## RADIATION PERSPECTIVE IN THE UNITED STATES OF AMERICA

Ralph Nader  
Public Interest Research Group  
Washington, D.C.

I would like this morning to discuss briefly some of the issues that nuclear power plants and x radiation in particular pose to the consumer movement in the United States. After all, consumers are to be either the prime beneficiaries or the prime victims of this source of risk and, until recently, they have not been even minor beneficiaries of adequate information or adequate participation in the decisions that have been made.

Our country rests its premise about our economic and governmental structure on citizen and consumer participation - not just at elections, but in between elections - not just on Congress, but on regulatory agencies at the Federal, State, and local level. Over the years a great deal of faith has been placed in the scientific and governmental establishments dealing with the use of x radiation. There has been a great deal of faith in the policymaking apparatus dealing with the development and promotion of peaceful uses of atomic energy. That faith has rested on ignorance. It has rested on as absolute a delegation of citizen responsibility to government and to professionals as has ever occurred in the history of the United States.

In the last few years the hazard of deploying faith based on ignorance has become increasingly apparent. It started with the very late revelation of the occupational risks to uranium miners; a hearing, which the Joint Committee on Atomic Energy finally held in 1967, bringing out facts which should have been brought out years earlier. This revelation was followed by a series of disclosures relating to medical and dental x-ray exposure in the United States Congress, preceded by a number of articles by a few health physicists and radiological technicians who spoke at their own professional peril.

The Radiation Control Act of 1968, supposedly put the Department of Health, Education, and Welfare into the business of setting standards for medical and dental x-ray machines and taking the lead in upgrading the performance of both technicians and medical personnel and used machinery; but that Act has suffered in its aftermath from the same kinds of deficient constituency and awareness that led to its enactment.

As far as we know, x radiation from medical and dental machines present the bulk of present radiation exposure to the American population. We know that dentists and doctors and their assistants handle these machines in a framework where the patient or the consumer is not expected - is not encouraged - to ask questions, much less to receive the answers about how well these machines have been checked, about how qualified are the operators of these machines, and about a number of other issues dealing with the use of these machines in diagnosis and treatment.

It has not been realized throughout the public that when it comes to radiation, ionizing radiation, the old saying that "every little bit counts" is directly applicable. Another phrase might also be applicable; that when it comes to this kind of radiation, "even if it doesn't pinch, it hurts."

Unfortunately, the public views trauma and other forms of industrialized violence largely as it has viewed street crime; if these do not provoke immediate sensory pain and anguish, they do not provoke public concern. Tornadoes and fires provoke immediate pain and anguish; low level radiation, with a few exceptions, does not. Radiation is a form of new and silent violence, at least as it comes from an industrialized economy, and not natural background. It is a new and silent form of cumulative violence that does not provoke immediate pain or anguish, therefore does not produce any immediate concerned constituency among the public. This means that our intellectual approach has got to be one that analyzes risks and probabilities and alternatives and remedies and that unlike street crime, it can not rely on a visceral and sensory response for the public's arousal and concern.

The medical and dental x-ray experience relating to the Radiation Control Act of 1968, I think alerted many people in this country that the assurances that had been given for years just weren't so. Instead, there is a disgraceful lack of systematic inspection, effective design, and of operating care; and the patients are exposed to ten times or more levels of ionizing radiation, unnecessary to fulfill the functions of the x ray in terms of taking a clear and adequate picture. This astounded many people because the professional corps had dissented very little up to that point. However, when this dissent began, the processes of Congressional hearings and legislation moved with almost incredible swiftness. That's a good lesson, perhaps, for health professionals to learn; that when knowledge is focussed on the Congress by people who have been working in these areas, who have not felt sufficiently free to be candid, that knowledge can be translated into policy awareness. I know no other institution in the U.S. Congress that has been more deprived, and that has delighted in being deprived of knowledge, of risks, probabilities, and alternatives, than the Joint Committee on Atomic Energy.

This abdication leads to the second section of my remarks relating to nuclear power plants. Nuclear fission power plants, fission; including the breeder proposal, have suffered the pain of a certain type of success. Whenever a new technology delivers its commercial objective (in this case, electricity), the risk of developing that technology in an imbalanced way increases when it relates to the costs to present and future generations of such a technology. It runs the risk of imbalancing the development of alternative technologies for energy because of its unquestioned success in producing electricity.

Once again the same pattern was repeated; a corps of professionals and government officials and later members of industry were built up around the promotion of nuclear power. The Atomic Energy Commission had the dual and conflicting role of promoting this peaceful use of atomic energy and safeguarding it at the same time. It embarked on a massive research and development project, benefiting from weapons research, as well as the nuclear reactor R&D program, and then transferred this knowledge and this hardware to the private utilities and the reactor manufacturers, directly and indirectly. Then the peculiar jargon of the atomic energy world began to be developed. In the 60's when questions were asked by the very few, the answers were "the risks are negligible," or "although there is a possibility of an accident, it is very, very unlikely to ever happen."

It is interesting to note that the jolt of the atomic energy establishment came almost accidentally from two sources: (1) Conservationists' concern over thermal pollution and (2) a statement by Dr. Ernest Sternglass about the number

of people, in terms of present and future generations, who would be seriously exposed to radiation from these plants. His estimates were considered so outlandishly wild that the Atomic Energy Commission commissioned Drs. John Gofman and Arthur Tamplin, members of an AEC laboratory, to refute him. In refuting him, they still came out with a level of risk estimate that was still considered outlandish, particularly by the Atomic Energy Commission.

It is interesting to know that those two controversies are no longer the most serious ones relating to nuclear power plants; that the controversies now have spread into almost every nook and cranny of this technology. What most provokes the AEC, the reactor manufacturers, and the utilities are questions by the public that have not been answered. The response of the Atomic Energy Commission to these questions has been filled with innuendo, insinuations, and sensationalism, and a great deal of secrecy. The AEC has not accepted a critical and cardinal foundation of our system of law - from the English common law to the present-day statutory law - that the burden of proof for going forward with a technology is on the promoter, not on the potential victims.

And so in the following areas we are faced with grave and unanswered questions and often grave and totally insupportable performances:

1. Power Plant Siting Policy. We have only to look to England to see how disastrous our siting policy has been in locating these plants in the suburbs and near large metropolitan areas. In England, I understand, the siting policy has been to locate nuclear power plants much further away from metropolitan areas. In Sweden, we have the reverse; there is even a proposal to build one of these plants in downtown Stockholm. Moreover, plants have been designed or are already built only a few miles away from major Swedish cities. Siting, a classic illustration of what happens when the government ignores the people in terms of their rightful role in decision making. They are the ones who have to live near these plants. They are the ones who will receive the most intense exposure in case of an accident.
2. Meltdown and Emergency Core Coolant System. This problem has received the most publicity; the problem of meltdown and emergency core coolant system. It is interesting to note that this issue was brought to public attention by a group of scientists, lawyers, and environmental citizens whose action led to the public hearings by the Atomic Energy Commission, which led to the first opportunity for a number of Atomic Energy Commission scientists to say what they should have said, and perhaps what they wanted to say publicly months, if not years prior, about the inadequacy of the Emergency Core Cooling System (ECCS) and the inadequacy of the safety and research program that was supposed to insure the ECCS's predictable deployment in an emergency.
3. Thermal Pollution. In this problem we have a very interesting conflict between State and Federal jurisdictions. I would expect a far more aggressive posture by the States in asserting their jurisdiction generally notwithstanding the recent Supreme Court decision denying it. That assertion may well come in the form of Congressional proposals as well as renewed State action.
4. Fuel Processing. On the question of the fuel processing plants, there has been probably less information publicly available than on the reactors themselves. Here the question of the transportation of radioactive materials is a critical one. A recent General Accounting Office (GAO) report noted a number of deficiencies dealing with the containerization and transportation practices. The GAO, as has been its tradition since the clampdown by Congressman Chet Holifield in

1965, does not name names. The GAO, however, does, and did in this instance, point to a number of unknowns, hazards, and deficiencies to which the Atomic Energy Commission should pay heed. But that, to me, is a drop in the bucket in terms of further inquiry by the Congress, hopefully, about the transportation risks here and the exposure of radioactive materials to waylaying nuclear highwaymen.

5. Waste Disposal. The problem of waste disposal is perhaps the most nagging technical problem of all. Once again, we have a classic illustration of going forward with the benefit of the technology and waiting and hoping and believing that the waste disposal problem will be solved sometime along the way. The containment of incomparably deadly materials such as Plutonium-239 from the environment for half a million years is a worthy task only if the society's survival is at stake. Instead, such waste makes society's survival the issue.
6. Earthquakes pose a problem which has plagued the utilities in California.
7. Sabotage and Theft pose a considerable risk.
8. Economic Costs of maintaining these plants are rising and causing concern among the utilities.

Witness the recent speech of Mr. Rodus, who is the President of Consolidated Edison in New York City and the more recent public discussion of the insurance of these plants. It is interesting to note that in our country's history, insurance coverage has always been considered a deterrent to hazardous practices in the sense that if the technology is too dangerous, it won't be able to get insurance. and if it can't get insurance, it would not operate. But, once again, the foresight of the nuclear power establishment led to the enactment in 1957 of the Price-Anderson Act, which limited the overall liability of a nuclear power plant accident to a current \$560 million level, with the private insurance participation under the \$100 million level. The inadequacy of this insurance coverage may be compared with a 1965 Atomic Energy Commission sponsored report (it was not released until this year), that a big accident in an average sized nuclear plant would lead to 45,000 deaths, over 100,000 injuries, and \$17 billion worth of property damage. Seventeen billion dollars worth of property damage is a long way from a limited \$560 million dollars that the Price-Anderson Act stipulates.

There have been throughout an increasing level of "leakage," not only of radioactive waste, but of Atomic Energy Commission documents. These documents have been showing that there have been people inside this world of atomic energy who have been very seriously concerned, but who have not felt free enough to speak out. The words "near misses" have occurred in these documents and, certainly, there is now quite a long list of "near misses," quite a long list of minor accidents that could have been big accidents.

It is perhaps instructive to our country that there would not be a single nuclear plant in operation today if it were left up to the free market. The nuclear technology would not have been developed without government (that, perhaps, is not too astounding, given the investment levels required). More significantly, however, is that these nuclear plants would not be in operation today without government interference via the Price-Anderson Act. As Commissioner Herbert Dennenberg of Pennsylvania, and the State's insurance department, said at recent hearings, and as Professor Harold Green also stated at those same hearings, "without the Price-Anderson Act, without the limitation of liability, no utility could afford to operate a nuclear power plant."



And so, the consumer pays going and coming. The consumer pays through his or her taxes for this \$30 billion R&D program developed since World War II, and, without adequate insurance coverage the consumer pays in the event of an accident.

This brings me to the subject of the health professionals. I think it could be said categorically that there would not have been a Radiation Control for Health and Safety Act of 1968 without possibly three health professionals participating, led by Dr. Karl Z. Morgan, then of the Oak Ridge National Laboratory. The technical information which broke through the barriers of fraternal secrecy and reached the public was absolutely a prerequisite for Congressional concern and action in this area. The Act has not worked well, but at least there is the authority on the statute books to make it work much better.

Why haven't we been hearing more from health physicists in all these areas, particularly nuclear plant operational risks? I think one reason is that they are not given enough decision making power at the plant level or in the councils of government. Bureaucratic leaders still consider them to be technical minions rather than coequal participants in fundamental and derivative decision making processes. The need to develop an independent professional base for health physicists then is critical; that is, independent of their occupational or employment base. They will not be able to speak and to participate according to their best knowledge, if they are restricted by contemporary organizational pressures, both corporate and governmental. I would urge that health professionals in the health physics community seriously consider establishing an independent, technical base of technology assessment and advocacy according to the highest canons of their professional mission and relating this work not only to nuclear plants, medical and dental x rays, but also to new consumer products that raise questions industry is not constrained to answer.

Now, in conclusion, the usual evaluation of nuclear power plants has been in a benefit-risk frame of reference with the benefit considered great and the risk "negligible." I think there are a number of areas we can all agree on verbally; whether we can agree in terms of action is another thing.

The first is that there is an intolerable level of secrecy surrounding this matter, which only recently has been broken by court decision, by deposition, by externally forced public hearings, and by information leaks. The suppression of the 1965 AEC estimate of damage from a nuclear power plant accident is a significant case in point of such secrecy, particularly the internal dialogue about how to release this information, whether to release it, how much to give to the Joint Committee on Atomic Energy, and so forth.

Second, we need a far more aggressive posture by the Joint Committee on Atomic Energy. Some people in the consumer and environmental areas believe that the Joint Committee on Atomic Energy has permitted such a deterioration in this area that it now has as its opportunity the prospect of saving this country. Nuclear reactor peril is, of course, of tremendous consequence to not only present but future generations. The Joint Committee might well heed Alfred Whitehead's memorable dictum when he counseled the scientific world to always have before them the option for revision -- the option for revision of theories or hypotheses.

At the present time, the Atomic Energy Commission, the reactor manufacturers and the utilities have not proved their case. Some might say they have only done so in a negative way. But, they have not proved their case, and the right of the consumer to have these questions answered so that the consumer realizes (1) what the risks are to himself and to future generations (he or she might care about their children and grandchildren) and (2) what the alternatives

are in terms of other energy sources, greater energy conservation and more explicit energy allocation; and he might also want to know what his participation rights are when a nuclear power plant or a waste disposal depot is proposed for construction under the licensing procedure.

Philosophically one might ask the following question: "Under what conditions should a society ever engage in the deployment of a technology which has to be perfect forever, such as a nuclear power plant, because the alternative is massive social disaster now and into the future?" In my judgment, the only condition when a society should engage in such a deployment is its utter survival, and nuclear power is not utter survival. Nuclear fission power is one approach. There are other approaches - solar and geothermal among them.

Perhaps the greatest cost of putting all our energy eggs for the future in one fragile nuclear basket is that the government has not deployed its billions in developing other options, whether these be geothermal, solar or until recently, fusion energy, or other alternatives. This is why I think the United States, as the leader in nuclear power development and sales, has got to rethink its program; rethink how it allocates its tax dollars for developing other energy sources, and to rethink its obligation of aggressive sales of nuclear reactors with export bank financing to underdeveloped countries where the technical infrastructure is far, far less adequate to care for estimable risks from the siting to the waste disposal of radioactive materials.

I would like to conclude with one hope directed to Chairman Melvin Price of the Joint Committee on Atomic Energy. He is aware of my disagreements with Congressman Holifield, and I am sure he is aware that the Joint Committee has not opened up its hearing processes to consumer and environmental testimony as it now intends to do under his direction in October. As of January 1964, these hearings have neither been held nor scheduled. But, I would hope that if Chairman Price is not convinced by recent disclosures of the need to consider a "go-slow" program at the very minimum, that he will at least focus on the need to ask the tough questions of the Atomic Energy Commission and to demand the answers openly and on the public record for the scientific, environmental, consumer communities to scrutinize, to cross-examine and to follow up. The risk is not one to which only a selected few are exposed. It is a risk for mankind.

## PERSPECTIVES ON RADIATION RISKS

Ralph E. Lapp  
Industrial Consultant and Science Writer

Gertrude Stein, looking over your program for this conference, might well sum it up, "A man-rem is a man-rem is a man-rem." This might be an assault on Women's Lib -- a double assault if the feminine perception unmask the wiles of the roentgen-equivalent-man.

It seems to me that full understanding of the man-rem concept will make society aware of the need to balance radiation risks. We must be concerned with the man-rem as a unit of national exposure and conclude that the consequences of radiation risk-taking should be evaluated independently of the radiation source. We should not discriminate between man-rem from radioactive fallout, from nuclear power plant effluents, or from diagnostic x rays.

The BEIR<sup>1</sup> report which your organization has so wisely taken as the main topic for discussion today needs to be translated from the technical jargon of the radiation specialist and injected into the public discourse. This is not an easy undertaking, for the nature and dimension of things familiar to scientists is strange territory for the average person.

### The BEIR Translation

The great value of the BEIR report is that it systematically reviews the data on somatic and genetic effects of ionizing radiation and provides us with an understandable linkage between radiation dose (man-rem) and injury to humans. Your organization will be discussing the genetic effects in detail, so I shall confine myself to somatic effects. Here the BEIR translation reads:

"Continuous exposure of a population of 1 million persons to the level of 1 rem per year may result in the incidence of 150 to 200 cancer deaths per year."

I shall make the assumption the 200 figure is valid.

You have titled this session "Radiation Perspective in the United States of America," and I shall proceed to address this issue on the following basis:

- (1) That the BEIR man-rem dose to cancer-death response applies to very low levels of radiation.
- (2) That perspective may be achieved by extrapolating human exposure data to the year 2000.

I shall not attempt to leap into the 21st century, even though it may happen that certain sources of ionizing radiation released in this century will persist into the next. I plan to rely on the U.S. Environmental Protection Agency report ORP/CSD 72-1, "Estimates of Ionizing Radiation Doses in the United States 1960-2000," as a data base with some modification. I shall use 1 million man-rem as a basic unit and disregard all sources of dose very much less than this unit.

### Natural Background Radiation Risk

The average whole-body annual dose due to the natural background radiation in the United States is taken to be 0.13 rem. Cosmic radiation dose increases with altitude and is taken as 0.045 rem per year for the United States. Radiation emanating from terrestrial sources external to man averages about 0.06 rem per year, while the internal radiation dose, primarily  $^{40}\text{K}$  and  $^{210}\text{Po}$  plus  $^{222}\text{Rn}$  averages 0.025 rem.

Within the 50 States the external dose to man varies from a high of 0.225 rem per year in Colorado to a low of 0.075 in Louisiana. Adding in the internal dose contribution, we see that the natural background radiation varies from about 0.1 to almost 0.3 rem per year within the borders of the United States. Man, of course, alters his environment and lives in structures of varying radioactive content, thus increasing his external dose.

The present U.S. man-rem dose from natural sources of radiation is taken as 27 million man-rem, and by extrapolation to a year 2000 population<sup>2</sup> of 280 million I arrive at an end-of-century dose total of 36 million man-rem. The cumulative 30-year dose approaches 1 billion man-rem, and this equates to almost 200,000 cancer deaths. This figure represents about 1 percent of the spontaneous cancer deaths due to all causes.

One might be tempted to look for some correlation in the cancer deaths in a city like Denver and a sea-level city with half the natural background, but this is a signal-to-noise problem that taxes the ingenuity of the biostatistician. Moreover, it turns out that lower level cities may exhibit higher rates of cancer mortality.

### Radioactive Fallout Risks

Global drizzle or protracted fallout from debris introduced into the stratosphere by atmospheric tests of nuclear weapons is, like the natural background, an unavoidable source of radiation exposure. Beginning with the high-yield megaton-class weapons detonated in 1952, there have been additions to this stratosphere burden of radioactive material. France and China continue to add to this source of global contamination, but the other nuclear powers agreed to a Limited Nuclear Test Ban in 1963. It will be recalled that grassroots support for this ban came from widespread public fear of fallout, symbolized by strontium-90.

Strontium-90 is a bone-seeker having a radioactive half-life of 29 years (138 curies per gram). A person born in 1954, the year when the U.S. initiated high fission-yield megaton tests in the Pacific, would accumulate a dose to the bone of about 1.2 rem by the end of the century.

Cesium-137, half-life = 137 years, deposited on the ground is the major source of external radiation from fallout. Other shorter-lived emitters contributed to the human dosage in the 1950s and 1960s. For example, in the U.S. in 1963 the total gamma radiation (external) plus dosage due to internal uptake of fallout nuclides produced a 0.013-rem dose per U.S. individual. This decreased to 0.004 rem in 1970, and it is assumed to increase slowly to 0.005 rem in the year 2000. The present 1-million man-rem dose per year is expected to reach 1.4 million man-rem at the end of the century. The 30-year dose is estimated to be 34 million man-rem, corresponding to a total of 6,800 cancer deaths of fallout origin.

### Radiation Risks in Jet Travel

The demands of modern living make it almost an involuntary act to travel by jet aircraft. Scheduled airlines in the U.S. customarily seek altitude in the 25,000- to 35,000-foot altitude range or higher for purposes of passenger comfort and fuel conservation. At such altitudes passengers are exposed to an average of roughly 0.004 rem per hour<sup>3</sup>. In 1971 domestic air travel in the U.S. amounted to 106 billion passenger revenue miles on scheduled carriers<sup>4</sup>. Air travel of this type increased over tenfold in the past 20 years<sup>5</sup>, and on the basis of a recent Department of Transportation report<sup>6</sup> I estimate that the annual radiation dose to U.S. air travelers will reach a 2-million man-rem total by the year 2000.

A 2-million man-rem dose per year would mean an annual cancer fatality rate of 400. To put this in perspective, CHART I illustrates the historic pattern of airline accident mortality. The lower curve plots the annual deaths due to accidents on scheduled airlines and indicates that the public is willing to accept airline fatalities at a rate of about 200 per year. The upper curve records the annual fatalities experienced in U.S. civil aviation; it would indicate that private parties are willing to accept an almost tenfold higher annual level of air fatality.

Extrapolation of the scheduled carrier mortality rate to the future is problematic, but with the increasing dependence on high-density flights, partly as a result of diminished availability of jet fuels in the future, the United States might well experience annual fatality rates approaching 1,000. At such a level the radiation risk would be less than half that for fatal accidents, and presumably the Surgeon General would not require imprinting "AIR TRAVEL INVOLVES RADIATION RISKS HARMFUL TO YOUR HEALTH" on your air ticket.

### Medical Diagnostic Radiation Risks

The present annual dose from medical diagnostic practice in the U.S. exceeds 15 million man-rem. Assuming that there is no significant change in the use of x rays as a diagnostic tool, then it is expected that the national dose will reach 20 million man-rem by the year 2000. A 30-year total of somewhat more than 500 million man-rem corresponds to 100,000 cancer deaths, although it is true that not all can be considered "extra" since the radiology might not be specific to the patient's cancer.

Assuming that all 100,000 cancer deaths are actually iatrogenic, it is pertinent to place some sort of dollar value on a human life. This is an uncertain calculation, but there is legal precedent for estimating such a dollar value in court cases. Often an estimate of 20 years life-income denied is used, and awards in the range of \$300,000 are made. (Assessment of the lifelong cost of maintaining a genetic defective would involve much larger sums, but only somatic radiation effects will be considered here.) On this reckoning the societal cost of excessive x radiation may be computed. One, of course, has to make some estimate of "excessive," and I shall assume that 50 percent of the diagnostic dose is unnecessary and that proper technique and well-regulated x-ray equipment could produce the desired diagnostic results. With no attempt at precision, but only to scope the problem, I estimate that 250 million man-rem multiplied by \$60 per man-rem represents a \$15 billion cost in the United States for the 1970-2000 period.

### Nuclear Power Effluent Risks

Here in the United States there has been rather heated controversy about the radiation risks posed by nuclear radioeffluents. It should be understood that routine release of radioactivity from a nuclear power plant is basically a matter of fuel clad failure with subsequent entry into the primary coolant of certain fission products. Release of short-lived noble gases is subject to temporary holdup to reduce the effluent activity. All U.S. nuclear power sites are subject to independent radiation monitoring, and the environmental surveys of the environs are available to the public. Critics<sup>7</sup> of nuclear power have charged that nuclear power effluents would permit irradiation of the U.S. population to the extent that 32,000 extra cancer deaths would be incurred annually.

The Atomic Energy Commission published earlier this year a projective evaluation of the radiological consequences of a large regional nuclear power plant operation in the year 2000 after having circulated a draft report of the study to critics. This WASH 1209 report<sup>8</sup> concludes that, "The average radiation potentially received by the total body of an individual in the study area in the year 2000, resulting from the operation of the assumed facilities, was calculated to be 0.17 millirem."

This AEC projected dose rate would yield 0.05-million man-rem dose to the U.S. population in the year 2000, corresponding to 10 cancer deaths per year. The 1970-2000 total would be about 90 cancer deaths associated with nuclear power, according to the dose-risk relation of the BEIR report.

The question of dose commitment of long-lived radionuclides introduced into the environment by the year 2000 is beyond the scope of my discussion. However, I call your attention to two very useful treatments of the nuclear fuel cycle:

ENVIRONMENTAL SURVEY OF THE NUCLEAR FUEL CYCLE, U.S. Atomic Energy Commission, Directorate of Licensing, November 1972.

SITING OF FUEL REPROCESSING PLANTS AND WASTE MANAGEMENT FACILITIES, Oak Ridge National Laboratories Document ORNL-4451, July 1970.

### Nuclear Power Plant Accident Risks -- Siting Policy

Public opposition to nuclear power has recently concentrated on risks associated with a catastrophic accident; i.e., a Class 9 accident according to the AEC's 1 to 9 classification of reactor accidents. I cannot do more than survey some of the highlights of this problem of a low-probability high-consequence accident. I call to your attention the fact that the Atomic Energy Commission will publish sometime next year a detailed analysis of Class 9 accidents, including both the probability of accidents and estimates of the extent of their consequences. In 1957 the AEC did bring out WASH-740, "Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants," but to use a Washington word, this report is considered "inoperative" today.

The lack of an authoritative and realistic evaluation of radiation risks attending major reactor accidents has allowed antinuclear spokesmen to seize upon the "worst case" postulated by the WASH-740 report and to focus on it as a probable occurrence. It is true that power reactors today are seven times more powerful than the 500-megawatt plant assumed in the AEC's 1957 study. Moreover, populations at risk in the vicinity of nuclear plants have grown since WASH-740 was published. CHART II, for example, illustrates the cumulative populations at risk near selected nuclear power sites. The Calvert Cliffs site on the

Chesapeake 45 miles from Washington, D.C., has a population at risk beyond 5 miles, very much less than that assumed in the WASH-740 analysis. On the other hand, Indian Point north of New York City represents a considerably greater risk in population distribution for the Burlington site near Philadelphia; this was not approved by the AEC. Currently pending is a construction permit application for Newbold Island reactors sited 6 miles from the defunct Burlington site. I have publicly opposed approval of this siting on the basis that the Atomic Energy Commission has not placed in the public domain evidence of nuclear safeguards reliable enough to compensate for the greater population at risk in the Newbold Island area. If the AEC approves Newbold Island, then the way is open for other utilities to press for closer metropolitan siting.

It is true that the AEC has issued its WASH-1250 report, "The Safety of Nuclear Power Reactors and Related Facilities" (July 1953), in response to the October 1971 request of the Joint Committee on Atomic Energy. This will serve as the basis for public hearings to be chaired by Congressman Melvin Price, beginning September 25th. But the WASH 1250 report does not deal with a Class 9 accident and its consequences.

On March 23, 1962, the AEC issued document TID-14844, "Calculation of Distance Factors for Power and Test Reactor Sites," as a guide for utilities to determine:

- (1) An exclusion area such that an individual exposed at the boundary fence would not receive more than 25 rem whole-body dose in 2 hours or more than 300 rem dose to the thyroid.
- (2) A low population zone such that cloud passage would not deliver more than 25 rem (whole-body) or 300 rem (thyroid) dose at its outer boundary.
- (3) Population center distances specifying distance to centers of large population concentrations.

This AEC siting criteria document did not take a conservative view of thyroid dosage since members of this organization recognize that 300 rem is a significant dose to the thyroid as evidenced by the experience with Rongelap natives exposed to fallout from the March 1, 1954, Bikini bomb test. I maintain that the AEC needs to put forth an updated version of its siting criteria.

#### Class 9 Probabilities

The Wash-740 report had very little to say about the probability of a major reactor accident. In the past few years there has been increased emphasis on probabilistic assessment of reactor risks. Reactor designers are trying to define how safe their nuclear machines are, and this they do in terms of postulating a spectrum of initiating events and following their consequences sequentially as they affect the pathway leading from the nuclear fuel pellet to the environment.

The only way for the fuel pellet's stored radioactivity to escape from the core is to overheat to the point where the clad deteriorates and fission products are released within the core. Pellet heatup can occur as a result of Loss of Coolant Accident (LOCA) unless coolant is resupplied to the core channels. The AEC requires that U.S. nuclear power reactors be equipped with Emergency Core Cooling Systems (ECCS) as safeguards to prevent fuel melting. If  $P_1$  is the probability of a LOCA and  $P_2$  is the probability that ECC systems will not prevent fuel melting, then the probability  $P_{12}$  of a LOCA + ECCS failure is  $P_1 \times P_2$ . Nuclear vendors estimate that the probability of a pipe break is about

$10^{-4}$  per reactor year and the chance that ECCS will fail to perform its function is  $10^{-3}$  per reactor year. Thus,  $P_{12}$  becomes  $10^{-7}$  per reactor year.

This vendor-estimated low probability for LOCA + ECCS failure has to be compounded by an additional probability; i.e., failure of containment safeguards, before the meltdown-released radioactivity is released to the atmosphere. Here one has to deal with a complex problem of containment failure in various modes affecting the time-release of specific fission products. For example, water sprays could effectively reduce the release of iodine-131.

I might interpolate at this point that for ocean-sited nuclear power plants the Advisory Committee on Reactor Safeguards (ACRS), which makes an independent review of each reactor application for licensing, has under discussion a requirement for a core-catcher to prevent loss of meltdown fission debris through containment.

Let us assume that vendor estimates are wrong by a factor of 10 in each  $P_1$  and  $P_2$  estimate; i.e.,  $P_{12} = 10^{-5}$  per reactor year. In other words, with 100 reactors operating, as will soon be the case in the U.S., the overall probability for a major reactor accident of a Class 9 type would be one chance in a thousand per year.

#### Class 9 Radiation Consequences

Would the public accept a risk of one in a thousand per year when 100 reactors are operating? It's premature to extrapolate this to one in a hundred per year when 1000 reactors are operating because that's several decades from now, and presumably new and more reliable safeguards will be available then. When the AEC grants an operating license to a utility to run a nuclear power reactor, it presumably makes a judgment that the public risk of accident comes within acceptable limits. But these limits have not been defined for the American people.

If one were to take a public opinion poll of Americans inquiring into attitudes about the probability of an accident to a nuclear reactor, I suspect that people would immediately ask, "How serious an accident?" In attempting to answer this question, we have to go back to our chain of probabilities; i.e., to  $P_{123}$  where  $P_3$  is the probability for release of X curies of various radionuclides through containment.

We must now introduce three additional probabilities:

$P_4$  = that allowing for the meteorology prevailing at the time of the accident release.

$P_5$  = that governing the population distribution in the downwind area of the nuclear site

$P_6$  = shielding factors reducing exposure to people in the downwind sector

Certain pessimistic values for  $P_3$  and  $P_4$  led to high-consequence estimates in the Wash-740 report, leading to an extreme projection of a lethal dose as far out as 15 miles from the accident site. I think that when one is dealing with a probability that is itself a compounding of six separate probabilities; i.e.,

$$P_{123456} = P_1 \times P_2 \times P_3 \times P_4 \times P_5 \times P_6$$

it is easy to postulate extreme consequences by taking very high values for each individual probability. In fact, some antinuclear spokesmen put  $P_1 \times P_2 = 1$ ,



then assume almost complete venting of the fission products and couple this with the highest possible values of  $P_4$ ,  $P_5$ , and  $P_6$ . Thus, they arrive at most improbable and in some cases impossible fallout patterns blanketing a state as large as Pennsylvania.

Allow me to cite a single example of such nuclear extremism. I have here a letter signed by Dr. John W. Gofman for the Committee for Nuclear Responsibility, Inc. (July 1972) on a letterhead listing the names of four Nobel Prize winners. It begins:

"Dear Friend,

Do you live within 100 miles of the locations indicated on the attached map? If so, you also live, or will live, within deadly range of a nuclear power plant . . . "

CHART III is adapted from the most recent AEC report on accident (Class 9) consequences, "An Evaluation of the Applicability of Existing Data to the Analytical Description of a Nuclear Reactor Accident--Core Meltdown Evaluation," BMI-1910 (July 1971). The upper curve represents a pessimistic core meltdown accident,  $P_1P_2 = 1$ , but assigned no probability in the report; and  $P_3$ , involving pressure vessel meltthrough and 50-percent fission product escape from the core with a 2-hour delay for release of the noble gases and inversion conditions for  $P_4 \times P_5$ , is not defined because the graph is for individual dosage and  $P_6 = 1$ . To illustrate the impact of a single variable ( $P_4$ ), I have plotted as the lower curve an estimate of how the dose distribution would look under average conditions of meteorology<sup>9</sup>. Note that the lethal distance (radius for LD<sup>50</sup> dose of 500 rem) is less than 1 mile.

#### Indian Point and Calvert Cliffs Estimates

Although it is not at all clear, based on AEC publications to date, as to which radionuclide dose is of greatest lethality in a Class 9 accident (i.e., noble gas external dose, ingested dose to the gut, radioiodine dose to the thyroid), for the purpose of illustrating the significance of multiplying  $P_4$  by  $P_5$  I shall apply the two curves in CHART III to a population distribution for the Indian Point and Calvert Cliffs site. I shall assume a uniform distribution of population in all directions from the reactor site. I deduce the following population doses:

<u>Meteorology assumed</u>	<u>Inversion</u>	<u>Average (millions of man-rem)</u>
Indian Point	10	1
Calvert Cliffs	1	0.1

Naturally, the real values for the population dose would depend on the wind direction. In the case of Calvert Cliffs a westerly wind would mean a tenfold or more reduction in dose. The man-rem dose for an accident situation cannot be directly related to dose as estimated for other sources of radiation since it is a single-shot affair. In making any such comparison, the doses given for the accident situation need to be reduced by a factor of 10.

So far I have said nothing about  $P_6$ . If we regard the rem dose as an open-field measurement, then we have to introduce shielding factors due to housing and the body itself. These are generally assumed to be 0.4 and 0.8 so that the effective dose is about one-third that of open-field dose. In an emergency

situation (remembering that a reactor accident could involve several hours of warning for much of the community nearby) there is the option of evacuation or of shelter-seeking.

To the best of my knowledge, the various States and cities in the U.S. have no plans for a Class 9 accident. The emergency plans I've seen are patterned to Class 8 situations which do not pose very serious radiation risks.

#### Comparison of Radiation Risks (1970-2000)

I shall now summarize the dose estimates for various sources of radiation thus far annualized.

<u>Source</u>	<u>30-year dose (millions of man-rem)</u>
U.S. natural background.....	1,000
Weapons test fallout.....	34
Domestic jet travel.....	36
Medical diagnosis.....	500
Routine nuclear effluents.....	0.45

It will be noted that test fallout and nuclear effluents plus waste products have dose commitments persisting beyond the year 2000.

There's nothing much we can do about the first two items in this tabulation, and since the third item is not a dose to the total population, I shall concentrate on a discussion of the radiation risks of medical diagnosis and nuclear power facilities, bearing in mind the admonition of the BEIR report (page 7):

"An additional important point, often overlooked, is that even if the benefit outweighs the biological cost, it is in the public interest that the latter still be reduced to the extent possible providing the health gains achieved per unit of expenditure are compatible with the cost-effectiveness of other societal efforts."

#### Nuclear Safety Costs

AEC technical specifications for power reactors force U.S. utilities to spend about \$40 million in capital costs and operations for added safeguards per 1,000-megawatt installation to provide a wide margin of safety against accidents. These costs are in addition to those that a utility would pay for normal insurance against damage to the reactor. One thousand reactors expected by the year 2000 would therefore entail expenditures of \$40 billion. In addition, I would estimate that the AEC, EPA, and HEW will probably spend up to \$10 billion on safety and radiation control.

#### A Double Radiation Standard?

In 1972 the 50 States spent a total of \$7.2 million implementing radiation controls, not all of which apply to the diagnostic use of x rays. Considering the very much greater population dose associated with medical diagnosis as compared to nuclear power dosage, it seems to me that our society has a split vision on radiation risks and is setting up a double standard for radiation risks. I am not advocating relaxation of the As Low As Practicable radiation

limits set forth by the AEC, nor am I suggesting cutbacks in nuclear safety expenditures, but it does seem to me that some standards have to be applied to the dominant controllable radiation risk in America; namely, the diagnostic use of x rays.

According to my reckoning, the excessive use of x rays will mean 50,000 cancer deaths in the rest of this century. All of these can be avoided if we as a nation put radiation risks in perspective and establish rational restraints on that most lethal weapon -- the x-ray machine. I agree with Dr. C. L. Comar who proposes<sup>10</sup> that such standards "should be established in terms of minimal exposures required to fill society's needs."

#### Costs for Alternative Sources of Energy

Although opponents of nuclear power argue that there are environmentally and economically acceptable alternative sources of power to substitute for nuclear power, their proposals are not viable options for utilities before the year 2000. Competent energy experts are in agreement that the single candidate option is coal. It is therefore valid to reckon the costs of the coal fuel cycle, assuming that coal-fired plants replace nuclear units. I estimate that in the year 2000 such plants would require an annual boiler feed of 2 billion tons of high-rank coal.

Burning 2 billion tons of coal subjects a society to a risk-chain stretching from the mine to the smokestack. Let's consider, first, the occupational hazards in a coal-vs-nuclear comparison. I shall use data just made available by the Council on Environmental Quality<sup>11</sup>.

Assuming that almost all year 2000 coal is strip-mined, I estimate that the occupational costs of mining, processing, transporting, and using coal for 1,000 plants would total 2,640 deaths per year. The CEQ estimate for Light Water Reactor occupational risks is 153 deaths per year. In other words, the coal substitution would be 17 times more costly than nuclear risks in the year 2000 -- meaning the conventional risks of the uranium fuel cycle,

Two environmental hazards predominate in the coal cycle, the acres disturbed in strip-mining and the stack emissions, primarily SO<sub>x</sub> effluents. Uranium ore has a specific energy content up to 40 times higher than coal so the acres disturbed are very much less for present generation nuclear plants, and with the advent of the power-breeder, nuclear power will enjoy a thousandfold or more advantage in reduced environmental impact as compared to coal.

The 1975 primary and secondary standards on stack emission will crack down on SO<sub>x</sub> effluents from coal-burning plants, and it is to be assumed that use of compliance fuels and new developments in sulfur control will serve to make year 2000 effluents much lower in level than even the 1975 standard. But some SO<sub>x</sub> will come out of the stack, and the biological damage of chemical pollutants will have to be assessed. The 0.45-million man-rem 1970-2000 dose from routine release of nuclear power effluents sets a very high standard for fossil fuel plants to achieve.

#### Conclusion

I have attempted to put radiation risks in perspective and, in particular, to suggest that it should be a national objective to reduce the man-rem dose to the U.S. population and to bring radiation risks into better balance. I have also attempted to scope the problem of a nuclear reactor accident, and I would suggest that at your next Congress you devote major attention to the Atomic

Energy Commission 1974 study of this problem. In the meantime I would suggest that members of the International Radiation Protection Association have an individual responsibility to (a) persuade the medical profession to reduce the diagnostic dose and (b) act as brokers for the communication of reliable information to the public in matters of nuclear safety.

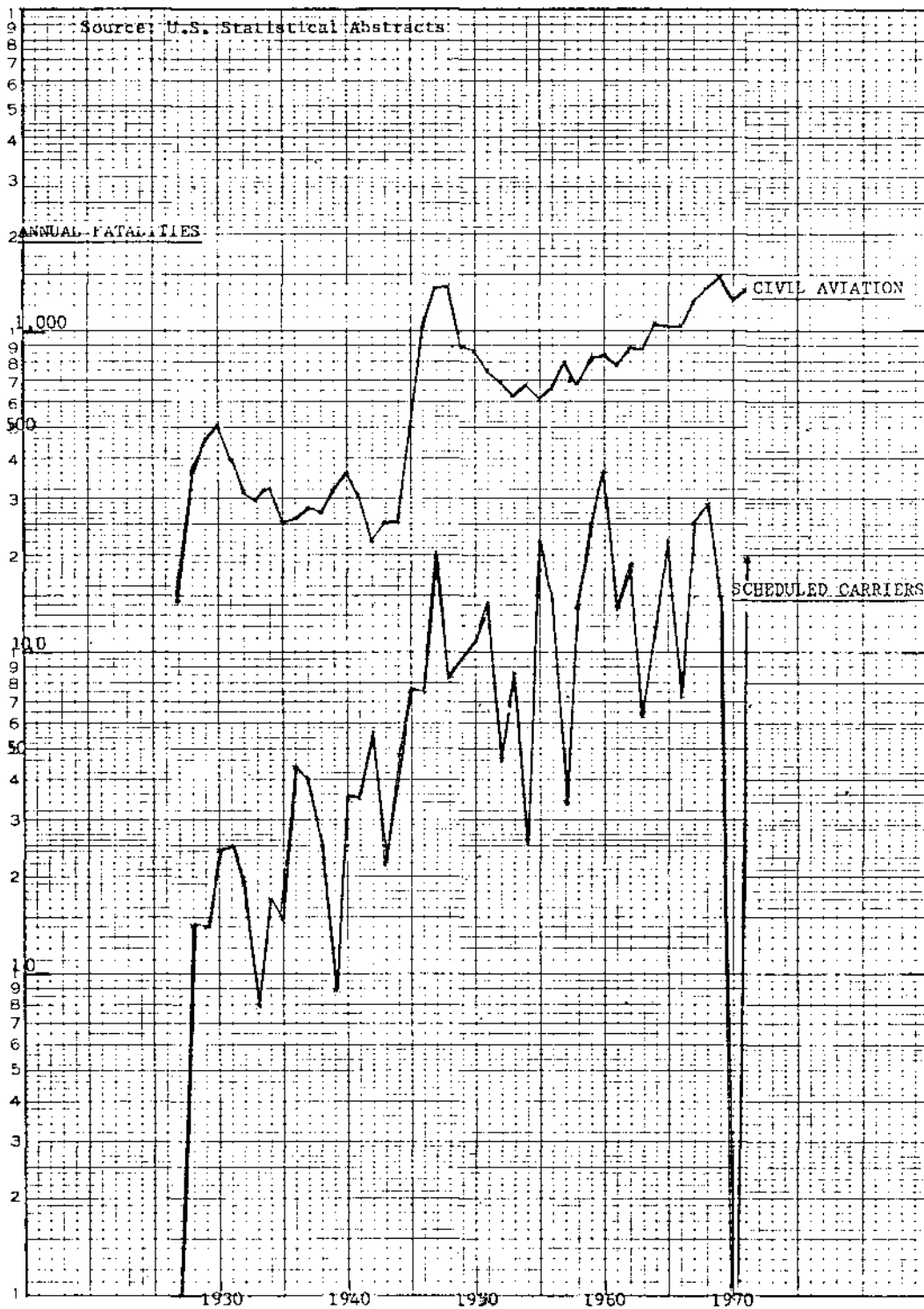
#### References

- <sup>1</sup> The Effects on Populations of Exposure to Low Levels of Ionizing Radiation. Report of the Advisory Committee on the Biological Effects of Ionizing Radiation, National Academy of Sciences.
- <sup>2</sup> The ORP/CDS 72-1 estimate is based on a year 200 population of 321 million.
- <sup>3</sup> H. J. Schaefer. Radiation in air travel, Science 173:780 (1971).
- <sup>4</sup> U.S. Statistical Abstract, Table 932 (1972).
- <sup>5</sup> R. E. Lapp. The Logarithmic Century, Figures 2-5, Prentice-Hall (1973).
- <sup>6</sup> Research and Development Opportunities for Improved Transportation Energy Usage, Figure III-1. Department of Transportation Report No. DOR-TSC-OST-73-14 (April 1973).
- <sup>7</sup> John W. Gofman and Arthur R. Tamplin, as cited in references listed on pages 204-5 of the BEIR Report, footnote 1.
- <sup>8</sup> The Potential Radiological Implications of Nuclear Facilities in the Upper Mississippi River Basin in the Year 2000. U.S. Atomic Energy Commission (January 1973).
- <sup>9</sup> Meteorology and Atomic Energy. U.S. Atomic Energy Commission Report TLD-24190 (1968).
- <sup>10</sup> Science 181:611 (August 1973).
- <sup>11</sup> Energy and the Environment. Electric Power (August 1973). (Data cited are taken from Tables A-2 and A-11).

- CHART I Historical Record of U.S. Air Fatalities (1927-1972). Data taken from U.S. Statistical Abstracts for recent years and Historical Statistics of the United States, Colonial Times to 1957 (Government Printing Office, Washington, D.C.).
- CHART II Population (Cumulative) at Risk Near Selected Nuclear Sites. Data are taken from U.S. AEC Public Dockets for Calvert Cliffs, from TID-14844, and from Figure 2 in article, "Siting Practice and Its Relation to Population," by H. B. Piper and F. A. Heddleson, to be published in Nuclear Safety, Vol. 14, No. 6 (1973).
- CHART III Whole Body Radiation Dosage -- Class 9 Accident. Upper curve is taken from Figure 14, page 31, BMI-1910 (July 1971). Lower curve is author's estimate based on data in fn. 9.

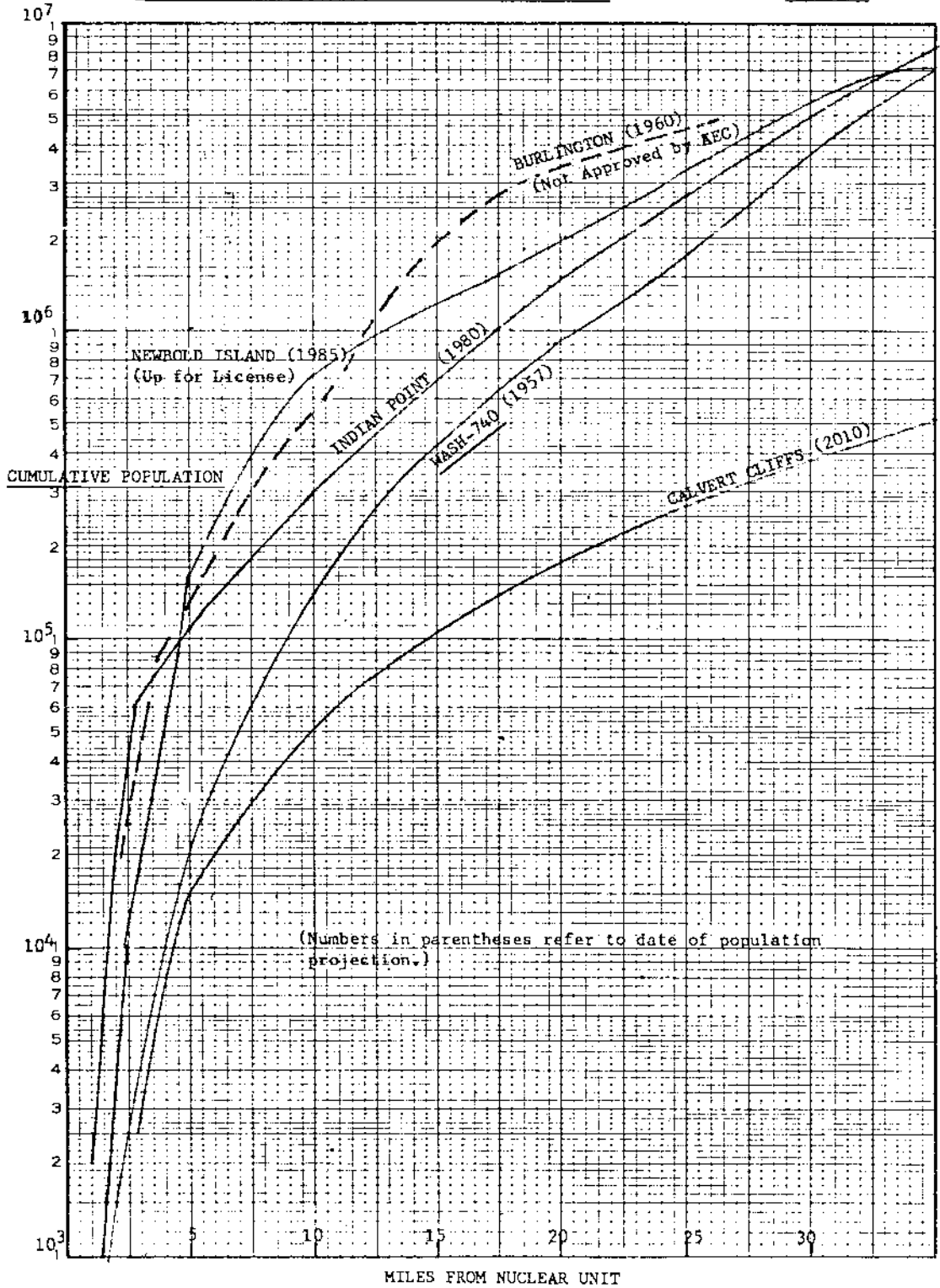
U.S. AIR FATALITIES (1927-1972)

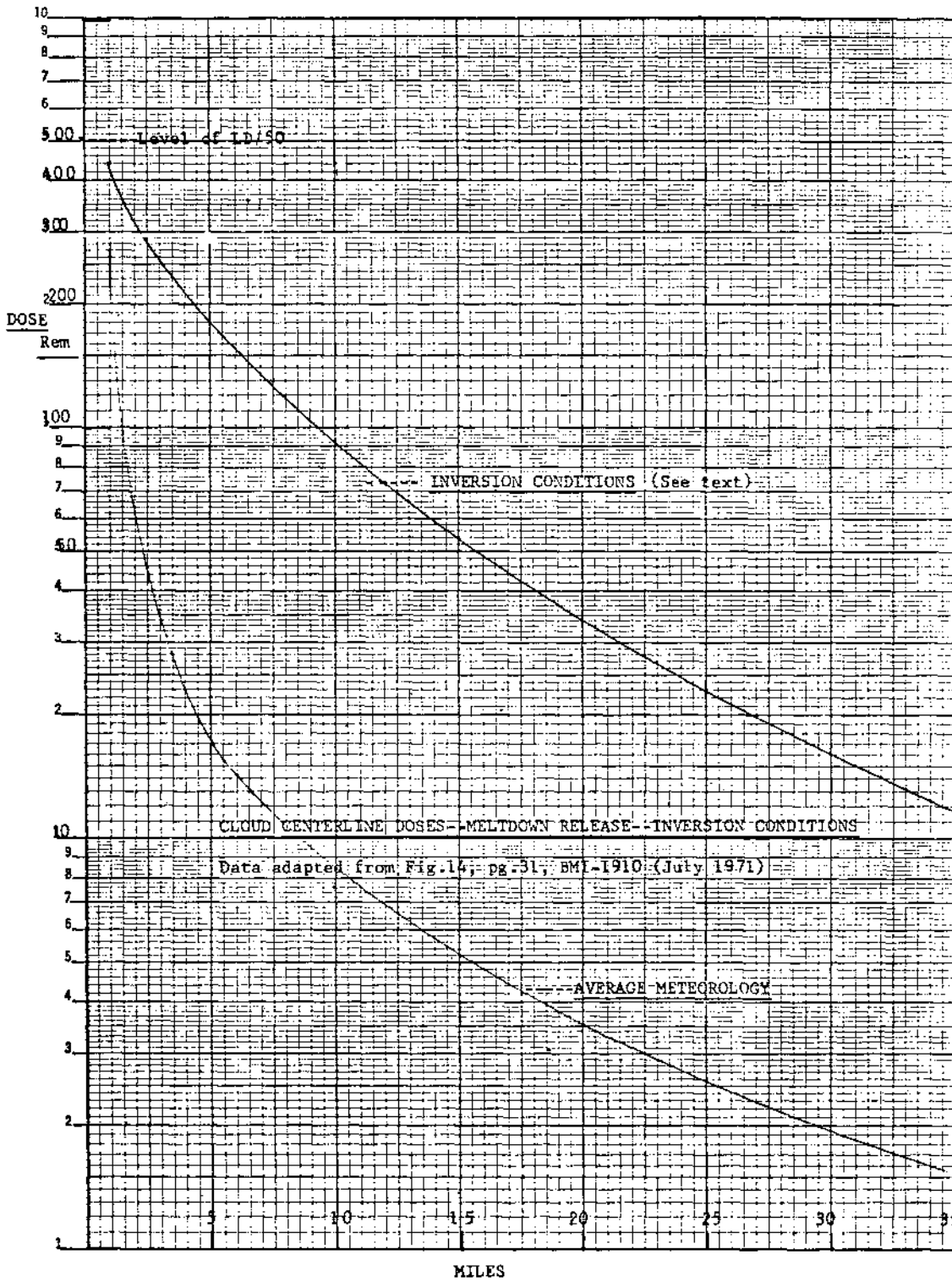
CHART I



POPULATION AT RISK NEAR SELECTED NUCLEAR SITES

CHART II







## RADIATION AND MAN

Presentation of U.S.A. National Academy of Sciences  
Report on The Effects on Populations of Exposure to Low Levels of  
Ionizing Radiation (BEIR Report)

### 1. General Review and Implications

C. L. Comar, Cornell University, Ithaca, N.Y.

#### Abstract

This paper together with that of Dr. James F. Crow on Genetic Effects and of Dr. Arthur C. Upton on Somatic Effects is an expository review of the report of the Advisory Committee on the Biological Effects of Ionizing Radiations of the National Academy of Sciences - National Research Council which was prepared by about 50 members of the Committee and its Subcommittees and completed in late 1972. It deals with the scientific basis for establishment of radiation protection standards encompassing a review and re-evaluation of existing scientific knowledge concerning radiation exposure of human populations.

Data, assumptions, and numerical risk estimates are presented for the genetic and somatic effects of low levels of exposure. It is calculated that exposure of the entire U.S. population to 170 mrem/year would cause at equilibrium (after many generations) between 1100 and 27,000 incidences per year of serious genetic disabilities. In the opinion of the Committee, the same exposure could cause from roughly 3000 to 15,000 cancer deaths annually based on the assumptions used including linearity and with no correction for dose-rate. The uncertainties and bases for these estimates are stressed in the papers of Drs. Crow and Upton. Interpretations are presented in regard to effects on growth and development and on the environment (on organisms other than man).

Future major contributions to radiation exposure of the population will continue to be natural background and medical applications. Medical radiation exposure can and should be reduced considerably without any deprivation of benefits. Radiation exposure to the U.S. population from the developing nuclear power industry can be kept at about 1% of natural background and the exposure of any individual kept to a small fraction of background provided that the technology operates as planned. Engineering and technical uncertainties in regard to nuclear power were not assessed by the Committee.

It is suggested that radiation protection standards not be set on an arbitrary basis such as related to background levels, (even though all agree that such levels will not produce observable effects), but rather should be established in terms of minimal exposures required to fill society's needs. Hopefully, it will be possible to make meaningful risk-benefit assessments, then to make cost-effectiveness assessments so that logical decisions can be made as to the worth of any given effort to reduce the risk, and finally to choose among the alternate options taking into account a comparison of the biological and environmental costs.

Ultimately, these techniques for dealing with radiation protection (actually estimating the risks and the worth of reducing them) may provide guidance for other pollutants, since the time is coming when priority decisions will have to be made in allocation of limited resources for the maintenance and improvement of the quality of life.

## I. Introduction

In late 1972 the Advisory Committee on the Biological Effects of Ionizing Radiation of the U.S.A. National Academy of Sciences - National Research Council completed its Report on The Effects on Populations of Exposure to Low Levels of Ionizing Radiation (BEIR Report). This paper, the first of three, is a personal interpretation of the approach, general findings and implications of the BEIR Report. It will be followed by detailed discussions of Genetic Effects by Dr. James F. Crow and of Somatic Effects by Dr. Arthur C. Upton. Appreciation is expressed to the Program Committee of the International Radiation Protection Association for the opportunity of presenting these papers and the generous time allotment.

## II. Background

Understanding of the objectives and frame of reference of the BEIR Report can be gained by remembering the "BEAR" Reports (on Biological Effects of Atomic Radiation) of an earlier committee of the National Academy that was established in 1955 to respond to public anxiety about the possible effects of nuclear weapons testing on the human population. The BEAR reports did so respond, were accepted by the public, and served mainly to: (a) place in perspective the extent of harm expected from fallout, (b) introduce the concept of regulation of average population doses on the basis of genetic risks, and (c) emphasize the significance of medical-dental radiation exposure.

In the late 1960's public concern again became aroused because of potential exposures from a developing nuclear power industry. Numerical genetic and somatic risk estimates were being used, and harmful effects were widely described as if it were planned that the total U.S. population were to be exposed to levels equivalent to present radiation protection guideline (namely 170 mrem/year). In February of 1970 the Federal Radiation Council (FRC) asked an existing NAS-NRC Advisory Committee to undertake a complete review of the matter. Thereafter the FRC was subsumed into the Environmental Protection Agency, the Advisory Committee was expanded into the BEIR Committee, and the review was undertaken.

Over the years, somewhat similar responsibilities in regard to radiation protection have fulfilled by other organizations including; the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), The International Commission on Radiological Protection (ICRP), and the National Council on Radiation Protection and Measurements (NCRP). Their reports have been most useful to members of the BEIR Committee and their personnel most cooperative.

It must be remembered that reports of various bodies may differ because of the charge, scope and composition of the committee involved, as well as the state of knowledge and public atmosphere existing at the time of the writing of a given report. In general the main differences between the BEIR Report and previous official documentation arise not so much from the inclusion of new data or new interpretations, but rather from a philosophic approach to radiation protection generated by changing conditions and public attitudes.

The BEIR report differs from the other reports in one or more of the following ways: (a) efforts are made to present the conclusions in such a way as to be useful to those who must take into account technological, economic, and sociological considerations in the development of regulatory programs in

the United States; (b) numerical risk estimates for human populations exposed to low levels of ionizing radiation are presented together with the assumptions and compilations of data on which they are based; (c) it is suggested that radiation protection standards should not be set on an arbitrary basis, such as in relation to background levels (even though all investigators agree that such levels will not produce observable effects), but rather that they should be established in terms of minimal exposures required to fill society's needs; (d) implications of possible effects of radiation on the environment - on organisms other than man - are considered; (e) it is proposed that medical-dental radiation exposure can and should be reduced considerably without impairing medical benefits; (f) emphasis is placed on the desirability of making meaningful risk-benefit assessments and then cost-effectiveness assessments so that logical decisions can be made as to the worth of any given effort to reduce the risk - choices could then be possible among alternate options involving radiation and non-radiation processes by comparison of biological and environmental costs.

Obviously the BEIR Committee and its subcommittees even though expanded to a membership of about fifty could not deal definitively with the broad non-scientific, sociological or economic issues. Yet it was felt that such issues must be brought forward. For example, in regard to the possibilities of catastrophic reactor accidents; it was not within our competence to render opinions on probabilities, effects, overall risks, financial expenditures justified, or rational of public acceptance. Nevertheless, we did call attention to this matter prominently in the summary statement.

### III. Risk Estimates

There is always the question as to whether numerical risk estimates should be included in official documentation. It is realized that no matter how carefully the estimates are qualified, the numbers will be used and quoted by others out of context. The decision to include numerical values was based largely on the fact that such estimates were already being widely used in public discussion and were having a significant emotional impact on decision-making. It was felt necessary first to assure that the most reliable numbers be used, but perhaps more important, to bring about the understanding that it may be a public disservice to misuse risk estimates, that is to make decisions on the basis of risk estimates for only one of the necessary options, or to spend large amounts of resources to reduce small risks even further while larger risks go unattended.

The philosophy in radiation protection to-date has been to state the matter in qualitative terms. For example, NCRP 39\* contains the following statement:

"In particular, it is believed that while exposures of workers and the general population should be kept to the lowest practicable level at all times, the presently permitted exposures (170 mrem/yr for the population) represent a level of risk so small compared with other hazards of life, and so well offset by perceptible benefits, that such approbation (public acceptance) will be achieved when the informed public review process is completed."

\*Basis Radiation Protection Criteria, National Council on Radiation Protection and Measurements, January 15, 1971, NCRP Report No. 39. Material in brackets added.

However, it appears now that public knowledge and public demand requires further detail.

The risk estimates of the BEIR Report are presented in greatly simplified form in Tables 1 and 2. In round figures it is stated that exposure of a population of 200 million to 170 mrem/yr could cause about 10,000 deaths or serious disabilities per year of somatic and genetic origin in the first generation. This can be expressed as a risk per year of 1 in 20,000. Given the numerical estimates for genetic and somatic risk the question arises as to how this information can be used as a basis for radiation protection guidance. Logically, the guidance or standards should be related to risk. Whether we regard a risk as acceptable or not depends on how avoidable it is. Obviously no risk, no matter how small, should be accepted if it is indeed readily avoidable. To the extent not avoidable, acceptability depends upon how the risk compares with those of alternative options and those normally accepted by society.

It is not difficult to compare the risks from radiation with those normally accepted by society - recognizing that any such comparison is not meant as a justification for acceptance of any unnecessary risk. As indicated in Table 3, there seem to be two natural boundary conditions. The high risk boundary of 1 in 100 is the statistical risk of deaths from all causes. The lower boundary of 1 in 1,000,000 appears to be the risk of deaths from external catastrophes such as floods, earthquakes, lightning, snake bites, etc. Thus, one can establish a rough scale of risk, as shown in Table 4.

Traditionally, society has treated low and negligible risks as acts of God and has focused attention on the high risk category. It is interesting to note that in the high risk range we find such voluntary activities as auto travel, plane travel, hunting, and skiing.

It is important to realize that public response will probably never be completely logical. For example, the public abhorrence of specific catastrophes may result in relatively large investments to avoid them, regardless of the quantitative importance. There may be other philosophic biases related to risk that are just not subject to rational agreement.

There are two main aspects of the analysis. First, it is necessary to assess risk-benefit relationships since a man-made risk can be regarded as avoidable unless we need the associated benefit. As indicated, we feel that the risk estimates for radiation are comparatively well established, but the question of benefit requires much more consideration. The BEIR Committee has now been specifically charged to undertake the study of benefits, the difficulty of which should not be underestimated. As an example, one benefit of radiation exposure from the nuclear power industry is, obviously, the avoidance of health effects from the combustion of fossil fuels, which immediately necessitates comparing the biological costs of the alternative options.

The second aspect concerns what might be called "cost-effectiveness analysis," which governs just how avoidable the risk is. It is obvious that any risk can be decreased at an increased financial cost. In a resource limited society (and believe me, we are becoming more so all the time) the allocations must be made where they will do the most good. It is a misuse of resources and a disservice to society to add costs for the purpose of decreasing the risks of any one system greatly below acceptable levels, when other societal activities with unacceptable risks are being ignored. Some examples of choices that could be made are: a national program to persuade people to use seat belts is estimated to cost less than \$100 for each death

averted; a program of early cancer detection and treatment is estimated to cost up to about \$40,000 for each death averted. At the height of fallout it was calculated that the removal of  $^{90}\text{Sr}$  from milk at a cost of 2 to 3 cents per quart would cost about 20 million dollars for each case of cancer averted. It has been estimated that money spent on improved collimation of X-ray machines would be 1000 to 10,000 times more effective in reducing radiation dosages than money spent on improving present reactor waste systems.

By way of summarizing risk considerations, certain points appear to be important. Our best estimates of risk from radiation exposure, even when expressed in numerical terms support previous views (implied by NCRP documentation) that such risks are indeed small when compared with the probable benefits from necessary activities. The public should somehow be educated to this truth and all attempts should be made to reduce any philosophic biases. As part of this process, regulatory procedures should assure that societies' needs are being met with the lowest possible risks. The interpretation of the words, "lowest possible," should rest upon development of cost-effectiveness analyses. It should be made clear that even when the benefits clearly exceed the risks, the risks should still be reduced to the lowest level compatible with cost-effectiveness considerations.

#### IV. Summary of BEIR Report Recommendations

The BEIR Report made ten major points which are paraphrased in abbreviated form:

- a. No exposure to ionizing radiation should be permitted without the expectation of a commensurate benefit.
- b. Public protection from radiation must not result in a substitution of worse hazards for the radiation avoided. Small risks should not be reduced below the point of cost-effectiveness.
- c. The upper limit of non-medical radiation exposure for individuals in the general population should produce risks that are small relative to those normally accepted.
- d. The above upper limit for individuals should be considerably lowered if the general population is to be exposed. This limits the total amount of harm that could be caused as well as the probability of harm to any individual.
- e. Medical radiation exposures can and should be reduced considerably without impairing medical benefits.
- f. Guidance for the nuclear power industry should be based on risk-benefit and cost-effectiveness analysis taking into account the alternate options.
- g. Extraordinary efforts to minimize the risk from catastrophic accidents in the nuclear power industry are called for.
- h. Occupational and emergency exposure limits should be based on numerical risk estimates to the individual.
- i. Populations of other living organs will most likely not be perceptibly harmed if the radiation protection guidelines acceptable for man are adhered to. Nevertheless, there are good reasons for

strengthening ecological studies.

- j. Every effort should be made to assure accurate estimates and predictions of radiation equivalent dosages from all existing and planned sources.

#### V. A Broader View

Society is becoming increasingly sensitive to health problems from contamination and pollution. Whenever a potential hazard is identified, becomes measurable, and is publicly recognized there is pressure for zero or near zero guidelines. Usually there is little or no information about low-level population effects and decisions may well be made that overall cause a lowering of the quality of life. Radiation is the one agent for which tremendous amounts of data are available. Now that we have risk estimates, fraught with uncertainty though they may be, the way we use, neglect, or misuse them for the public good may set an example for the course we are to follow with the great variety of potential harmful agents that modern man exposes himself to. In a real sense then, radiation workers have societal responsibilities that go far beyond the effects of ionizing radiation.

Table 1

Simplified Summary of Radiation Effects

170 mrem/yr to U.S. population ( $200 \times 10^6$ )

would cause an upper limit of 3600 genetic disabilities/year in 1<sup>st</sup> generation

could cause as a most likely estimate 6000 cancer deaths/year

Table 2

Radiation Effects Expressed as Risk per year

170 mrem/yr could cause

~ 10,000 cases of harm in  $200 \times 10^6$  per year  
or 1 case in 20,000 per year

Table 3

Comparative Population Risks in the U.S.

<u>Sources</u>	<u>Risk of Death per Year</u>
From all causes	
Under 1 year	1 in 46
1 year old	1 in 735
10 years old	1 in 3600
35 years old	1 in 470
55 years old	1 in 85
Average population	1 in 100
From 170 mrem/year	1 in 20,000
From natural disasters	1 in 1,000,000
From 1 mrem/year	1 in 3,400,000



Table 4

Arbitrary Categories of Risks

<u>Scale</u>	<u>Risk of Death per Year</u>
High	1 in 100 to 1 in 1,000
Medium	1 in 1,000 to 1 in 100,000
Negligible	1 in 100,000 to 1 in 10,000,000

PRESENTATION OF THE U. S. A. NATIONAL ACADEMY OF SCIENCES REPORT ON THE  
EFFECTS OF IONIZING RADIATION (BEIR REPORT). 2. GENETIC EFFECTS

James F. Crow  
Genetics Laboratory, University of Wisconsin  
Madison, Wisconsin 53706

Abstract

Information for assessing the magnitude of the human genetic risk from radiation still comes almost entirely from nonhuman sources, mainly the mouse. The Committee recommended that estimates for low dosages be made on the assumption of a linear relationship between the effects at the lowest doses where reliable measurements exist and the effect at zero dose. This was chosen as a plausible assumption for some effects (mutation and chromosome breakage) and as a conservative procedure for others (nondisjunction and chromosome loss).

The Committee considered four bases for risk estimation. In order of reliability these are: (1) The risk relative to natural background radiation, (2) The risk relative to specific genetic conditions, (3) The risk relative to current incidence of serious disabilities, and (4) The risk in terms of overall ill health.

The Original BEAR Report

The current report is a follow-up of the original report of the National Academy of Sciences Committee on the Biological Effects of Atomic Radiations (BEAR).<sup>1</sup> This Committee, along with a corresponding group in Britain working at the same time and not entirely independently, introduced the idea of regulating the *average* dose to the population.

The BEAR Committee recommended that man-made radiation be kept at such a level that the *average* individual in the population receive less than 10 r before the mean age of reproduction, a period of time taken to be 30 years. Moderate variation in exposure from person to person was not regarded as very important as long as the population average is kept low for the reason that the damage is to later generations. The concern is not so much that of the individual for his own children, for which the risk is slight, but of society for an overall disease and disability rate in future generations. Prior to this report, the main emphasis had been on the protection of the person receiving the radiation. The individual dose limit was set so as to be well below that for which there is any observable harm. The BEAR Genetics Committee emphasized the linear, non-threshold concept for genetic effects and its implication that there is no "safe" dose, a concept that had been discussed earlier by the NCRP.

The general principles guiding the Committee were: (1) Mutations, spontaneous or induced, are much more often harmful than beneficial. This is to be expected on the grounds that mutations, being random events, are more likely to make well regulated systems worse rather than improving them. It has also been

observed experimentally that mutations whose effects are large enough to be visible are almost always harmful. (2) Any amount of radiation, however small, that reaches the reproductive cells entails some genetic risk. (3) The number of mutations produced is directly proportional to the dose, so that linear interpolation from high dose data provides a valid estimate for low dose effects. (4) The effect is independent of the rate at which the dose is administered or of the spacing of the total amount. The last of these has turned out to be wrong, as will be discussed later.

With these principles, the number of mutations is the simple product of the number of genes in the population *times* the dose *times* the mutation rate per gene per unit dose. For the last quantity, mouse data were becoming available at the time of the BEAR study, and the effects were considerably higher than those in *Drosophila*, which had constituted the main quantitative evidence before this time. But there was no estimate of the number of genes in any mammal. There wasn't any very good evidence in *Drosophila* either. Some *Drosophilists* suggested that the bands on the salivary gland chromosomes might correspond to genes and that counting them might give an estimate of the number of genes; but this generally was regarded as naive. A more indirect way was to estimate the ratio of the total mutation rate to the specific locus rate, but this had its problems because of the difficulty in measuring the total mutation rate. This ratio was taken to be about 10,000 and the risk estimates therefore were for a hypothetical organism whose mutation rate is that of the mouse and whose gene number is that of *Drosophila*.

H. J. Muller strongly advocated the principle that each mutant must ultimately be eliminated from the population, and therefore for each mutation there must eventually be one gene extinction, or "genetic death". The Committee included this kind of calculation in its report, but with mixed enthusiasm. Some thought it to be the only way of trying to assess the *total* impact of mutation. Others thought the problem of finding the correspondence between gene extinctions and tangible measures of human suffering and frustration to be completely insoluble, and therefore the method essentially worthless. The Committee also estimated the mutation rate doubling dose and applied this to the estimated mutational component of human disease and disability.

#### What Has Been Learned Since?

What has been learned in the nearly two decades since the BEAR Committee met? An enormous amount by any standards! The BEAR Report was written early enough to miss both the molecular revolution and the development of human cytogenetics. I don't believe the letters "DNA" appear anywhere in the Genetics Report, and the chromosome number is given as 48.

We now know the chemical basis of the gene with an amount of detail that would have been utterly unbelievable in the 1950's. The chemical basis of mutation is deeply understood and the systems of mutation repair, especially of UV damage, are models of clarity and beauty. The human species has joined *Tradescantia*, maize, and *Drosophila* in becoming cytologically respectable.

With such deep fundamental knowledge one might expect that estimation of radiation risks would be correspondingly more precise. Yet, there remain large gaps, the most serious being (1) the almost complete absence of information on radiation mutagenesis from human sources, and (2) our inability to determine the relationship between an increase in the mutation rate and the effect on human welfare in the future. In some ways the situation seems worse than it did in the 1950's because researches in the meantime have brought out complexities that were not suspected at that time. It can no longer be assumed that the number of mutations is independent of the dose rate or of fractionation.

Furthermore, we are more cognizant of differences in different kinds of cells, between the sexes, and among different organisms.

#### How Valid Are Mouse Data?

Since we still don't have any reliable human radiation data we still have to rely on other organisms, particularly the mouse. The BEIR Report<sup>2</sup> and the United Nations Report (UNSCEAR)<sup>3</sup> do this. Is there any reason to believe that mouse rates are equivalent to man? I should like to present some data, recently assembled by Abrahamson, Bender, Conger, and Wolff<sup>4</sup>, that should add to our confidence in extrapolation from other organisms to man. They plotted the mutation rate per rad per locus as a function of the amount of DNA in the haploid genome. The results for *Escherichia*, yeast, *Neurospora*, *Drosophila*, mouse, tomato and barley, on a log-log plot, fall very close to a straight line at a 45 degree angle to the axes. The amounts of DNA in these species ranges over a factor of 1000; so do the mutation rates, ranging from  $10^{-9}$ /rad for *E. coli* to  $10^{-6}$ /rad for barley. Yet, the ratio is nearly constant. The human species has about 20 percent more DNA per cell than the mouse, so placing ourselves at the appropriate place on the line gives a single-locus mutation rate of  $2.6 \times 10^{-7}$ /rad. This is the value for high dose-rates; chronic radiation would produce effects 1/3 to 1/4 as high.

Is there any way to make sense out of this remarkable observation? There is perhaps one way. We must remember that what is constant when normalized for the amount of DNA is the *per locus* rate, not the genome rate.

It is known that in bacteria the genome is a continuous string of DNA and that there are roughly 3000 genes. There is now good evidence from *Drosophila* that the number of gene loci is equal to the number of salivary gland chromosome bands -- just as the more naive geneticists used to think. The evidence comes from the work of Judd and his colleagues<sup>5</sup> who for several years collected all mutants that were located in a small region of the X chromosome that could be delimited by a deletion. They now seem to have found all the gene loci in this area, since for some time all the new mutants have been mapped at one of the previous sites. These lethal, or in a few cases, visible, mutants fall into 16 distinct groups, as defined by a complementation test. It is also true that there are exactly 16 salivary chromosome bands in this region. Unless this is a fantastic numerical coincidence, the simple idea that the number of genes is equal to the number of chromomeres appears to be correct. There is supporting evidence from other *Drosophila* chromosome regions.

The number of salivary gland chromosome bands is a little over 5000. Thus, the *Drosophila* has only about twice as many genes as *E. coli*. Yet, the amount of DNA per cell is an order of magnitude higher. There is also good evidence that the *Drosophila* chromosome is a continuous strand of DNA -- some 20 Angstroms in diameter and about a centimeter long! Thus, it looks as if a *Drosophila* gene is at least ten times as long as a bacterial gene.

If we accept this inference, then as organisms get larger and more complex, they don't get many more genes, but rather, the genes get longer. If this is true, the gene in higher organisms presents a larger target for radiation. Perhaps this is the explanation of the puzzling results of Abrahamson *et al.*

In any case, whether the explanation is correct or not, the fact that the data from these diverse organisms lies so close to the line adds to our confidence in extrapolating to man from the mouse.

#### Risk Estimates

All quantitative estimates that the BEIR Genetics Committee used were derived from mouse low dose-rate data. No correction was made for the larger amount of DNA in the human cell, although this would have made only a trivial difference (about 20 percent) among much larger uncertainties. Cytogenetic estimates were usually made directly as if humans were mice, although adjustments were made in those cases where there was some reason to think humans are different.

Estimates of genetic disease other than cytogenetic was done by estimating the relative risk for one rem. This is the proportion by which the mutation rate is increased by one rem; its reciprocal is the doubling dose. This was estimated by taking the specific locus rate for mice, averaged over the two sexes, as the radiation induced rate. The spontaneous rate was estimated directly from human spontaneous mutation rate studies. From this we derive 1/200 to 1/20 as the relative risk of one rem -- or 20 to 200 as the doubling dose.

For any category of disease the Committee attempted to estimate the *mutational component* of its incidence. Conceptually, we think of the disease incidence as divided into two discrete compartments, one of which has an incidence directly proportional to the mutation rate and the other whose incidence is independent of the mutation rate. (Nobody thinks that this is a correct picture of the true situation, but it seemed to us to be a reasonable model for the purpose of risk assessment.) For conditions that are caused by dominant or X-linked mutations, the mutational component as defined above is very nearly one. For congenital anomalies and constitutional diseases the fraction is taken to be from 5 to 50 percent.

The Committee recommended four bases for risk estimates: (1) The risk relative to the natural background radiation; (2) The risk for specific genetic conditions; (3) The risk for severe malformation and disease; and, (4) The risk in terms of overall ill health.

These are in decreasing order of robustness and accuracy and increasing order of social relevance. Unfortunately, the closer we come to estimating tangible human dangers, the more uncertain the estimates become.

1. The Risk Relative to That From Natural Background Radiation.

Of course, this is not a risk estimate at all; but it may be very useful as a policy guide, nonetheless. The idea is this: The human species has lived with this amount of radiation throughout its evolutionary history. Although we don't think this has been good for us, nevertheless we have managed to survive, even thrive. Most people don't take background radiation levels into account when they decide where to work or live; in other words, the risk is comparable to other risks that are commonly, usually unthinkingly, accepted. As the Report says: "If the genetically significant exposure is kept well below this amount, we are assured that the additional consequences will neither differ in kind from those which we have experienced throughout human history nor exceed them in quantity."

2. The Risk For Specific Genetic Conditions.

The basis for this estimate is the radiation-induced rate for mice, averaged over both sexes. For chronic radiation of spermatogonia in males and oocytes in females, the average is taken to be  $.25 \times 10^{-7}$  per rad. Using the incidence of dominant and X-linked diseases and making informed guesses as to the persistence of these genes in the population, the human risks were estimated.

3. The Risk Relative to the Current Incidence of Serious Disabilities.

With a 20-200 rem doubling dose, an exposure of 5 rem per generation (170

mrem per year) would cause an eventual increase of from 2.5 to 25 percent in the burden of disease that owes its incidence to mutation. About one percent of children have a dominant or X-linked disease or disability, and this incidence is essentially proportional to the mutation rate. Recessive diseases are rarer and their incidence is only very indirectly related to the mutation rate. Disease of more complex etiology -- congenital anomalies, anomalies expressed later in life, constitutional and degenerative diseases -- are partly genetic, but there is great uncertainty as to how directly their incidence reflects the mutation rate. It is unlikely that more than half the incidence has this cause. Some would estimate it as low as 5 percent. The estimates are summarized in Table 1, which is taken from the BEIR Report.

Table 1. Estimated effect of 5 rem per generation on a population of one million. This includes conditions for which there is some evidence of a genetic component.

Disease classification	Current incidence	Effect of 5 rem per generation	
		First generation	Equilibrium
Dominant diseases	10,000	50-500	250-2500
Chromosomal and recessive diseases	10,000	Relatively slight	Very slow increase
Congenital anomalies	15,000	}	50-5000
Anomalies expressed later	10,000		
Constitutional and degenerative diseases	15,000		
TOTAL	60,000	60-1000	300-7500

4. The Risk in Terms of Ill Health.

In addition to the categories above, we have illnesses of many sorts, ranging from so mild as to constitute only a minor inconvenience to severely incapacitating and fatal. The mutational component can only be guessed. Dominant genes probably play a smaller part in this than they do in the conditions in Table 1. Rather arbitrarily we took 20 percent as the mutational component. This leads to an estimate, at equilibrium, of an increase in all disease of between .5 percent and 5 percent if the population were exposed to 5 rem per generation. The Committee also suggested how a dollar value might be placed on a rem through this estimate.

One factor that is left out of these calculations, and which we have no way of assessing, is what appears to be the majority of mutants in *Drosophila* -- namely, mutants with a very mild effect on viability. These mutants show very little recessiveness, so their impact is partly in the first generation after the mutation occurs and is spread over the next 50 to 100 generations. Extensive mouse experiments offer no evidence for any measurable contribution from such mutants. The Committee had this admonition: "We remind all who may use our estimates as a basis for policy decisions that these estimates are an attempt to take into account only known tangible effects of radiation, and that there may well be intangible effects in addition whose cumulative impact may be appreciable, although not novel."

As regards public policy toward radiation protection, the Committee had this to say: "It seems clear that the genetically significant radiation exposure from fallout, from nuclear power developments, and from occupational exposure (treated as a part of the over-all population average) is now very small relative to that from natural radiation. There is no reason to think that the dose commitment for the development of nuclear power in the next few

decades should be more than about a millirem annually. The 1956 report and the guides that grew out of it were the result of an effort to balance genetic risks against the needs of society. It now appears that these needs can be met with very much less than the 170 mrem per year of the current Radiation Protection Guides. Accordingly, the 170 mrem seems to provide an unnecessarily large cushion.

Likewise, we believe that the currently much higher level of radiation from medical sources (mainly diagnostic) should be examined in view of the same concept. If it can be reduced further without impairing essential medical services, then the present level is unnecessarily high."

#### References

1. The Biological Effects of Atomic Radiation. Summary Reports. National Academy of Sciences--National Research Council. 1956.
2. The Effects on Populations of Exposure to Low Levels of Ionizing Radiation. Report of the Advisory Committee on the Biological Effects of Ionizing Radiations. National Academy of Sciences--National Research Council. 1972.
3. Ionizing Radiations: Levels and Effects. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation. United Nations. 1972.
4. Abrahamson, S., M. A. Bender, A. D. Conger, and S. Wolff. The uniformity of radiation-induced mutation rates. Submitted for publication, 1973.
5. Shannon, M. F., T. C. Kaufman, W. W. Shen, and B. H. Judd. Lethality patterns and morphology of selected lethal and semi-lethal mutations in the zeste-white region of *Drosophila melanogaster*. Genetics 72: 615-838, 1972.

PRESENTATION OF U.S.A. NATIONAL ACADEMY OF SCIENCES REPORT  
ON THE EFFECTS ON POPULATIONS OF EXPOSURE TO  
LOW LEVELS OF IONIZING RADIATION.

3. SOMATIC EFFECTS

A.C. Upton

State University of New York at Stony Brook  
Stony Brook, New York

Introduction

Consideration is given herein to effects of ionizing radiation that are manifest in exposed individuals themselves (i.e., somatic effects) as contrasted to effects that are manifest in subsequent generations (i.e., genetic, or inherited, effects). In general, moreover, acute effects of irradiation are not considered, since these occur only at dose levels well above protection standards.

With few exceptions, the somatic effects of interest manifest themselves only after an interval of years or decades following irradiation and are not detectable except in a statistical sense. In any given individual, a particular effect cannot be attributed conclusively to radiation, as opposed to some other cause, and the smaller the dose, the less the likelihood of radiation being the cause.

Because no somatic effects causing significant disease or mortality are known to be induced by ionizing radiation at dose rates approaching natural background, the risks of such effects at these dose rates can be estimated only by extrapolation from observations at higher radiation levels, based on assumptions about the relevant dose-effect relationships, the mechanisms through which the effects are produced, and the susceptibility of the populations at risk.

Principles Underlying Induction of Somatic Effects

For none of the effects of interest can the dose-response relation be defined over a wide range of dose and dose rate. For some effects, however, such as the induction of cataract of the lens and impairment of fertility, the relationship between effect and dose is nonlinear, these effects presumably depending on the killing of sufficient numbers of cells in the lens and gonads, respectively, so that there is little or no risk of the effects at dose rates approaching natural background radiation levels.

For induction of certain tumors, on the other hand, a linear non-threshold dose-effect relationship cannot be excluded, nor can the possibility that such effects might result from subtle injury in only one or a few cells of the body. The most important effect of radiation on the mortality of human populations, furthermore, apparently results from carcinogenic effects.

In assessing the induction of cancer, the following problems are noteworthy: (1) cancers induced by radiation are indistinguishable individually from those occurring naturally, their existence being demonstrable only in terms of an excess above the natural incidence; (2) the natural incidence of cancer varies by orders of magnitude, depending on the type of neoplasm, age and sex of population at risk, and other factors; (3) cancer of any one type occurs with sufficiently low incidence in man that few irradiated populations are large enough to provide relevant quantitative dose-incidence data; (4) the time elapsing between irradiation and the clinical appearance of a neoplasm is a matter of years or even decades, complicating the prospective follow-up of irradiated populations for tumor development and the retrospective evaluation of cancer patients for relevant radiation exposure history; (5) many of the data on radiation-induced tumors come from individuals exposed to internally deposited radionuclides, in whom the dose-incidence relation is obscured by



nonuniformities in temporal and spatial distribution of the dose; (6) other data come from studies of therapeutically irradiated patients, in whom effects of radiation may be confounded by effects of underlying disease processes or of treatments other than radiation; and (7) some of the available data concern cancer mortality, whereas others concern cancer incidence, hence radiation-induced malignancies that do not greatly alter the death rate (e.g., thyroid carcinoma) must be distinguished from those that are more generally fatal (e.g., leukemia).

#### Cancer Incidence and Radiation Dose

Despite the difficulties mentioned above, the incidence of several types of cancer in human populations has been shown unequivocally to increase with increasing dose. With few exceptions, however, the observed dose-incidence data pertain to relatively high doses and high dose rates. Nevertheless, the findings for any given neoplasm are reasonably consistent from one irradiated human population to another, suggesting that the observed relationship may be applicable within limits to the general population for purposes of risk evaluation.

In Japanese atomic-bomb survivors and in British patients treated with spinal irradiation for ankylosing spondylitis, the incidence of all leukemias except the chronic lymphocytic type has been increased, the relationship between incidence and dose at the relatively high doses and high dose rates in question, being compatible with a linear dose-incidence function with a slope corresponding to about 1 case of leukemia per  $10^6$  exposed persons, per year, per rem. Data for other irradiated populations, although far less quantitative, imply a comparable excess of leukemia per unit dose to the marrow, despite wide differences in the conditions of exposure; however, there is evidence that susceptibility may be several times higher in utero, during childhood, or late in adult life than at intermediate ages.

Tumors of the thyroid gland also have been found to be increased in incidence in irradiated populations. The dose-effect relationship at relatively high doses and high dose rates, like that for leukemia, can be represented by a linear, non-threshold function, corresponding to a risk of roughly 2-9 cases of cancer per  $10^6$  exposed children, per year, per rem to the thyroid gland, averaged over the fifth to twenty-fifth years after exposure. In those irradiated during childhood, susceptibility appears several times higher than in those irradiated as adults.

For tumors of other types and sites, dose-response data are more limited, and estimates of risk correspondingly cruder. For cancer of the lung, mortality at high doses has been estimated to approximate one death per  $10^6$  exposed persons per year, per rem. For cancer of the breast, mortality at high doses has been estimated to approximate two deaths per  $10^7$  exposed persons per year, per rem. For cancer of the GI tract, including the stomach, mortality at high doses has been estimated to approximate one death per  $10^6$  persons per year, per rem. For cancer at all other sites combined, mortality has been estimated to approximate one death per  $10^6$  persons per year, per rem, which implies that either susceptibility to such malignancies is low, by comparison with susceptibility to the types mentioned earlier, or that the latent periods for such malignancies extend well beyond 25 years of follow-up.

Some studies suggest that after prenatal irradiation the overall juvenile cancer mortality may be increased by about 50 cases/10<sup>6</sup>/rem/year, averaged over the first 10 years of life; however, there is also evidence that the observed excess may be dependent on factors other than radiation.

The variations in rate of induction of different types of cancer by irradiation are apparently unrelated to variations in the natural incidence of the respective types. Hence it is clear that the doubling dose of radiation is not uniform for all types of cancer.

#### Probability of Cancer Induction at Low Doses and Low Dose Rates

The dose-mortality figures cited above, which pertain chiefly to populations exposed at high doses and high dose rates, may be used to estimate the probability of cancer at lower doses and lower dose rates, if it is assumed that the relationship between mortality and dose remains the same irrespective of changes in dose, dose rate, and population at risk. However, there are cogent radiobiological reasons for doubting that the dose-incidence relationship remains constant in the face of such changes. One reason is the widespread occurrence of repair of most types of injury induced at low doses and low dose rates by low-LET radiations. The dose rate characteristic of background radiation (approximately 0.1 rem/year) is 10<sup>8</sup>-10<sup>9</sup> times lower than the dose rate at which effects have been observed in most irradiated populations, and at background levels ionizing events in individual mammalian cell nuclei occur at a frequency of less than one per day, whereas at the higher dose rates mentioned, thousands of such events occur every second. Because of this difference, and its implications for the production and repair of radiation damage at the molecular level, the risk of cancer induction at low doses and low dose rates may be appreciably smaller per unit dose than at high doses and high dose rates (as has been observed to be the case in certain radiation-induced tumors of experimental animals). The possibility of zero risk at low dose rates is not excluded by the data.

#### Relative Biological Effectiveness

Another source of uncertainty complicating extrapolation from available data is the variation in relative biological effectiveness among different types of radiations. This problem pertains to the interpretation of data from atomic bomb survivors of Hiroshima, underground miners exposed to radon gas and its radioactive decay products, and populations with high body burdens of alpha-emitting radionuclides.

In Hiroshima, the numbers of survivors are larger (and the statistics correspondingly better) than in Nagasaki; but the radiations at Hiroshima included an appreciable component of fast neutrons. Hence it is necessary to estimate the relative biological effectiveness (RBE) of this component in order that the dose-effect data for the two cities can be compared. The best estimate of the RBE, derived from intercomparison of the Hiroshima and Nagasaki data for leukemia, is between 1 and 5; however, for many radiobiological effects the risk-per-rad of low-LET radiations, such as x-rays and gamma rays, decreases to a greater degree with decrease in the dose and dose rate than does the effectiveness of high-LET radiations, which may decrease little if at all. Hence the RBE value of 1-5 for leukemia induction may be considerably smaller than the RBE value applicable to low doses and dose rates. Nevertheless, since RBE values of 1 and 5 have been assigned in this report to the Hiroshima neutrons for the purpose of calculating the risk per rem, the resulting estimates of risk may err on the conservative side.

#### The Linear Hypothesis

Although there is experimental evidence that the dose-effect relationship for x-rays and gamma rays may not be invariant with dose and dose rate, the use of a non-linear hypothesis in estimating risks for purposes of radiation

protection would be impractical in the present state of knowledge, since it would require allowance for individual variations in temporal and spatial distribution of tissue dose, as well as for other variables which cannot be analyzed at this time.

Furthermore, it is the whole population from birth to death that is to be protected, and no body of human observations provides risk estimates for longer than about 25 years. Moreover, the human fetus may be especially susceptible to radiation carcinogenesis. Thus, in a situation that calls for a careful weighing of costs and benefits it has seemed prudent to present risk estimates on the basis of human data exclusively, with the use of a linear interpolation into the region of low dose.

### Risk Estimation

In the Japanese atomic-bomb survivors, the excess mortality from all forms of cancer, including leukemia, corresponds to roughly 50-78 deaths per  $10^6$  exposed persons per rem over the 20-year period from 1950-1970; i.e., from the fifth to the twenty-fifth year after exposure. In the irradiated spondylitics, the excess mortality corresponds to a cumulative total of roughly 92-165 deaths from cancer per  $10^6$  persons per rem during the first 27 years after irradiation. If such rates, extrapolated to low-dose levels without allowance for the possible dependence of the effect on dose and dose rate, are assumed to apply generally, than exposure of the U.S. population of about 200 million persons to an additional 0.1 rem during one year (approximately equivalent to a doubling of irradiation from background sources) could be expected to cause 1350-3300 deaths from cancer during the 25 years following irradiation, or about 50 to 130 deaths per year. Continual exposure of the population to the additional 0.1 rem per year could be expected ultimately to cause 1350 to 3300 deaths annually, provided that the effect of a given increment of dose did not persist beyond 25 years after exposure. However, use of a factor to allow for the influence of dose and dose rate on the dose-effect relationship might reduce these estimates appreciably.

In assessing the cumulative effects of low-level irradiation on an entire population, attention must be paid to differences in age at exposure, duration of the latency for carcinogenesis, and size and duration of the carcinogenic effects; however, only tentative allowances can as yet be made for these variables. Nevertheless, a range of values can be assumed for each parameter (Table 1), enabling the effects of chronic low-level exposure of the U.S. population to be estimated, at least for illustrative purposes. These estimates (Table 2) imply that exposure of the entire population continuously throughout life at a dose rate of 0.1 rem per year could cause up to 1,700-9,000 cancer deaths per year, corresponding to 0.6-2.9% of the natural cancer death rate. For individuals exposed continuously from age 20 to age 65 years at a dose rate of 5 rems per year, the same approach yields an estimate of 380-930 excess cancer deaths per  $10^6$  persons per year (Table 3), corresponding to 1-2% of the natural cancer death rate at age 60-64 years.

Because the extrapolation model used in the above calculations made no allowance for the influence of repair at low doses and low dose rates, the derived estimates may be too high. For other reasons also, the estimates may be too high or too low: (1) insofar as high dose data have provided the primary basis for the estimates, the risks may have been overestimated, owing to side effects at the high dose levels which may have enhanced the carcinogenic action of radiation; (2) longer periods of follow-up may lead to estimates of risk that differ in magnitude from those above; (3) the data on most radiation-induced tumors are too scanty to allow construction of dose-incidence curves adequate for extrapolation; (4) uncertainty attaches to the RBE values used for alpha and neutron radiations; (5) uncertainty attaches to the relevant tissue dose, owing to nonuniformity in the distribution of the dose throughout

the body; and (6) the carcinogenic effects per unit dose might, under certain conditions, conceivably be even higher at low doses and low dose rates, owing to less killing of the cells that are most susceptible to cancer induction.

Comment

The figures presented in the foregoing are not to be taken as precise estimates of risk, since they are derived from evidence that is now incomplete. Moreover, the values are based largely on mortality data; and if expressed in terms of cancer incidence, the estimates could be higher by a factor of 2. Despite the limitations indicated, the current estimates suffice to indicate that the mean dose to the individual, as well as the mean dose to the population, should be kept as low as practicable.

Whether other somatic effects deserve to be considered in the same category with cancer in evaluating the risks of low-level irradiation remains to be determined. For those effects that may be conceived to fall into this category, however -- induction of cataracts, disturbances in the growth and development of the embryo, life-shortening from causes other than cancer, and impairment of fertility -- existing dose-effect data suggest that these are not likely to occur at dose levels compatible with present radiation protection guides. Hence, it seems reasonable to limit consideration to cancer alone for this evaluation.

Table I  
Assumed values used in calculating estimates of risk shown in Tables 2 and 3.

Age at Ir- radiation	Type of Cancer	Duration of Latent Period (years)	Duration of Plateau Region (years) <sup>a</sup>	Risk Estimate	
				Absolute Risk <sup>b</sup> (deaths/10 <sup>6</sup> / yr/rem)	Relative Risk (% incr. in deaths/rem)
In Utero	Leukemia	0	10	25	50
	All other cancer	0	10	25	50
0-9 Years	Leukemia	2	25	2.0	5.0
	All other cancer	15	(a)30 (b)Life	1.0	2.0
10 + Years	Leukemia	2	25	1.0	2.0
	All other cancer	15	(a)30 (b)Life	5.0	0.2

<sup>a</sup> Plateau region = interval following latent period during which risk remains elevated.

<sup>b</sup> The absolute risk in those aged 10 or more at the time of irradiation, for all cancer excluding leukemia, can be broken down into respective sites as follows:

<u>Type of Cancer</u>	<u>Deaths/10<sup>6</sup>/year/rem</u>
Breast	1.5*
Lung	1.3
GI incl. Stomach	1.0
Bone	0.2
All other cancer	1.0
Total	<u>5.0</u>

\* This is derived from a value of 6.0, corrected for a 50% cure rate and the inclusion of males as well as females in the population.

(From Report of U.S.A. National Academy of Sciences Committee  
on the Biological Effects of Ionizing Radiation, 1972)

Table 2

Estimated numbers of deaths per year in the U.S. population attributable to continual exposure at a rate of 0.1 rem per year, based on mortality from leukemia and from all other malignancies combined.

Age at Irradiation	ABSOLUTE RISK MODEL <sup>a</sup>		RELATIVE RISK MODEL <sup>a</sup>	
	Excess Deaths Due to:		Excess Deaths Due to:	
	Leukemia	All other Cancer	Leukemia	All other Cancer
<u>In Utero</u>	75	75	56	56
0-9 years	164	(a) 73 (b) 122	93	(a) 715 (b) 5,869
10 + years	277	(a) 1,062 (b) 1,288	589	(a) 1,665 (b) 2,415
Subtotal	516	(a) 1,210 (b) 1,485	738	(a) 2,436 (b) 8,340
TOTAL	(a) 1,726 = 0.6% increase (b) 2,001 = 0.6% increase		(a) 3,174 - 1.0% increase (b) 9,078 - 2.9% increase	

- <sup>a</sup>The figures shown are based on the following assumptions:
- (1) 1967 U.S. vital statistics can be used for age specific death rates from leukemia and all other cancer, and for total U.S. population.
  - (2) Values for the duration (a or b) of the latent period (the length of time after irradiation before any excess of cancer deaths occur), duration of risk ("plateau region"), and magnitude of average increase in annual mortality for each group are as shown in Table 1.

(From Report of U.S.A. National Academy of Sciences Committee on the Biological Effects of Ionizing Radiation, 1972)

Table 3

Estimated excess annual numbers of cancer deaths for individuals exposed from 20 to 65 years of age.

Population Exposed and dose rate	ABSOLUTE RISK MODEL			RELATIVE RISK MODEL		
	Excess Deaths Due to:			Excess Deaths Due to:		
	Leukemia	All other	Cancer	Leukemia	All other	Cancer
U.S. Pop'n 0.1 rem/yr	195	(a) 721		436	(a) 1,444	
		(b) 808			(b) 1,793	
10 <sup>6</sup> people: 5 rem/yr.	81	(a) 300		181	(a) 601	
		(b) 336			(b) 746	

(From Report of U.S.A. National Academy of Sciences Committee on the Effects of Ionizing Radiation, 1972)

# NON-IONIZING RADIATION

## BIOLOGICAL EFFECTS AND EXPOSURE STANDARDS FOR NON-IONIZING ELECTROMAGNETIC ENERGIES\*

Sol M. Michaelson  
University of Rochester  
School of Medicine  
Department of Radiation Biology and Biophysics  
Rochester, New York 14642

### Abstract

Non-ionizing radiant energies at certain frequencies, intensities, and durations of exposure can produce biological effects which may be beneficial as well as harmful. For the general population and those persons exposed or with potential for exposure to these energies, personnel exposure guidelines and product emission standards have been promulgated for some of these energies. Personnel protection guides or exposure standards are usually those established by the American National Standards Institute (ANSI), American Conference of Governmental Industrial Hygienists (ACGIH) or Department of Defense. Some industrial organizations have standards of their own which may be modifications of the national standards. Legislation for personnel exposure and product emission levels for NIR are covered under the Occupational Safety and Health Act of 1970 and the Radiation Control for Health and Safety Act of 1968, respectively. It is important that distinction be carefully made between product emission standards and personnel exposure standards and how they relate to potential injury. A proper perspective and realistic assessment of the biomedical effects of these radiant energies is essential so that the individual or general public will not be unduly exposed nor will research, development and beneficial utilization of these energies be hampered or restricted.

### Introduction

During the last quarter century there has been a marked development and increased utilization of equipment and devices for military, industrial, consumer use, and medical applications that emit a large variety of non-ionizing radiant (NIR) energies; these include ultraviolet, infrared, visible light, microwaves, and radio-frequency. Because of the biological consequences of these energies, the "Radiation Control for Health and Safety Act of 1968" (PL-90-602) and the "Occupational Safety and Health Act of 1970" (PL-91-596) (OSHA) were enacted to protect the general public as well as the worker.

---

\*The work upon which this paper is based was performed pursuant to Contract No. FDA 73-30 with the Public Health Service, Food and Drug Administration, Department of Health, Education and Welfare, with support by the Bureau of Medicine and Surgery, U.S. Navy, and with the U.S. Atomic Energy Commission at the University of Rochester Atomic Energy Project and has been assigned Report No. UR-3490-247.



The Radiation Control for Health and Safety Act requires the Secretary of Health, Education and Welfare (HEW) to prescribe performance standards for U.S. produced and imported electronic products, if he determines that such standards are necessary for the protection of public health and safety. An electronic product, under the Radiation Control Act, is any product that uses an electronic circuit and that may generate ionizing or NIR, or sound waves. Any manufactured or assembled product is covered by the Act if it emits radiation and contains an electronic circuit or functions as part of an electronic circuit. Responsibility for day-to-day administration of the Act has been delegated to the Bureau of Radiological Health (BRH).

To assure safe and healthful working conditions, OSHA provides broad authority to the Departments of Labor and HEW to develop criteria for dealing with potentially toxic materials and harmful physical agents, such as NIR, indicating safe exposure levels for workers for various periods of time.

Some NIR energies at certain frequencies, intensities, and exposure durations can produce biological effects or injury depending on multiple physical and biological variables. Although devices which utilize or emit NIR provide immeasurable benefits to mankind, they may also create potential hazards to the individual through uncontrolled and excessive emissions. Consequently, questions are being raised such as: 1) Are there any problems, and if so, how serious are they? 2) How adequate is our present knowledge about hazard to personnel from these energies? 3) How can exposure be reduced? 4) How can better regulation be obtained to reduce exposure?

For the general population and those persons exposed or with potential for exposure to these energies in the course of their occupations, personnel exposure guidelines and some product emission standards have been promulgated. Personnel protection guides or exposure standards are usually those established by the American National Standards Institute (ANSI), American Conference of Governmental Industrial Hygienists (ACGIH) or Department of Defense as well as BRH. Some industrial organizations have standards of their own which may be modifications of the national standards.

Standards

A summary of the various guidelines and standards is shown in Table I.<sup>1-5</sup>

TABLE I  
Summary of Guidelines and Standards for Non-Ionizing Radiation Exposure

Agency	Standard	Frequency	Intensity	Exposure	Comments
ANSI	Z39.18-1971	30-300 MHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects
		300 MHz-10 GHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects
ACGIH	1971	30-300 MHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects
		300 MHz-10 GHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects
DOE	1971	30-300 MHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects
		300 MHz-10 GHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects
BRH	1971	30-300 MHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects
		300 MHz-10 GHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects
OSHA	1971	30-300 MHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects
		300 MHz-10 GHz	0.01-100 W/m <sup>2</sup>	10 min	Eye and skin effects

In spite of the fact that this compilation is oversimplified and many details are omitted, it does indicate the complexity and variety of protection guides for NIR.

To insure uniform and effective control of potential health hazards from exposure to NIR, it is necessary to establish standards or protection guides. Detailed discussion of exposure standards is presented by Matelsky<sup>5</sup>, Michaelson<sup>3,7</sup>, and Schwan.<sup>8</sup>

Ideally, effect or threshold values should be predicated on firm human data. If such data are not available, however, extrapolation from well-designed, adequately-performed and properly analyzed animal investigations is required. In

discussing standards for NIR, it is necessary to keep in mind the essential differences between a "personnel exposure" standard and a "performance"

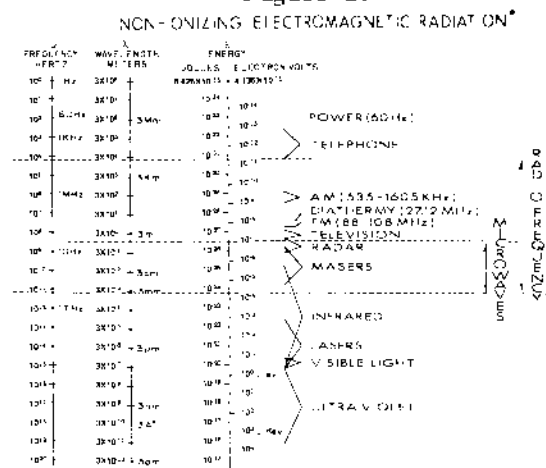
standard for a piece of equipment and how they relate to each other. An exposure standard refers to the safe (incorporating a safety factor of at least 10) level of whole-body exposure and exposure time. This standard is a guide to people on how to limit exposure for safety. An emission standard (or performance standard) refers not to people but to equipment and specifies the maximum emission close to a device which ensures that likely human exposure will be at levels far below this limit which essentially is several orders of magnitude below the personnel exposure standard. As an example, one can cite the standards for microwaves. For personnel exposure the standard is 10 mW/cm<sup>2</sup>. For microwave ovens the emission or product performance standard is 1 mW/cm<sup>2</sup> at manufacture and a maximum of 5 mW/cm<sup>2</sup> throughout the life time of the oven. This level is measured at 5 cm from the external surface and should be considered in relation to a restricted field with only a small area of the body potentially exposed.

Conceptually, as well as practically, these guidelines bear no relationship to the use of these energies in the context of medical diagnosis and treatment and should not be applied for such purposes. These standards for product emission and personnel exposure are designed to protect the general public and the worker, and are based on entirely different criteria than one would apply for diagnostic and therapeutic purposes. In the medical context, on the basis of occupational and general personnel protection standards, individuals are grossly "over-exposed" to radiant energies to achieve a specific diagnostic or therapeutic result. Diathermy at 2450 MHz creates incident energy exposures on a watt level to achieve desired tissue heating;<sup>9</sup> ultraviolet erythral doses are pushed to the limit to control serious cases of acne vulgaris and psoriasis;<sup>9</sup> Q-switched lasers are used by ophthalmologists to literally "cook" the back of the eye to restore a detached retina to a semblance of its normal anatomic position.<sup>10</sup> To draw a parallel with ionizing radiation, used therapeutically, the localized exposures of cancer patients to incident <sup>60</sup>Co gamma radiation grossly exceed current guidelines for general population and occupational exposures. This is brought out simply to emphasize that current standards are in no way applicable to medical uses of any of the non-ionizing radiant energies, nor should they be.<sup>11</sup>

Biophysics

To provide a basis for understanding the biologic effects of NIR, review of some fundamental aspects of electromagnetic radiation is indicated. The non-ionizing electromagnetic (EM) spectrum encompasses wavelengths from 3x10<sup>8</sup> m to 3x10<sup>-2</sup> nm (fig. 1).<sup>12</sup>

Figure 1.



\*Adapted from Air Force Manual AFM 161 0, 1969

As the frequency decreases, the EM energy of the emitted photons is insufficient, under normal circumstances, to dislodge orbital electrons, and produce ion pairs. The minimum photon energy capable of producing ionization in water and atomic oxygen, hydrogen, nitrogen, and carbon is between 12 and 15 electron volts (eV). Inasmuch as these atoms constitute the basic elements of living tissue, 12 eV may be considered the lower limit for ionization in biological systems. Since the energy value of 1 quantum of NIR is considerably less than 12 eV, the type of electronic excitation necessary for ionization is not possible no matter how many quanta are absorbed. NIR absorbed into the molecule either

affects the electronic energy levels of its atoms, or changes the rotational, vibrational, and transitional energies of the molecules. Changes are produced in biological systems through either photochemical (ultraviolet) and/or thermal modes (infrared, microwaves).<sup>13</sup>

A factor that has been a source of continuing concern has been the problem of measurement of energy absorbed by biological tissue. Knowledge of the incident energy is inadequate to explain what is happening within biological structures, and these occurrences must be correlated with absorbed energy. In some cases of NIR exposure we are incapable of describing the incident energy, not to speak of its absorption, as is the case in the near-field of a microwave source.

The phenomena of reflection, transmission, and energy absorption occur in biological tissues that are exposed to some NIR energies. In the case of microwaves, these phenomena occur not only at the initial entry point or exposed area, but also at deeper tissue interfaces such as the fat-fascia-muscle layers, and within tissues themselves. When microwaves are used, frequency specificity of interactions create complex problems. Considerable effort will have to be expended in this area of dosimetry before problems, controversies, and existing confusion can be resolved.

### Ultraviolet Energy

For ultraviolet (u.v.) exposure, the critical organs are the skin and eyes, resulting in erythema of the skin and skin cancer, rapid skin aging, photosensitization, and keratoconjunctivitis.

Specific absorbed wavelengths of u.v. that can elicit a specific biologic response constitute the "action spectra" for that response. These action spectra define the relative effectiveness of different wavelengths in eliciting a specific response when absorbed.<sup>6</sup> Determination of exposure criteria for u.v. effects has become increasingly difficult with the proliferation of action spectra. A great number of uncertainties still exist in what constitutes a "threshold" effect.<sup>13</sup>

In the intact animal, incident u.v. does not penetrate through the skin. Below 290 nm absorption in humans is entirely in the epidermis. Between 290 and 320 nm, less than 10 percent reaches the dermis; above 400 nm, over 50 percent reaches the dermis. Whole-body exposure to u.v. is possible, however common articles of clothing are effectively opaque to ultraviolet.

In 1948, the Council on Physical Medicine of the American Medical Association issued criteria for safe exposure to radiant energy from u.v. germicidal lamps.<sup>2</sup> This group recommended that for the primarily used wavelength, 253.7 nm, exposures should not exceed 0.5  $\mu\text{W}/\text{cm}^2$  for periods <7 h, nor 0.1  $\mu\text{W}/\text{cm}^2$  in the case of continuous exposure.

The American Conference of Governmental Industrial Hygienists (ACGIH) has also proposed threshold-limit values (TLV) for u.v.<sup>1</sup> The TLV for occupational exposure to u.v. incident upon skin or eye where irradiance values are known and exposure time is controlled are as follows: 1) For the near u.v. spectral region ( $\lambda$  320 to 400 nm) total irradiance incident upon the unprotected skin or eye should not exceed 1  $\text{mW}/\text{cm}^2$  for periods  $>10^3$  s and for exposure times  $<10^3$  s should not exceed 1  $\text{J}/\text{cm}^2$ . 2) For the actinic u.v. spectral region ( $\lambda$  200 to 315 nm), radiant exposure incident upon the unprotected skin or eye should not exceed 100  $\text{mJ}/\text{cm}^2$  for  $\lambda$  200 nm to 1000  $\text{mJ}/\text{cm}^2$  for  $\lambda$  315 nm within an 8 h period. However, the relative spectral ( $S_\lambda$ ) effectiveness of the radiation has to be taken into consideration, i.e. for 270 nm  $S_\lambda = 1.0$ , for 254 nm  $S_\lambda = 0.5$ . These are described as follows:<sup>1,13</sup>

	$\mu W/cm^2$	$\lambda$ (nm)	$S\lambda$
Maximum	$2 \times 10^4$	297	1.0
Erythematol	$3 \times 10^3$	300	0.3
Dose	$1.26 \times 10^4$	254	0.5
Maximum	$10^4$	288	1.0
Keratitic	$3 \times 10^3$	270	1.0
Dose			

A criteria document for a recommended standard for Occupational Exposure to Ultraviolet Radiation has been published by the U.S. Dept. of HEW, National Institute for Occupational Safety and Health (NIOSH). This document incorporates the ACGIH proposed TLV for Ultraviolet Exposure. These recommendations have as yet not been acted upon by OSHA.<sup>13</sup>

The above are related to exposures to the non-ionizing, non-coherent sources of u.v., where the mode of interaction with matter has been primarily a photochemical one. It is likely that exposure to low-power lasers emitting in the u.v. will also produce photochemical reactions. However, the highly collimated aspect of such generators may produce energy densities on the cornea sufficient to give rise to thermal effects. With higher output lasers, sufficient energy may penetrate the cornea of the eye and the epithelium of the skin to produce effects not known at present.<sup>13</sup>

#### Visible Energy (Light)

The hazards to man from visible light are relatively few and mostly come from artificial sources such as lasers and certain high intensity light sources which can produce absorbed energy levels greater than  $50 \text{ cal/cm}^2/\text{min}$ . Items which would probably fall in this category include high intensity reading lamps, movie and slide projector bulbs, spot lights, flood lights, etc.

The penetrating ability of visible light is slight except for transparent materials such as the lens and humors of the eye. Light entering the eye from a bright source is focused on the retina and therefore, the thermal irradiance is independent of the inverse square law for image sizes greater than the diffraction limit.<sup>14</sup> Because of its narrow depth of penetration, visible light in general does not manifest itself as a potential hazard. There are situations, however, in which it can become hazardous. For example, pulsating light at certain frequencies has been reported as a potential source for producing psychological effects. Epileptiform responses have been produced in animals and children exposed to pulsating light near the alpha rhythm frequency of the EEG.

Due to the optical properties of the eye the heat energy per unit area on a small part of the retina may be greater by a factor of  $10^5$  than on the cornea. For visible light a power density of  $1 \text{ W/cm}^2$  will exceed the threshold for pain within 1 s; with a thermal time constant of 0.1 s, the threshold energy density per pulse will be  $0.1 \text{ J/cm}^2$ . These factors become exceedingly important in relation to coherent light sources (laser). The sensation of heat, however, serves as an effective warning system under those conditions where there is time to react.

Normally, intense and bright sunlight causes maximal constriction of the pupil thus reducing the energy density on the retina. Bright sunlight, furthermore, causes painful photophobia which will not permit prolonged direct and fixed observation of the sun. The lid reflex (approximately 150 ms) is another mechanism to protect the eye. The continuous action of these measures would be adequate under normal conditions to avoid burn injuries to the retina.<sup>15</sup>

There appear to be three predominant factors controlling potential hazard to the eye: 1) intensity, 2) pupil dilation (that is, the area of exposure), and 3) length of exposure. If these factors are controlled to keep the absorbed energy below the threshold of thermal burning (reported to be between 40 and 50 cal/cm<sup>2</sup> per min), no eye injury should be expected.

### Infrared Energy

Infrared (i.r.) extends from beyond the red end of the visible portion of the EM spectrum (750 nm) to about 1x10<sup>6</sup> nm. The i.r. spectrum is frequently arbitrarily divided into three bands: the near i.r. (750-3000 nm), the middle i.r. (3x10<sup>3</sup> - 3x10<sup>4</sup> nm), and the far i.r. (3x10<sup>4</sup> to approximately 1x10<sup>6</sup> nm).

There is little evidence that photons in the i.r. (i.e., less than 1.5 eV) are capable of entering into photochemical reactions in biological systems, probably because they are too low in energy to affect the electron energy levels of these atoms. The interaction that does occur upon absorption involves an increase in the kinetic energy of the system, producing a degradation of the radiant energy to heat.<sup>6</sup>

Most biological materials are considered opaque to wavelengths shorter than 1500 nm because of the almost complete absorption of these energies by water. Radiant energies in the short wavelength region of the near i.r. can be transmitted into the deeper tissues of the dermis and the eye.

The most prominent direct effects of low wavelength i.r. on the skin include acute skin burn, increased vasodilation of the capillary beds, and an increased pigmentation which can persist for long periods of time. Under conditions of continuous exposure to high intensities of i.r., the erythematous appearance due to vasodilation may become permanent. Many factors mediate the ability to produce actual skin burn, and it is evident that for this immediate effect, the rate at which the temperature of the skin is permitted to increase is of prime importance.<sup>6</sup>

The threshold for warmth perception in the skin is reached at a warming of the skin at a rate of about .001-.002°C/s at a skin temperature of 32°C-37°C. Threshold and intensity of temperature sensation depend to a large extent on the size of the skin area changing temperature. Similarly, the minimal time of warming the skin before a temperature sensation is elicited depends on the size of the area affected and on the density of the specific temperature receptors in that area. Experimental evidence indicates that temperature sensation is little influenced by the absolute temperature of the skin and is governed by the rate of change of the skin temperature.<sup>16</sup> Results of Cook,<sup>17</sup> however, indicate that skin temperature is the vital factor in determining pain, though only in so far as this is a measure of the temperature of the thermal pain receptors below the skin surface.

The cornea of the eye is highly transparent to energies between 750 nm and 1300 nm and becomes opaque to radiant energy above 2000 nm. Thermal damage to the cornea is dependent upon the absorbed dose, and probably occurs in the thin epithelium rather than in the deeper stroma. A dose of 7.6 W s/cm<sup>2</sup> of  $\lambda$  880-1100 nm was found to elicit minimum regressive corneal damage; whereas only 2.8 W s/cm<sup>2</sup>  $\lambda$  1200-1700 nm produced this response.<sup>18</sup> These values are consistent with absorption characteristics. With excessive exposure to these critical wavelengths, there may be complete destruction of the protective epithelium, with opacification of the stroma due to coagulation of the protein. Obviously, such denaturization in an area over the pupil would seriously interfere with vision. The probability of incurring such an insult is low except where highly collimated sources can irradiate the eye without producing the sensation of pain in the surrounding skin tissue.<sup>6</sup>

Damage of the lens of the eye from i.r. has been the subject of considerable investigation over a period of many years. The term "Glass-Workers" cataract has become generic for lenticular opacities found in individuals exposed to processes hot enough to be luminous.<sup>5</sup> In 1907, Robinson<sup>19</sup> published the results of his investigations in England on the incidence of opacities on the posterior surface of the lens in the eyes of glass workers that were different than senile cataracts in appearance. It was upon his recommendation that the disease, radiation cataract, became scheduled in England as occupational in origin and by 1921 was copied into the U.S. Workman's Compensation Act. Although some serious dissent has arisen as to the validity of the data obtained by earlier investigators,<sup>20</sup> the weight of evidence as of now favors the concept that i.r. emitted from hot sources in industry is the etiological agent responsible for i.r. cataractogenesis.<sup>21</sup>

Protection guides for i.r. exposure are designed primarily for protection against ocular effects. The main difficulty, however, in devising protection standards against i.r.-induced cataract is to correlate the information on the radiation emitted during industrial processes with cataract formation. The intensities of i.r. which cause cataract are unknown. Only a small amount of experimentation on animals has been done but it has provided some knowledge of the way cataract is formed; the numerical data obtained cannot be used in devising standards, due to the relatively massive and frequent doses used in experiments, and possible physiological and anatomical differences in rabbit and human eyes.<sup>22</sup>

The tolerance limits of the human body for i.r. have been determined. An incident intensity of 0.04 cal/cm<sup>2</sup>/s of short-wave i.r. can just be tolerated by epigastric and interscapular skin areas of 144 cm<sup>2</sup>.<sup>23</sup> Approximately 25% of this energy flux would be reflected, so this corresponds to a tolerated transmitted intensity of 0.03 cal/cm<sup>2</sup>/s. It can be estimated that the maximum incident intensity of long-wave i.r. that can be tolerated by a lumbar area, 12x12 cm, is also approximately 0.03 cal/cm<sup>2</sup>/s.<sup>24</sup>

Transmission and absorption factors of the ocular media for the i.r. spectrum and threshold doses to elicit minimum damage have been determined:<sup>5,18</sup>

- 1) For corneal damage: 7.6 J/cm<sup>2</sup>, 800-1100 nm; 2.8 J/cm<sup>2</sup>, 1200-1700 nm.
- 2) Corneal exposure to produce damage in the iris: 10.8 J/cm<sup>2</sup>, 800-1100 nm.
- 3) Corneal exposure for production of retinal burns: 1 J/cm<sup>2</sup> (this value determined with a 0.1 s exposure to 20-40 J/cm<sup>2</sup> causing a 1 mm burn).

### Laser

The acronym LASER (light amplification by stimulated emission of radiation) is commonly applied to a device which produces an intense, coherent directional beam of light by stimulating electronic or molecular transitions to lower energy levels.<sup>5</sup> The characteristics of lasers which influence their effect upon biological systems include the duration of the pulse, the time interval between pulses, the specific wavelength emitted, and the energy density of the beam. The degree of damage produced depends upon the absorbing tissue, its absorption characteristics, the size of the absorbing area, and its vascularity.<sup>6</sup> It has become common practice to describe the output of pulsed lasers in terms of energy (joules), and that from continuous wave (CW) lasers in terms of power (watts). The J/cm<sup>2</sup> is used to express absorbed energy density, and the W/cm<sup>2</sup> to describe power density.<sup>6</sup>

Biologic effects can occur through three mechanisms of interaction: a) thermal effect; b) acoustic transients; or c) other phenomena.<sup>6</sup> The latter

two effects are only seen with high power density laser pulses. When laser light impinges on tissue, the absorbed energy produces heat. The resultant rapid rise in temperature can easily denature tissue protein. Since tissue is not homogeneous, light absorption is not homogeneous and the thermal stress is greatest around those portions of tissue that are the most efficient absorbers. Rapid and localized absorption produces high temperatures. Steam production, evident only at high exposure levels, can be quite dangerous if it occurs in an enclosed and completely filled volume such as the cranial cavity or the eye. A second interaction mechanism is an elastic or acoustic transient or pressure wave. As the light pulse impinges on tissue, a portion of the energy is transduced to a mechanical compression wave (acoustic energy), and a sonic transient wave is built up. This sonic wave can rip and tear tissue and if near the surface, can send out a plume of debris from the impact. Other phenomena such as free radical formation, are believed to exist during laser impact on biological systems, but this has not yet been conclusively demonstrated.<sup>2,6</sup>

The primary hazard from laser radiation is exposure of the eye. Exposure levels, if kept below those damaging to the eye, will not harm other tissues and organs of the body. Eye damage can range from mild retinal burns, with little or no loss of visual acuity, to severe lesions with loss of central vision, and total loss of the eye from gross over-exposure. Long-term exposure of the retina to wavelengths in the visible spectrum, at levels not far below the burn threshold, may cause irreversible effects.

Excessive i.r. (1.4-1000  $\mu\text{m}$ ) exposure causes a loss of transparency or produces a surface irregularity in the cornea. Damage results from heating of the cornea by absorption of the incident energy by tears and tissue fluid in the cornea. Although the critical temperature threshold is not known, it does not appear to be much above normal body temperature, and there are indications that it is a function of exposure time.<sup>5</sup>

Excessive u.v. (0.2-0.4  $\mu\text{m}$ ) exposure produces photophobia accompanied by redness, tearing, conjunctival discharge, surface exfoliation, and stromal haze. Damage to the corneal epithelium by absorption of u.v. probably results from photochemical denaturation of proteins or other molecules in the cells. Some of the most important molecules are the desoxyribonucleic acids (DNA) and ribonucleic acids (RNA). The absorption is probably by selective sensitive portions of single cells. Thus the action of the u.v. is photochemical rather than thermal, since the temperature rise calculated for experimental exposure is negligible.<sup>5</sup>

The type of damage inflicted on the eye by laser beams ranges, therefore, from a small and inconsequential retinal burn in the periphery of the fundus, to severe damage of the macular area, with consequent loss of visual acuity, up to massive hemorrhage and extrusion of tissue into the vitreous, with possible loss of the entire eye.<sup>2,7</sup>

The large skin surface makes this tissue readily available to accidental and repeated exposures to laser energies. The biological significance of exposure of the skin to lasers operating in the visible and i.r. regions is considerably less than exposure of the eye, as skin damage is usually repairable or reversible. Effects may vary from a mild reddening (erythema) to blisters, and charring. Depigmentation of the skin, ulceration and scarring and damage to underlying organs may occur from extremely high powered laser sources.

Latent and cumulative effects of laser exposure are not known at this time. Little or no data are available describing the reaction of skin exposed to lasers in the 0.2-0.4  $\mu\text{m}$  spectral region, but chronic exposure to u.v. in this range can have a carcinogenic action on skin as well as eliciting an erythematous response.

On the basis of studies with non-coherent u.v., exposure to wavelengths in the 0.25-0.32  $\mu\text{m}$  spectral region is most injurious to skin. Exposure to the shorter (0.20-0.25  $\mu\text{m}$ ) and longer (0.32-0.40  $\mu\text{m}$ ) u.v. is considered less harmful to normal human skin. The shorter wavelengths are absorbed in the outer dead layer of the epidermis (stratum corneum), and exposure to the longer wavelengths has merely a pigment-darkening effect. The sensitivity of skin, however, to the longer wavelengths may be increased by known or inadvertent usage of photosensitizers.<sup>5</sup>

One cannot discuss potential hazards from laser energies without mentioning operationally associated hazards such as: 1) compressed gases, 2) cryogenic liquids, 3) ionizing radiation that may emanate from laser power supplies and components, 4) toxic materials used in laser targets or laser system elements. Attention should also be paid to adequate ventilation to eliminate or reduce exposure to toxic materials to safe levels.<sup>5</sup>

ANSI, ACGIH, and BRH have or are in the process of developing laser standards. Selected maximum permissible exposure (MPE) laser levels for ocular effects are shown in Table II. These have to be understood, however, in the context and with consideration of the laser classification system that has been developed.<sup>5</sup> Because of the complexity of these standards, the ANSI standard<sup>5</sup> and the ACGIH, TLV<sup>7</sup> should be consulted.

TABLE II  
Maximum Permissible Exposure (MPE) to Laser for Direct Ocular  
Intrabeam Viewing for Single Pulses or Exposures<sup>a</sup>

Spectral Region	Wave Length	Exposure Time, (t) Seconds	MPE
Ultraviolet	200 nm to 302 nm	$10^{-2}$ to $3 \times 10^{-4}$	$3 \text{ mJ} \cdot \text{cm}^{-2}$
	305 nm	" "	$10 \text{ mJ} \cdot \text{cm}^{-2}$
	310 nm	" "	$100 \text{ mJ} \cdot \text{cm}^{-2}$
	315 nm	" "	$1.0 \text{ J} \cdot \text{cm}^{-2}$
	315 nm to 400 nm	$10^{-2}$ to $10^{-4}$	$1.0 \text{ W} \cdot \text{cm}^{-2}$
Visible and Near Infrared	400 nm to 1.4 $\mu\text{m}$	$10^{-9}$ to $2 \times 10^{-5}$	$5 \times 10^{-3} \text{ J} \cdot \text{cm}^{-2}$
	" "	$10$ to $10^4$	$1.0 \text{ mJ} \cdot \text{cm}^{-2}$
	" "	$10^6$ to $3 \times 10^4$	$10^{-6} \text{ W} \cdot \text{cm}^{-2}$
Far Infrared	1.4 $\mu\text{m}$ to $10^3 \mu\text{m}$	$10^{-9}$ to $10^{-7}$	$10^{-7} \text{ J} \cdot \text{cm}^{-2}$
	" "	$10$ to $3 \times 10^4$	$0.1 \text{ W} \cdot \text{cm}^{-2}$

<sup>a</sup>Adapted from ANSI ( 5 ); ACGIH ( 7 ).

### Microwaves/Radiofrequency

The radiofrequency (rf) portion of the EM spectrum is considered to extend from 0.03 MHz (very low frequency -VLF) to 300,000 MHz (extremely high frequency -EHF). On a functional or operational basis, frequencies in the region from 100 MHz to 300,000 MHz (300 GHz) are designated as microwaves.

Of the various NIR energies, the rf and microwave bands have elicited the greatest interest and concern as well as confusion in consideration of the real and substantiated effects vis a vis unsubstantiated or speculative effects. When considering the bio-

logical effects of rf energy, the wavelength of the energy and its relationship to the physical dimensions of exposed objects become important factors. Absorption of the energy depends upon the dielectric properties of the tissues and the relative absorption cross section of the exposed subject. Thus, the size of the object with relation to the wavelength of the incident field plays an important role.<sup>2,8</sup>

In biological systems absorbed rf is transformed into increased kinetic energy of the absorbing molecules, thereby producing a general heating of the tissue. Such heating results from both ionic conduction and vibration of the dipole molecules of water and proteins.<sup>9</sup> The absorption of rf is dependent upon the electrical properties of the absorbing medium, specifically, its dielectric constant and electrical conductivity. These properties change as the frequency of the applied electric field changes. Values of dielectric constant and electrical conductivity and depth of penetration have been determined for many tissues.<sup>30</sup> The absorption of rf energy is high and the depth of penetration low in tissues of high water content such as muscle, brain tissue, internal organs, and skin, while the absorption is lower in tissues of low water content such as fat and bone. Reflections between interfaces separating tissues of high and low water content can produce standing waves



accompanied by "hot spots" that can be maximum in either tissue, regardless of dielectric constant or conductivity.<sup>29</sup>

Extensive investigations into microwave bioeffects conclusively show that for frequencies between 200 MHz and 24,500 MHz, exposure to power density of  $>100 \text{ mW/cm}^2$  for 1 h or more could have pathophysiologic manifestations of a thermal nature. At power densities  $<100 \text{ mW/cm}^2$ , however, evidence of pathologic change is non-existent or equivocal. According to the best evidence available, the most important, if not the only, effect of microwave absorption in the mammal is the conversion of the absorbed energy into heat. Whole-body exposure of various species of animals to microwaves at levels  $>10 \text{ mW/cm}^2$  is characterized by a temperature rise which is a function of the thermal regulatory process of the animal. The end result is either reversible or irreversible change depending on the conditions of the exposure and the physiologic state of the animal.<sup>31</sup> Smaller animals show a greater temperature response than do larger animals at equivalent exposures.<sup>32</sup>

Irradiance levels of 10,000 MHz and 3000 MHz microwaves required to produce a threshold sensation of warmth have been obtained.<sup>33,34</sup> These data indicate that when a  $40 \text{ cm}^2$  area of the face is exposed to microwaves, thermal sensation can be elicited within 1 s at a power density of  $21 \text{ mW/cm}^2$  for 10,000 MHz and  $58.6 \text{ mW/cm}^2$  for 3000 MHz. Within 4 s the threshold is lowered by approximately 50%, i.e.  $12.5 \text{ mW/cm}^2$  (10,000 MHz) and  $33.5 \text{ mW/cm}^2$  (3000 MHz). On this basis, if the entire face were to be exposed, the threshold for thermal sensation to 10,000 MHz would be  $4-6 \text{ mW/cm}^2$  within 5 s or approximately  $10 \text{ mW/cm}^2$  for a 0.5 s exposure. Threshold for pain reaction to 3000 MHz exposure of a  $9.5 \text{ cm}^2$  area of the forearm ranges from  $830 \text{ mW/cm}^2$  for exposures longer than 3 min to  $5.6 \text{ W/cm}^2$  for a 20 s exposure period. If a larger area ( $53 \text{ cm}^2$ ) is exposed, the pain threshold for a 3 min exposure is  $560 \text{ mW/cm}^2$ .<sup>17</sup> These data and other information on microwave sensation suggest that cutaneous perception of microwaves may provide a protection factor with sufficient margin of safety constituting a warning mechanism to prevent exposure to microwaves at levels that could be injurious.<sup>3</sup>

Microwaves have been shown to produce cataracts in some experimental animals, notably rabbits, and there are also reports of microwave-induced cataracts in man. In animal studies, the techniques used and interpretation of the results and conclusions, however, are quite often equivocal. Careful review of the reports on human cataractogenesis indicates that there has been insufficient quantitation and correlation of pathophysiology with the level of microwave exposure.

Carpenter and his associates<sup>35</sup> have reported that single or repeated exposures of rabbits' eyes with 2450 MHz pulsed or CW can cause opacity when the lens temperature increases  $4^\circ\text{C}$ . These authors have suggested a "cumulative" effect on the lens from repeated "sub-threshold" exposures of rabbits' eyes to microwaves.

In order not to confuse this suggested "cumulative" effect with that recognized for ionizing radiation, it is important to define the cumulative effect produced by ionizing radiation to put this point in its proper perspective. Cumulative injury from exposure to ionizing radiation is a manifestation of the irreparability of a certain fraction of the injury which has been designated as Residual Radiation Injury. Such Residual Radiation Injury is additive with frequency of exposures and is not dependent on intervals between exposures once the full recovery potential has been realized.<sup>36</sup> A cumulative effect is the accumulation of damage resulting from repeated exposures each of which is individually capable of producing some degree of damage. Careful analysis of the work of Carpenter *et al*<sup>35</sup>, as well as Williams *et al*<sup>37</sup> and Birenbaum *et al*<sup>38</sup> reveals that whenever lens opacity is produced in animals, a threshold ( $>100 \text{ mW/cm}^2$ ;  $>1 \text{ h}$ ) becomes obvious. No one

has yet been able to produce cataracts even by repetitive exposures when the power density is really below threshold.

It is important to note that lens opacity has consistently been produced in only one species, namely the rabbit. One can question whether the rabbit is the most appropriate animal model. According to Cogan *et al.*<sup>39</sup>, with local microwave exposure the cataractogenic level for monkeys has been found to be higher than for rabbits.

Most epidemiological studies in the U.S. have involved the ocular lens. The few reports<sup>40,41,42,43,44</sup> that are available are highly questionable and have not been found acceptable by competent ophthalmologists.

The suggestion that microwaves may interact with the central nervous system (CNS) by some mechanism other than heating has been made by several investigators, mostly in East European countries, who stress that the CNS must be considered as being moderately or highly sensitive to rf or microwave energy absorption. The first report on the effects of microwaves on conditional response activity of experimental animals was made by Gordon *et al.*<sup>45</sup> In subsequent years, the study of the "nonthermal" effects of microwaves gradually occupied the central role in electrophysiological studies in the Soviet Union.<sup>46</sup>

Many investigators do not accept the possibility of nonthermal neural stimulation by microwaves and explain these effects entirely upon local heating.<sup>47,48,49</sup> They suggest that thermal stimulation of the peripheral nerves could produce the neurophysiological and behavioral changes that have been reported.

Eastern European investigators have contributed most of the reports of effects of rf and microwave energies in man.<sup>45,46,50,51</sup> The greatest emphasis is placed on effects produced at less than "thermogenic" power flux densities (<10 mW/cm<sup>2</sup>). According to these investigators, the basic symptomatology and neuropathology underlying all of the reported syndromes is described as due to the functional disturbance created in the CNS by "non-thermal" mechanisms. These effects are reported to occur in occupational exposures at levels far below those required to produce a temperature rise. The symptoms are manifested by weakness, fatigue, vague feelings of discomfort, headache, drowsiness, palpitations, faintness, memory loss, and confusion. These syndromes are apparently completely reversible in most cases, with little or no time lost from work.<sup>52</sup> Much of these reports is based on subjective rather than objective findings, and measurement of field intensity in most cases is not comparable from worker to worker or factory to factory.<sup>53</sup>

Dodge,<sup>54</sup> in his review of the Soviet research in this area, has stated, "An often disappointing facet of the Soviet and East European literature on the subject of clinical manifestations of microwave exposure is the lack of pertinent data presented on the circumstances of irradiation...important environmental factors (heat, humidity, light, etc.) are often omitted from clinical and hygienic reports." A point that should be noted is that in the West the effects reported by East European investigators have not been observed, even at much higher exposure levels.

Microwave exposure standards for most of the Western world are based, with minor variations, on standards developed in the U.S. (table III). The original U.S. standard was tentatively adopted about 15 years ago on the basis of theoretical considerations by Schwan and his associates. This standard was based on the amount of exogenous heat which the body could tolerate and dissipate without any resulting rise in body temperature. This tolerance level was calculated to be 10 mW/cm<sup>2</sup> for continuous exposure. Intensive investigation into the biological effects of microwaves was subsequently carried out by the U.S. Department of Defense. None of these investigations was able to produce any evidence for a biological effect at levels even approaching the theoretical level of 10 mW/cm<sup>2</sup> and, indeed, no conclusive evidence was established for any effect below the level of 100 mW/cm<sup>2</sup>.<sup>31</sup>

The ANSI standard of 10 mW/cm<sup>2</sup> for radiofrequency exposure recommended in 1966 and reaffirmed in 1973,<sup>4</sup> is roughly a factor of ten below thresholds of damage by thermal effects, assuming a long duration of exposure--i.e., one quarter h or more. The 10 mW/cm<sup>2</sup> level is based on thermal equilibrium conditions for whole-body exposure. For normal environmental conditions and for incident electromagnetic energy of frequencies from 10 MHz to 100 GHz, the radiation protection guide is 10 mW/cm<sup>2</sup> and the equivalent free-space electric and magnetic field strength: approximately 200 V/m RMS and 0.5 A/m RMS, respectively. For modulated fields, power density and the squares of the field intensities are averaged over any 0.1 hour period, i.e. none of the following levels should be exceeded in any 0.1 hour period: Electric Field Strength Squared - 40000 V<sup>2</sup>/m<sup>2</sup>; Magnetic Field Strength Squared - 0.25 A<sup>2</sup>/m<sup>2</sup>; Power Density - 10 mW/cm<sup>2</sup>; Energy Density - 1 mWh/cm<sup>2</sup>; this guide applies whether the radiation is CW or intermittent.<sup>4</sup>

Table III.

RECOMMENDED MAXIMUM PERMISSIBLE EXPOSURE LIMITS FOR RADIO-FREQUENCY RADIATION			
MAXIMUM PERM. TO THE WORKER TO	FREQUENCY (MHz)	COUNTRY OR SOURCE	SPEC. LOCATIONS
10 mW/cm <sup>2</sup>	10-100,000	U.S.A., S. I., 1966; CANADA 1966	1 mW/cm <sup>2</sup> FOR EACH 8 MIN.
	30-100,000	SWISS PATENT 1966	HAIRY EXPOSURE
	1000-10000	SCANDINAVIA 1958	WHOLE BODY
ALL	ALL	U.S. ARMY AND AIR FORCE 1945	10 mW/cm <sup>2</sup> WENT. EXP.
			10-100 MHz, LIM. OCCUP.
			NIR - 1000 (mW/cm <sup>2</sup> )
ALL	ALL	SWEDEN 1961	INDU. EXP. (OCCUPAT.)
		GERMAN FED. REPUBLIC 1951	.....
		SWEDEN 1958	GENERAL PUBLIC PROLONGED OCCUPAT. EXP.
1 mW/cm <sup>2</sup>	> 100	USSR 1965; POLAND 1964	1-100 MIN/DAY
		JSSR 1965; POLAND 1951	2-11 HR/DAY
		CZECHOSLOVAKIA 1965	10-11 HR/DAY
0.1 mW/cm <sup>2</sup>	> 100	USSR 1965; POLAND 1965	8 HR/DAY
		CZECHOSLOVAKIA 1965	ENTIRE DAY PL. SPD & HR/DAY
0.01 mW/cm <sup>2</sup>	0.01-100	USSR 1965	PHLEBO-8 HR/DAY
		CZECHOSLOVAKIA 1965	
		JSSR 1965	

There is no evidence in the scientific or medical literature of the Western world, that the present U.S. standards represent a hazardous exposure level. The ANSI standard<sup>4</sup> has been accepted by OSHA and with very little modification throughout the Western world. Microwave exposure standards for most of the Eastern European nations are based, with minor variations, on limits established by the USSR (table III). These limits, promulgated in 1959 by the USSR Ministry of Health are: a) 0.01 mW/cm<sup>2</sup> for an entire workday; b) 0.1 mW/cm<sup>2</sup> for 2 h; c) 1.0 mW/cm<sup>2</sup> up to 20 min. These standards are based on vague "asthenia" syndromes reported by individuals who work with microwave/rf energies. These effects have not been demonstrated by Western investigators.

The apparent discrepancy in maximum allowable exposures between Eastern European and Western countries may be due to differences in industrial hygiene philosophy. Magnuson et al<sup>5,5</sup> have noted that in the USSR, MPE is based on presence or absence of biological effects without regard to the feasibility of reaching such levels in practice. The MPE represents a desirable level for which to strive rather than an absolute value to be used in practice.

### Conclusion

It is important to maintain a proper perspective, and assess realistically the biomedical effects of NIR so that the worker or general public will not be unduly exposed nor will research, development and beneficial utilization of these energies be hampered or restricted. There is a need for scientific competence and integrity. Although there is considerable agreement among scientists concerning the biologic effects and potential hazards of NIR, there are areas of disagreement, especially in relation to rf and microwaves. It is essential that research into the biologic effects of NIR be fostered and advanced to counteract the often voiced "what we don't know can hurt us" attitude with consequent overly restrictive and unrealistic standards. Free international exchange and closer personal contact between scientists would be invaluable in resolving discrepancies and divergence of opinion that exist in the understanding of some of the biologic and clinical implications of exposure to NIR and approaches to the setting of standards or protection guides.

Acknowledgement - The assistance of Mrs. Margaret Anderson Bush in organizing the material and typing this manuscript is acknowledged with appreciation.

## References

- [1] Threshold Limit Values for Chemical Substances and Physical Agents. Am. Conf. Govern. Industr. Hyg., Cincinnati, 1972, 94p. [2] Coun. Phys. Med. Amer. Med. Assoc., J.A.M.A. 137, 1600, 1948. [3] S.M. Michaelson. Proc. IEEE 60, 389, 1972. [4] U. S. Amer. Stand. Instit. USASI C95.1, New York, 1966. [5] Amer. Nat. Stand. Instit. Standard for the Safe Use of Lasers. ANSI Z/136.1, New York, 1972, 61p. [6] I. Matelsky. In: Industr. Hyg. Highlights. vol. 1, G.V. Cralley, G.D. Clayton (eds.), Pittsburgh, 1968, p. 140. [7] S.M. Michaelson. Amer. Indust. Hyg. Ass. J. 33, 156, 1972. [8] H.P. Schwan. IEEE Trans. Bio. Med. Eng. BME-19, 304, 1972. [9] F.H. Krusen et al (eds.). Handbook of Physical Medicine and Rehabilitation. W.B. Saunders, Philadelphia, 1971, 725p. [10] P.E. McGuff. Surgical Applications of Laser. Charles C. Thomas, Springfield, 1966, 200p. [11] S.M. Michaelson. In: Health Physics Society 7th Midyear Topical Symp. - Health Physics and the Healing Arts. San Juan, Puerto Rico, Dec. 1972, p. 44. [12] Laser Health Hazards Control. USAF, AFM-161-B, Wash., D.C., 1971, 45p. [13] I. Matelsky. The Non-Ionizing Ultraviolet Radiations. Amer. Industr. Hyg. Ass. Refresher Course, 1973. [14] K. Buettner, H.W. Rose. Sight Saving Rev. 23, 194, 1953. [15] W.J. Geeraets. Industr. Med. 39, 441, 1970. [16] E. Fischer, S. Solomon. In: S.H. Licht (ed.). Therapeutic Heat. E. Licht, New Haven, 1958, p.116. [17] H.F. Cook. J. Physiol. 118,1, 1952. [18] J.H. Jacobson et al. AMRL-TDR-62-96, 6570th Aerospace Med. Res. Lab., Wright-Patterson AFB, Ohio, 1962. [19] W. Robinson. Brit M.J. 2,381, 1907. [20] K.L. Dunn. Arch. Ind. Hyg. Occup. Med. 1,166, 1950. [21] D.G. Cogan et al. Arch. Ophthalmol. 47,55, 1952. [22] C.M. Edbrooke, C. Edwards. Arch. Occup. Hyg. 10,293, 1967. [23] D.L. Lloyd-Smith, K. Mendelsohn. Brit. M.J. #4559,975, 1948. [24] H.K. Whyte. Clin. Sci. 10,333, 1951. [25] L. Goldman. Biomedical Aspects of the Laser. Springer, New York, 1967. [26] W.F. Van Pelt et al. Laser Fundamentals and Experiments. U.S.D. HEW, PHS, BRH, Publ. No. BRH/SWRHL 70-1, 1970. [27] T.L. Curtin, D.G. Bayden. Amer. J. Ophthal. 65,188, 1968. [28] A. Anne et al. In: M.F. Peyton (ed.). Biological Effects of Microwave Radiation. Plenum, New York, 1961, p.153. [29] C.C. Johnson, A.W. Guy. Proc. IEEE 60,692, 1972. [30] H.P. Schwan, K. Li. Proc. IRE 41,1735, 1953. [31] S.M. Michaelson. IEEE Trans. Microwave Theory and Techniques MTT-19,131, 1971. [32] S.M. Michaelson. Thermal effects of single and repeated exposures to microwaves - a review. International Symposium on Biologic Effects and Health Hazards of Microwave Radiation, Warsaw, 1973. [33] E. Hendler. In: J.D. Hardy (ed.). Thermal Problems in Aerospace Medicine. Unwin, Ltd., Surrey, 1968, p. 149. [34] E. Hendler et al. In: J.D. Hardy (ed.). Temperature Measurement and Control in Science and Industry. Pt. 3. Biology and Medicine. Rheinhold, New York, 1963, p.221. [35] R.L. Carpenter et al. Proc. 3rd Int. Conf. Med. Elect. (London) 3,401, 1960. [36] H.A. Blair. Ann. N.Y. Acad. Sci. 114,150, 1964. [37] D.B. Williams et al. Arch. Ophthal. 54,863, 1955. [38] L. Birenbaum et al. IEEE Trans. Bio. Med. Eng. BME-16,7, 1969. [39] D.G. Cogan et al. Arch. Ind. Health 18,299, 1958. [40] M. Zaret. 39th Ann. Sci. Meeting Aerospace Med. Ass., Miami, 1968. [41] M. Zaret. 40th Ann. Sci. Meeting Aerospace Med. Ass., San Francisco, 1969. [42] F.G. Hirsch, J.T. Parker. Arch. Indust. Hyg. 6,512, 1952. [43] G.H. Kurz, R.B. Einaugler. Am. J. Ophthal. 66,866, 1968. [44] I.S. Shimkovich, V.C. Shilyayev. Vestn. Oftal. 72,12, 1959. [45] Z.V. Gordon. Biological Effect of Microwaves in Occupational Hygiene. Izdatel'stvo "Meditsina" Leningrad Otdelenie, 1966, 164p. [46] Yu.I. Novitsky et al. Radio Frequencies and Microwaves. Magnetic and Electrical Fields. NASA Tech. Trans. TT F-14,021, 1971. [47] L.R. Pinneo et al. The Neural Effects of Microwave Radiation. RADC-TDR-62-231, AD 722684, Arlington, 1962. [48] R.D. McAfee. IEEE Trans. Microwave Theory and Techniques MTT-19,251, 1971. [49] R.D. McAfee. Am. J. Physiol. 203,374, 1962. [50] I.R. Petrov (ed.). Influence of Microwave Radiation on the Organism of Man and Animals. Meditsina, Leningrad, 1970. [51] A.S. Presman. Electromagnetic Fields and Life. Izd-vo Nauka, Moscow, 1968. [52] Yu.A. Osipov. Occupational Hygiene and the Effect of Radio-Frequency Electromagnetic Fields on Workers. Izd. Meditsina, Leningrad, 1965. [53] J. Pazderova. Prac. Lek. 20,447, 1968. [54] C.H. Dodge. In: Symp. Proc. (PB 193-898). USDHEW, PHS, BRH, Rockville, 1970. [55] H.J. Magnuson et al. Amer. Indust. Hyg. Ass. J. 25,185, 1964.

A PROGRAMME ON NON-IONIZING RADIATION PROTECTION OF THE  
WORLD HEALTH ORGANIZATION REGIONAL OFFICE FOR EUROPE

Dr Michael J. Suess  
WHO Regional Office for Europe  
Copenhagen, Denmark

Abstract

Increased and wider attention is being paid at present to a relatively new physical hazard, namely, non-ionizing radiation (NIR), arising from different devices. To promote the study of the health effects from NIR the WHO Regional Office for Europe has developed, within its long-term programme on environmental pollution control, a sector programme on NIR with activities extending to the year 1979. The mode of action for the control of NIR and the programme's priorities are based on the recommendations of a Working Group convened by this Office in November 1971.

Non-ionizing Radiation

During recent years there has been an increased development and use of equipment which can produce a number of types of radiation which may be hazardous both to the user and to the general public if necessary precautions are not taken, especially when large quantities of energy are being transmitted. None of these radiations can, after absorption, transmit enough energy to produce ion pairs, as is the case with the ionizing radiations. Free radical formation has been observed after exposure to ultrasound, and photochemical effects may be observed after exposure to ultraviolet radiation, but in most cases the energy absorption tends to be manifest in the form of heat. The differences in modes of production and in areas of use are, however, so great that it is necessary to discuss each type of radiation separately. The differences in energy even within a single type of radiation are of particular importance for the evaluation of the hazards from exposure to NIR. The ability to penetrate into the human body and the places of absorption of the radiation will depend on this characteristic and will differ from one type of radiation to another.

The NIR devices are finding an ever increasing use in industry, engineering, telecommunication, medicine, research, education, and by the general public. This gives rise to a number of questions:

- How serious are the problems linked with NIR, what are their dimensions and what acute and/or chronic effects on the human body are involved?
- How adequate is our present knowledge about occupational risks and public health hazards from these radiation types?
- How can the radiation exposure be reduced?

- How can better regulations be drafted and enforced to reduce the exposure to NIR?

Moreover, while the implications to occupational personnel and the general public from the use of these devices may be mainly limited to developed countries, it is important to start as soon as possible, preferably on an international basis:

- to collect and evaluate informative data in a systematic manner,
- to support further investigations into the health hazards involved,
- to discuss possible biologic criteria for damage,
- to develop guides and criteria for health protection, and
- to assist in the establishment of rules and regulations and the proper means of enforcing them.

Ultraviolet Radiation. This has been used extensively for sterilization of equipment and air, and in different types of medical apparatus. The use appears at present to be somewhat declining, but a certain risk to the occupational worker in rooms supplied with open ultraviolet sources is still present. The damage will be localized to the eye and to the skin, but with a certain long-term risk for ultraviolet carcinogenesis. It is evident that the largest exposed group is that of persons working in open air and attention should be drawn to protective cosmetics. This stands true also for entertainers as an occupational group. No quantitative knowledge of dose-effect relationship and of latency periods is available as far as human cancer is concerned. The ultraviolet lamps for private use are widely disseminated among the public and should be supplied with appropriate warnings.

Infrared Radiation. Exposure to this light can occur in almost any industry from direct infrared sources as well as from other heat sources, and the risks under certain working conditions are well known. Still unsolved is the question as to whether infrared radiation can produce cataract. In any case, the presence of well developed temperature sensors in the skin around the eye represents a good biological warning system.

Microwave Radiation. The recent great expansion in use of microwave ovens presents a new serious health hazard. The damage will affect the eye (cataract formation) and the skin and other locations by heating. In certain instances genetic effects may perhaps result. The risk from big radar installations emitting much energy should also be considered. Microwaves are recognized as the type of NIR which represents the largest risk. At present there exists no possibility for adequate measurements of absorbed dose and the special measuring problems of near and far fields cannot be neglected. In general, the thermal type of effects has been considered the most important one. However, additional effects, such as those on the nervous system, have been reported and require further attention.

Lasers. The risks from lasers must receive much attention with their increasing use. The emitted light can give rise to damage to the eye and the skin, and under certain conditions perhaps also to more deep lying organs. A special risk appears to be connected with lasers emitting non-visible light where unnoticed temperature gradients may be produced. The difficulties of evaluating the risks from lasers are partly due to the lack of agreement in the translating of animal experiments to the human eye. In the case of pulsed exposure the differences of effectiveness of heat transfer from different localities will decide the biologic effects in connexion with the duration of the pulses, but existing experiments are not too convincing. Also the question of a risk of shock waves from a pulsed laser is not sufficiently well studied to be fully understood.

Ultrasonic Radiation. This type of radiation is a relatively new energy source and its versatility has led to its widespread employment in various industrial, medical and scientific products for measurement and control applications and to modify the material by the dissipation of energy. While the possible damage to the human eye has already been observed, the rapidly expanding use of ultrasonic power calls for further study of levels and other biologic effects of stray radiation. Ultrasound can be produced in a large spectrum. It is partly made safe by its inability to pass an air-water interface. However, whether potential adverse effects exist from immersion of hands in ultrasonic fields during cleaning are not known. It is not definitely solved whether chromosome aberrations can be produced but the evidence tends to be negative. So far no major adverse effects have been recognized from diagnostic exposure of children in utero.

#### The Long-term Programme

It was the favourable acceptance of the Office's long-term programme on environmental pollution control and its approval by the Regional Committee of the European Region at its 19th session in Budapest in 1969 that eventually led to the development of a programme on NIR protection. The first activity on the subject was the convention of a Working Group in The Hague in November 1971, held to discuss the health effects of ionizing and non-ionizing radiation. The members of that Working Group reviewed and assessed the situation prevailing in Europe, studied trends and developments, discussed needed activities of special importance, and recommended actions and projects to be undertaken. The Working Group's deliberations were summarized in a report published by the Regional Office which also included detailed conclusions and recommendations.<sup>1</sup>

The first half of the Working Group's recommendations deal with NIR in general and touch on needed health studies, the establishment of reference centres, the preparation of model codes of practice, education and licensing, while the second half concentrates on the specific types of radiation. Noting the lack of regular action on NIR protection by any international agency or committee, the Working Group urged the WHO to initiate the setting-up of an organ with terms of references in the field of NIR similar to those of the ICRP and ICRO in the area of ionizing radiations (recommendation 8). Recommendation 5 is also of much interest, as it calls for the establishment of an international reference centre which should, among other activities, co-ordinate badly-needed studies on an international level. Many of the Working Group's recommendations have been transformed into a working plan as shown in figure 1. This working plan identifies the various activities which are expected to be carried out by the WHO Regional Office for Europe. Because of their inter-regional nature (i.e. concerning more than one WHO regional office) some recommendations of the Working Group cannot be implemented at the regional office level but may be carried out only through WHO headquarters. For example, this would be the case for the establishment of an international reference centre.

The NIR sector within the long-term programme can be divided into six activity components, namely, review, survey, study, manual, education and evaluation. The activities which have already taken place, or those in progress, are described in more detail in the coming sections. With respect to field and laboratory studies planned for 1974 to 1978, their objectives will be more closely identified following the recommendations from the various NIR scientific meetings already planned to discuss specific sources. In 1979, when the present series of planned activities will have been exhausted, a meeting will take place to evaluate the work already done and make recommendations on further needed action within the NIR sector of the long-term programme to the

implementation of which the WHO Regional Office for Europe can contribute (Fig. 2).

Range Limits of NIR. At the conclusion of the first Working Group's meeting, the inconsistency of the range limits of the various types of NIR as defined by different investigators could not be overlooked. It is, of course, recognized that any such limits were approximations, as no exact end-point can be defined. However, to advance future fruitful discussions and collaborations at an international level, the various NIR ranges have now been fixed and are being adhered to by WHO for its own activities on NIR (table 1). Thus, for example, the microwave radiation range with the given limit of 300 GHz to 300 MHz has been adopted for use by the forthcoming International Symposium on Biologic Effects and Health Hazards from Microwave Radiation to be held near Warsaw, 15 - 18 November, 1973.

Survey of Institutions. The worldwide identification of the existing institutions and the affiliated scientists active, at least to some extent, in the study of the biologic effects and health hazards from NIR and in the development of appropriate dosimetry and protection measures is a must if progress is to be expected in the establishment of internationally agreed units and procedures for dose measurement and analysis of results. Moreover, such agreements are necessary for the comparison of, for example, epidemiological studies of long-term effects from exposure to all types of NIR, and in general, for the meaningful collaboration between the various institutions on exchange of information, correlation of findings, joint research projects and the development of internationally accepted safety guidelines and protection standards.

Towards this goal, the European Regional Office has embarked on a survey of institutions and specialists in the field of NIR which, when completed, will provide the information for a Directory of Institutions. Meanwhile, a preliminary survey has revealed that only in a relatively small number of countries are there institutions dealing with NIR. In addition to the two countries long involved, namely, the USA and the USSR, only less than twenty or so other countries in the world may have institutions which are concerned with the study of NIR. Of these, about half are European countries. Over twenty institutions have already identified themselves in the USA. They include the National Institute of Environmental Health Sciences and the Bureau of Radiological Health, both of the Department of Health, Education and Welfare, the Environmental Protection Agency, research centres of the three branches of the armed forces, various universities and a few industrial corporations.

Survey of Legislation. A second survey is now under preparation by the WHO Regional Office for Europe and is expected to bring forward, in a concentrated orderly form, the detailed legislation and the administrative regulations which govern the manufacturing, licensing, use and maintenance of NIR devices in the various countries related to employees, patients and the general public, at national, regional and local levels.

Study of Health Effects. As part of its discussions, the Working Group did, of course, review the possible health effects from specific NIR sources. However, a more elaborate study on this subject was considered necessary. Consequently, a report was drafted for the WHO Regional Office for Europe on the potential hazard to human health from the exposure to microwaves, lasers, and radiofrequency-type radiation. In this report attention has been given equally to work published in the USA, Western Europe, the USSR and Eastern Europe. This report is now under review by a number of well-known experts and will be discussed, together with the review comments received by future working groups, and eventually serve as an input for the Manual on NIR Protection.



Health Effects from Microwave Radiation. WHO, together with the US Department of Health, Education and Welfare, and the Polish Scientific Council to the Minister of Health and Social Welfare, are sponsoring the forthcoming International Symposium on Biologic Effects and Health Hazards from Microwave Radiation. This symposium which will meet in Warsaw, 15-18 November, 1973, will serve as the first international forum for the personal exchange of research information and professional experience. The symposium sessions will include papers on thermal and biologic effects, influence on the nervous system and behaviour, effects on the cellular and molecular level, measurements of the radiation and the biologic effects, occupational exposure and public health aspects.

The material submitted to this symposium and the conclusions to be drawn from its discussions will be the subject of a special evaluation meeting which should provide the feedback into the NIR sector of the programme of the European Regional Office. Moreover, it is anticipated that during this evaluation meeting the research gap and the need for complementary information on the study of microwave radiation will be identified, and possibly taken up through an expansion of the presently foreseen activities.

Health Effects from Laser Radiation. To study the health effects from lasers, a Working Group will be convened by the WHO Regional Office for Europe in May 1974. This meeting will discuss and make recommendations on hazards to the eye and skin, risks from the use of lasers for engineering, industry, medicine, research and education, dose measurement and analysis of results, development of international standards, needs for additional investigations, and legislation and administrative regulations. This Working Group will also finalize the draft chapter on lasers for the Manual on NIR Protection.

Manual on NIR Protection. All the printed material resulting from the work on NIR will be incorporated into one manual, which should then serve as a comprehensive reference volume. To ensure that the manual has the maximum impact at all levels of governmental administration, it will be divided into two parts. The first part, addressed to senior administrators (i.e. decision-makers and appropriators of funds) is a short part containing well-integrated summaries of the following chapters and emphasizing the points which should be taken into consideration when promoting a NIR protection programme, namely, principles, available methodology, approaches and possible alternatives. The second part, comprising of the detailed chapters themselves, will provide the background and detailed information on the respective subjects, references, present practice, etc. Although this manual is being prepared by the Regional Office for use in Europe, it could, of course, also be adapted for use by countries outside the European Region. In its final form the second part of the manual may contain the following chapters:

- Directory of institutions,
- Potential hazards to human health from exposure to ultraviolet radiation, lasers, microwaves, radiofrequency radiation and ultrasound,
- Health criteria and guides,
- Occupational risks and public health hazards,
- Model code of practice for the safe handling of NIR sources,
- Existing laws and regulations,
- Licensing of NIR sources and law enforcement measures.

## International Health Criteria and Protection Standards

At the conclusion of the United Nations conference on the Human Environment in Stockholm, 1972, the WHO was entrusted with the enormous task of developing international health criteria and standards. In a meeting in Geneva, the scope of such a programme and its priorities were discussed.<sup>2</sup> To enable the preparation of the criteria documents, among others on NIR, it was considered essential to first prepare critical reviews of the existing knowledge on health effects from microwaves (as part of the category of first priority) and from ultraviolet radiation, lasers and ultrasonic radiation (as part of the category of second priority).

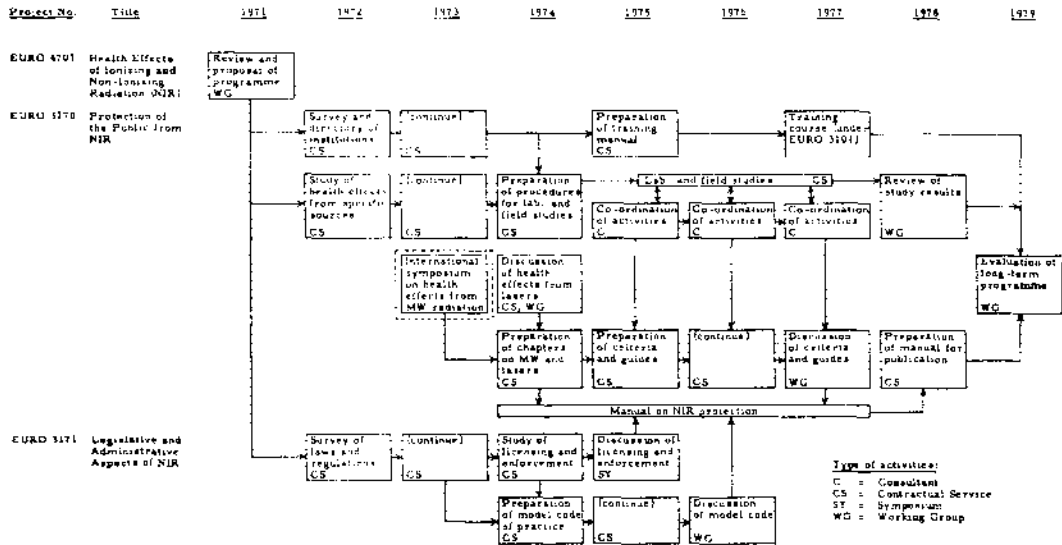
In view of the potential hazards connected with the use of emitters of NIR, especially whenever large amounts of energy are transmitted, it is felt that international rules for the use of these radiations are urgently needed. The rules should cover the construction, handling, licensing and maintenance of the equipment used for the production of the radiations. At present laws covering the protection of both workers and the general public against the different types of NIR exist only in the USA and USSR and are expected to be promulgated shortly in a few other countries. It is, however, evident that in most countries definite regulations concerning the manufacture of apparatus emitting NIR are deficient. It should be stated that before workable international rules for maximum exposure and regulations for manufacture can be set up, a number of conditions have to be fulfilled. Most important of these conditions is the adequate agreement on the level of exposure that represents the lowest hazardous dose to man. On the basis of this knowledge, it should be possible to formulate standards with a sufficiently large margin of safety which would then be the protection guidelines.

It is evident that the most general expression for the result of energy deposition after radiation is that of a thermal nature. It is however, very difficult to give in all cases a satisfactory description of the distribution of energy inside the body due to differences in absorption in different tissues the occurrence of interference or resonance in a single organ or at interfaces between tissues, and for other reasons. A principal reason for the lack of information is the lack of proper instruments for measurement at the biologically appropriate place. This problem is of great importance in all types of NIR, but is probably felt strongest in the study of microwave absorption. Therefore, in order to obtain the maximum amount of reliable information, it will be necessary to plan the necessary studies as multidisciplinary collaborative studies in which representatives from the following disciplines could participate: physicists and engineers well grounded in the theory of the types of radiation under study and in electronics; biologists experienced in disciplines such as physiology, biochemistry, pathology, genetics and biostatistics; and in some cases, knowledge of ophthalmology, behavioural science and human and veterinary medicine, and if possible, a biophysicist would be useful.

### References

1. World Health Organization Regional Office for Europe, Health Effects of Ionizing and Non-ionizing Radiation, Copenhagen 1972. (Report on a Working Group, The Hague, 15 - 17 November 1971, WHO document EURO 4701)
2. World Health Organization, The WHO Environmental Health Criteria Programme Geneva, 1973. (Report on a WHO meeting, Geneva, 20 - 24 November 1972, WHO document EP/73.1)

Figure 1: THE NIR SECTOR OF THE LONG-TERM PROGRAMME ON ENVIRONMENTAL POLLUTION CONTROL OF THE WHO REGIONAL OFFICE FOR EUROPE<sup>a</sup>



<sup>a</sup> This programme is periodically reviewed and as such subjected to changes as appropriate.

Figure 2. THE ACTIVITY COMPONENTS OF THE NIR SECTOR OF THE LONG-TERM PROGRAMME ON ENVIRONMENTAL POLLUTION CONTROL OF THE REGIONAL OFFICE FOR EUROPE

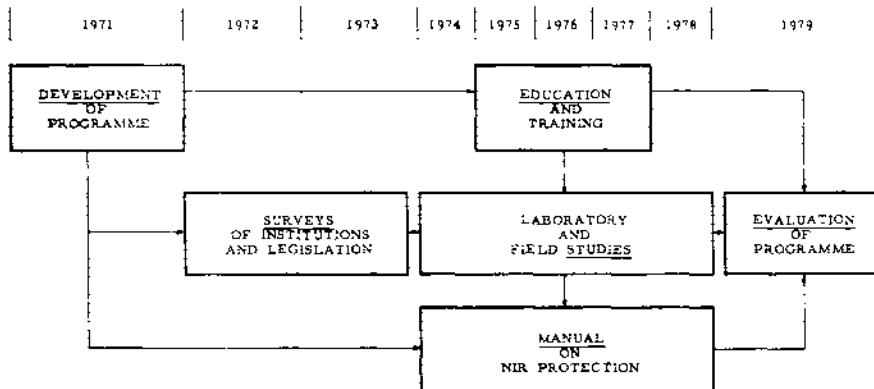


Table 1: CHARACTERISTICS AND SOURCES OF ELECTROMAGNETIC TYPE RADJATIONS

Type of radiations	Frequency range*	Wave-length range*	Energy range per photon	Typical source
Ionizing	above 30 000 THz	below 10 nm	above 124 eV	Electronic tubes, nuclear decay, nuclear fission
Ultraviolet	30 000 THz to 790 THz	10 nm to 380 nm	124 eV to 3.3 eV	Sun, gas discharge tubes
vacuum	3 000 THz to 1 600 THz	100 nm to 190 nm	12.4 eV to 6.5 eV	
far	1 600 THz to 1 000 THz	190 nm to 300 nm	6.5 eV to 4.1 eV	
near	1 000 THz to 790 THz	300 nm to 380 nm	4.1 eV to 3.3 eV	
non-ionizing portion	1 800 THz to 750 THz	170 nm to 400 nm	7.3 eV to 3.1 eV	
Visible	790 THz to 400 THz	380 nm to 750 nm	3.1 eV to 1.6 eV	Sun, thermally excited atoms
Infrared	400 THz to 300 GHz	750 nm to 1 mm	1.6 eV to 1.2 meV	Sun, hot bodies
near	400 THz to 100 THz	750 nm to 3 μm	1.6 eV to 0.4 eV	
middle	100 THz to 10 THz	3 μm to 30 μm	0.4 eV to 41 meV	
far	10 THz to 300 GHz	30 μm to 1 mm	41 meV to 1.2 meV	
Laser	1 500 THz to 15 THz	200 nm to 20 μm	6.2 eV to 62 meV	
Microwaves	300 GHz to 300 MHz	1 mm to 1 m	1.2 meV to 1.2 μeV	Klystron, Magnetron
EHF**	300 GHz to 30 GHz	1 mm to 10 mm	1.2 meV to 0.1 meV	
SHF**	30 GHz to 3 GHz	10 mm to 100 mm	0.1 meV to 12 μeV	
UHF**	3 GHz to 300 MHz	100 mm to 1 m	12 μeV to 1.2 μeV	
Radar	56 GHz to 220 MHz	5.4 mm to 1.3 m	0.2 meV to 0.9 μeV	
Radio-frequencies	300 MHz to 300 kHz	1 m to 1 km	1.2 μeV to 1.2 neV	Tubes, transistors and tuned circuits

\* The given ranges are only approximations, as no exact end-point can be defined,

\*\* Extremely high frequencies, Super-high frequencies, Ultra-high frequencies

Conclusions and Recommendations of a WHO Working Group on NIR\*Health Studies

(1) The knowledge of the noxious effects of NIR is scattered and incomplete. The Working Group felt that our knowledge concerning low level effects and the possibility of cumulative effects is very defective.

IT IS RECOMMENDED THAT the World Health Organization should organize the collection of case-histories of accidental exposure to all types of NIR in order to make it possible to give a better description of the clinical disorders to be expected after such exposure.

(2) With the growing use of NIR, the populations at risk will be increasing in size. Due to the continuous introduction of new procedures in which NIR is used, the populations at risk will also be changing both in size and distribution, which means that new groups may present previously unrecognized health problems.

IT IS RECOMMENDED THAT the World Health Organization should initiate appropriate surveys into the size and distribution of populations at risk, among both workers directly concerned and the general public.

(3) In view of the lack of quantitative information on possible late effects of NIR in man, further information is urgently needed.

IT IS RECOMMENDED THAT the World Health Organization should encourage and co-ordinate both prospective and retrospective long-term epidemiological studies on groups of workers exposed to NIR.

(4) When considering the available equipment, which generates NIR, the degree of its use and the potential hazard involved, research priorities for the study of health hazards from NIR must be given to lasers and microwaves.

IT IS RECOMMENDED THAT the World Health Organization should encourage such most urgently needed research.

Reference Centres

(5) In order to co-ordinate such studies and to correlate the findings and in general to act as observers in the field of NIR, centres of outstanding quality should be selected as international and national reference centres, both for the total field of NIR and wherever possible for the single types of radiation also.

IT IS RECOMMENDED THAT the World Health Organization should establish appropriate international reference centres to undertake these tasks, and encourage the setting up of similar centres at national level.

---

\* Taken from the report on a Working Group convened by the Regional Office for Europe of the World Health Organization, The Hague, 15 - 17 November 1972 <sup>1</sup>

#### Dose measurements and units

(6) For such studies, it is necessary to collect groups of exposed persons, for whom sufficient physical information on the exposure dose is available, in order to act as a basis for future epidemiological and other studies.

IT IS RECOMMENDED THAT the World Health Organization should encourage the development and use of measuring equipment for the registration of exposure to doses from NIR in exposed persons.

(7) It is felt that there is a need to extend the collaboration between centres active in health protection against NIR into the field of units and measurements also.

IT IS RECOMMENDED THAT the World Health Organization should initiate collaboration between such centres in order to harmonize the units used in dose measurements.

(8) It was evident from the discussions of the Working Group that the work of the International Commission on Radiological Protection and the International Commission on Radiation Units and Measurements, in introducing the concepts of normal man and of the maximum permissible dose and in classifying dose definition and dose measurements, has been fundamental in the control and protection of workers against ionizing radiation. In order to develop the understanding of the hazards of NIR, an organ with similar terms of references in the field of NIR is urgently needed.

IT IS RECOMMENDED THAT the World Health Organization should initiate the setting up of such an organ, either within its own organization or through international collaboration.

#### Safety guidelines and protective standards

(9) The use of NIR is rapidly expanding. The Working Group surveyed present safety codes and guidelines and found the guidelines in general sufficient to prevent injury under present conditions. It is doubtful, however, whether they will be adequate in the future, in view of the expected growth in the use of all types of NIR due not only to increased production of the types of equipment already in use but also to the constant introduction of new techniques using NIR.

IT IS RECOMMENDED THAT the World Health Organization should survey the field of health protection from NIR with the long-term goal of producing model codes of practice for this field, both for workers directly concerned and for the general public.

#### Education and licensing

(10) The number of apparatuses able to produce NIR is increasing rapidly in both industry and medicine. The number and types of such apparatus offered to the general public is also increasing. This increase in use requires technical personnel for construction, operation, maintenance and control. For all these groups, adequate training will be needed in the use of the equipment and in protection against the radiation emitted.

IT IS RECOMMENDED THAT the World Health Organization should conduct a survey of existing training programmes and courses on the use of NIR in order to ensure adequate international standards of education.

(11) To ensure the proper handling of equipment emitting NIR, a licensing system may be appropriate for manufacturers, engineers, physicians and technical personnel.

IT IS RECOMMENDED THAT the World Health Organization should investigate whether adequate licensing procedures for operation and maintenance are available and encourage the international formulation of the requirements for such licensing.

(12) The population at risk is that part of the general population which may be exposed either by the use of equipment emitting NIR or by accidental contact with the radiation emitted and is increasing with the growing use of such equipment. In order to minimize the risk involved, it will be necessary to educate the general public in the use of such equipment and inform them of the dangers which may be involved.

IT IS RECOMMENDED THAT the World Health Organization should encourage the dissemination of such information on the safe use and the hazards of NIR to the general public.

#### Ultraviolet radiation

(13) For the setting of proper standards for protection, an unambiguous dose-effect relationship for the production of skin cancer is urgently needed.

IT IS RECOMMENDED THAT the World Health Organization should encourage quantitative work on UV skin carcinogenesis.

(14) The evidence suggesting the possibility of cataract formation from UV radiation is not too convincing.

IT IS RECOMMENDED THAT the World Health Organization should encourage studies into possible cataract formation from UV radiation.

#### Infrared radiation

(15) The knowledge concerning possible long-term effects of IR radiation is inadequate.

IT IS RECOMMENDED THAT the World Health Organization should encourage studies into possible cataract formation and carcinogenesis from IR radiation.

(16) The significance of the effect of ambient climate on the possible damage from IR radiation is not yet very well understood.

IT IS RECOMMENDED THAT the World Health Organization should encourage studies into the relation between damage from IR radiation and the temperature, moisture and other environmental factors.

## Microwaves

(17) Agreement must be reached on how to establish common criteria for assessing the action of microwaves on man. Only this would make the accurate comparison of results of scientific research from different laboratories possible, and thus allow guidelines for the protection of health to be established.

IT IS RECOMMENDED THAT the World Health Organization should take steps to develop the relevant recommendations for criteria of microwave effects on man.

(18) The need for better personal dosimetry for the measurement of absorbed doses both by an integrating dosimeter and by a small implantable dosimeter for in vivo measurements in animals is felt very strongly in the case of microwaves.

IT IS RECOMMENDED THAT the World Health Organization should stimulate research on the development of accurate dosimeters for both near-field and far-field energy determination, including the development of implantable dosimeters.

## Lasers

(19) Insufficient knowledge is available for a complete description of the effect of laser radiation on the human eye.

IT IS RECOMMENDED THAT the World Health Organization should initiate a collaborative study on the effects of laser radiation on the human eye.

(20) Lasers are at present manufactured and sold in kits for home construction without appropriate licensing to purchasers, who may not understand the risks involved.

IT IS RECOMMENDED THAT the World Health Organization should take steps to introduce international standards for control of such sales.

## Ultrasound

(21) Although the Working Group knew of no reports of accidents or well-established damage arising from occupational exposure to ultrasound, the scientific evidence on this subject was felt to be inconclusive. It is uncertain at present whether adequate provisions exist for reporting occupational injuries in such a manner that ultrasonic exposure could be identified as a cause.

IT IS RECOMMENDED THAT the World Health Organization should encourage the systematic and meaningful reporting of such injuries.

(22) At present there is no knowledge of the acoustic intensity levels in human tissues resulting from occupational exposure to ultrasound, although adequate measuring techniques for this purpose exist.

IT IS RECOMMENDED THAT the World Health Organization should encourage the determining and reporting of such exposure levels in typical occupational situations.

(23) A conflict of opinion exists in the literature concerning the possibility that exposure to ultrasound could lead to biological changes of a mutational nature.

IT IS RECOMMENDED THAT the World Health Organization should encourage the critical scientific review of this topic and, if necessary, additional definitive experimental studies.

## Sound

(24) There are strong indications that sound levels, which are safe from the viewpoint of possible decrease of hearing level, may produce other somatic or functional effects or unfavourably influence the quality of sleep.

IT IS RECOMMENDED THAT the World Health Organization should promote international collaboration between the few institutes in the world active in this field.



## PITFALLS IN THE ASSESSMENT OF MICROWAVE RADIATION AS A HAZARD

Russell L. Carpenter, Esther S. Ferri and Gary J. Hegan. Northeastern Radiological Health Laboratory, 109 Holton St., Winchester, Mass. 01890

### ABSTRACT

The extent to which microwave radiation can constitute a health hazard is a question amenable to laboratory investigation, but the investigator must constantly be on guard against pitfalls peculiar to the investigation of this problem. Among them are: the reliability of power density measuring devices; near field and far field differences in field patterns and in perturbations of the microwave field by the experimental animal itself or by accessory supports or restrainers; whole body heating as a factor influencing results; determination of the relative roles of power density and duration of exposure as dose factors; and limitations on extrapolation to man of results from animal experiments. Attempts to define and to limit these problems will be described and illustrated.

. . . . .

The question of whether microwave radiation can constitute a health hazard is easily answered by placing an animal in a microwave field. With sufficient energy absorption and duration of exposure, the animal's body temperature will rapidly rise to a lethal level and it will expire, death being due to hyperthermia. Although this demonstrates that microwaves can be hazardous to the animal's health, the same result may also be obtained by placing it in the oven of an operative gas or electric range. The question we really wish to have answered is how much - or how little - microwave radiation can cause damage to living tissue. This question is one to which we have directed our efforts for several years, with the lens of the rabbit eye as the target tissue. Damage becomes visible in the lens as small opacities in its otherwise transparent tissue. In the course of our research, we have encountered a number of pitfalls, some of which I shall here briefly describe.

In a series of 136 experiments, 2450 MHz radiation was directed primarily upon the target by positioning the rabbit with its eye two inches distant from the dipole antenna of the radiation source. The head was thus in the near zone of the microwave field. For eight different power settings, we found the shortest duration of exposure which would cause a lens opacity to develop, and expressed this information graphically as a curve showing time and power thresholds for opacity induction.<sup>1</sup> This curve was similar to one obtained in like manner by Williams, et al.,<sup>2</sup> but differed with respect to values for field power. They calculated field energy from measurements made with a dipole antenna and a tunable bolometer detector. Ours were done calorimetrically, the calorimeter being a fluid-filled plastic sphere placed in the same position in the microwave field as was occupied by the eye during irradiation. The sphere was filled with a saline solution having a dielectric constant similar to that of the eye. Temperature changes reflecting energy absorbed

or lost were measured by a thermistor-bridge circuit, with the thermistor enclosed in the tip of a 24 gauge hypodermic needle inserted in the center of the sphere. Power density was then calculated employing the cross sectional area of the plastic sphere.

To assess microwave radiation as a hazard to the lens, it would have been most useful to know which of the two threshold curves represented the true state of affairs.

With the development of instruments for the direct measurement of power density in a microwave field, it appeared possible to re-evaluate our previous calorimetric measurements. Using a Narda Model 8110 Electromagnetic Radiation Survey Meter<sup>3</sup> and its Model 8122A probe having two crossed dipoles, we found that under identical conditions of geometry and power output, measurements were approximately 50 percent higher than when calculated from calorimetric measurements.

It should be noted at this time that the unit of  $\text{mW}/\text{cm}^2$  as applied to the calorimetric measurement is conventional. However, when using an electromagnetic survey meter and probe as a measuring device in the near field, a meter reading in  $\text{mW}/\text{cm}^2$  should not be considered a measure of the actual power density. The electromagnetic survey meter and probe measure, the electric field, and meter readings in the near field discussed in this paper are a measure of the electric field (E) and equal to  $E^2/377 \times 1000$ .

A few years later, it became possible to further evaluate the near field zone by means of an Electromagnetic Hazard Meter developed for the Bureau of Radiological Health by the National Bureau of Standards<sup>4</sup>. The probe of this instrument employs three crossed dipoles and, if desired, output of each can be read separately on the meter. This instrument gave readings which were an average of 34 percent higher than those of the Narda Model 8110 meter. This could be accounted for by the observation that there was a longitudinal and radial component of the field at the two-inch distance. This view was corroborated some months later when we acquired a Narda Model 8315A Broadband Electromagnetic Radiation Monitor<sup>5</sup> with its Model 8323 isotropic probe. Measurements made at the two-inch distance with this instrument averaged 41 percent greater than those made with the Narda Model 8110 instrument. Measurements with electric field sensors, such as the Narda and NBS probes, give an indication of the electric field strength; calorimetric devices measure absorbed energy. Therefore it would be inappropriate to compare measurements obtained by these different means.

There is a futility of attempting to define hazardous power levels on the basis of past reported experiments in the near zone field, if only because of the inability to measure the actual power density. The far field, on the other hand, exhibits a much more uniform and regular radiation pattern and permits a more reliable calculation of the power density from measurements of the E field.

One difficulty when performing experiments in the far field is that the entire body of the experimental subject is illuminated. The rabbit which, without anesthesia, will tolerate having its head subjected to a given exposure field for an hour in the near zone will strenuously seek escape after 15 minutes or will succumb from exposure to whole body radiation only one third of that tolerated in the near field.

Still another pitfall lies in the perturbation of the radiation field by the presence of the experimental subject itself. At a distance of 150 cm the field pattern of our standard gain horn at 2450 MHz is quite

uniform in power density, being highest in the center of the field and falling off in a gradual manner along the x and y axes so that 50 cm from the center, it is reduced by 75 percent. However, a rabbit sitting quietly in the field perturbs the field in such an irregular manner that the power density may be either increased or decreased by as much as 50 percent in some areas. For example, in one instance we have found that the presence of the rabbit appears to reduce the power density at a location of the rabbit's eye by about 50 percent. Inasmuch as the pattern of field perturbation depends in part on the geometry of the perturbing factor, it is not surprising that there is a difference in perturbation of the field when the rabbit's ears are held erect or are down flat against the body. We have found that the eye is subjected to less radiation when the animal's ears are down than when they are up. Perturbation of the field also occurs from the presence of such experimental accessories as plastic cages or animal restraints.

The relationship of this perturbed exposure field to an absorbed dose may be difficult to determine. However, a total absorbed dose would depend on a complex relationship between the exposure field (the magnitude, direction, and phase of the electric field at all points on the surface of the object), the dielectric constant or constants, geometry and surrounding media of the object of exposure.

#### REFERENCES

1. R. L. Carpenter, D. K. Biddle, and C. A. Van Ummersen: "Biological effects of microwave radiation, with particular references to the eye." Proc. Third Int. Conf. on Med. Electronics, London, pp. 404-408. (1960)
2. D. B. Williams, J. P. Monohan, W. J. Nicholson, and J. J. Aldrich: "Biologic effects of microwave radiation: Time and power thresholds for the production of lens opacities by 12.3 cm. microwaves." U.S.A.F. School of Aviation Medicine Report No. 55-94. (1955).
3. E. Aslan: "Electromagnetic leakage survey meter." Jour. Mic. Power, Vol. 6, No. 2, pp. 169-178. (1971)
4. R. R. Bowman: "Quantifying hazardous electromagnetic fields: Practical considerations." Biol. Effects and Health Implications of Microwave Radiation, Symposium Proceedings, Richmond, Va. (BRM/DBE-70-2) pp. 204-209 (1969).
5. E. Aslan: "Broadband isotropic electromagnetic radiation monitor." IEEE Trans. on Instrumentation and Measurements, Vol. IM-21, No. 4, pp. 421-424. (1972)
6. R. L. Carpenter and C. A. Van Ummersen: "The action of microwave power on the eye." Jour. Mic. Power, Vol. 3, No. 1, pp. 3 - 19. (1968)

## CONTROL AND USE OF LASERS IN N.Y.S. INDUSTRY

F. J. Bradley and S. N. Roberto  
Division of Industrial Hygiene  
N.Y.S. Department of Labor  
80 Centre Street  
New York, New York 10013

### Introduction

#### General

Since the important and historic paper of Solon, Aronson & Gould<sup>1</sup> pinpointed the potential for eye damage from lasers, it has been recognized that the eye is the critical body organ for laser radiation exposure in the visible and near-infrared. Experimental data by Ham, and others have confirmed the early predictions of eye damage and that eye exposure especially to the retina is a limiting factor in working around lasers<sup>2</sup>. Damage of the retina is usually irreversible and since the eye is crucial for an individual's well-being extreme care is indicated to ensure that such damage does not occur. An additional inducement for care in laser radiation exposure is the fact that maximum permissible exposure values are based on acute damage as evidenced by standardized objective tests on experimental animals. Little human data and fortunately no epidemiological data are available to confirm the suggested maximum permissible exposure limits. Very little data is available as a result of long term exposures at sub-acute levels of laser radiation and none to date have been incorporated except through safety factors into maximum permissible exposure limits.

Laser radiation that is not amplified by the optics of the eye causes surface damage either to the cornea or skin. Sufficient biological data is available to set corneal maximum permissible exposure limits for infrared radiation in the wave length range above 1400 nm and skin maximum permissible exposure limits in the visible and infrared wave length range above 400 nm. Insufficient data is available to set corneal or skin maximum permissible exposure limits in the laser UV region. Most of the skin damage data is based on short term exposures and little information is available on what might result from long term exposures.

To ensure that the benefits of laser radiation will be reaped with little human eye or skin damage New York State has promulgated Industrial Code Rule 50, Lasers, to regulate the industrial use of lasers in the State. The Code Rule is generally a performance-type Code but some specification type regulations for certain types of lasers and certain laser working conditions are incorporated.

The Code Rule separates lasers into three categories based on their emission characteristics. These are exempt, low intensity and high intensity lasers. There is an additional classification of the low and high intensity categories into fixed and mobile lasers. The rest of this paper expands on the control measures specified in Industrial Code Rule 50 and the industrial use of lasers in the State.

## Control of Lasers

### Exempt Lasers

There are two conditions which make a laser exempt from the provisions of Code Rule 50. Exemption #1 covers storage, shipment or sale of inoperable lasers. Exemption #2 covers the case of lasers which by reason of their design and construction cannot emit radiation that exceeds  $1 \times 10^{-7}$  J/cm<sup>2</sup> or  $1 \times 10^{-5}$  W/cm<sup>2</sup> when measured at 10 centimeters from the exterior surface of the laser.

### Low Intensity Lasers

Approvals. Low intensity lasers have an emission level which exceeds the above values for exempt lasers but do not exceed  $1 \times 10^{-1}$  J/cm<sup>2</sup> for 1 nsec to 1  $\mu$ sec pulse, 1 J/cm<sup>2</sup> for 1  $\mu$ sec to 0.1 sec pulse and 3 W/cm<sup>2</sup> for CW or repetitively pulsed lasers. The values are approximately set at the values where skin burns for visible and infrared electromagnetic radiation start to become a factor in control measures. Low intensity lasers other than those in research and development status will need Board of Standards and Appeals approval prior to distribution and use but with approval the laser installation will not need to register.

Mobile Lasers. Code Rule 50 places special requirements on mobile lasers - lasers used outside fixed installations. Such lasers have additional potential for causing injury to the laser worker and the general public and also have a high nuisance potential if not properly controlled. Because of these factors the Code requires that individuals using mobile lasers must be certified by the Industrial Commissioner. To assist him in implementing this provision the commissioner has appointed a Laser Examining Board whose functions are (a) examination of applicants and their experience and make recommendations thereon, (b) holding of hearings on denials, (c) holding of hearings on suspension or revocation of certificate, and (d) reporting findings and recommendations to the commissioner. There are two categories of certificates of competence. Class A certificate of competence for operators of low intensity mobile lasers and Class B certificate of competence for operators of high and low intensity mobile lasers.

### High Intensity Lasers

Fixed Laser Installation and Mobile Laser. All such installations and mobile laser operations utilizing high intensity lasers must register with the commissioner and designate a laser safety officer. In addition prior notification is required for all field work with mobile high intensity lasers. In case of fixed installation a laser radiation area must be designated and posted with the standard laser hazard symbol. Special precautions that may be necessary are (a) remote viewing apparatus, (b) special termination materials for high intensity beams, (c) interlocks on equipment and doors to laser radiation areas, and (d) "fail safe" electronic circuitry wherever it is warranted. Because of its importance in determining the extent of the outdoor laser radiation area a survey is required of the output power or energy density of high intensity mobile lasers.

Each laser shall be safeguarded against unauthorized use and no person shall dispose of a high intensity laser except by making it permanently inoperative or by transferring to another person authorized to receive it.

Each person who possesses a laser shall report (a) any theft or loss of intact laser, (b) any injury to individual resulting from operation of laser or associated equipment.

Personal Protection. It is obvious that persons using lasers must be adequately instructed in the safe use of the laser. One must never look directly into a laser beam above the maximum permissible exposure limits without adequate eye protection. Code Rule 50 recommends the use of approved safety eyewear by those individuals who may be exposed to laser radiation above the maximum permissible exposure limits. Minimum standards for such eyewear are (a) adequate optical density to reduce laser radiation to safe levels, (b) designed and tested to insure that eyewear retains its protective properties during use, (c) legibly labeled with the optical density of the lens and wave length at which it was measured. In the case of a high intensity laser protective eyewear should not be relied on as the primary protective barrier between an individual and the laser beam. Protective eyewear should be primarily for accidental exposures and the laser application should be designed to ensure that the probability of eye exposure is minimal. Reliance on protective eyewear is a poor substitute for adequate control measures such as enclosures or remote viewing equipment. Furthermore in today's laser world protective eyewear can be very tricky because of the large number of potential laser wavelengths which one may encounter.

Associated Hazards. At least 2 deaths from electrical shock have been reported in the literature around lasers. The electrical hazard is especially great in research and development work and therefore all laser equipment must be designed, constructed, installed and maintained so as to minimize the possibility of electrical hazards.

With high intensity laser beams adequate protection must be provided for air contamination arising from vaporized target materials, toxic gases, vapors and fumes. Two areas of special concern are vaporized fire brick which may contain beryllium, and UV laser beams and UV pumping lamps emitting radiation in the wavelength range from 185-210 nm which will produce ozone from oxygen in the atmosphere.

Besides adequate radiation shielding an explosion shield around the resonating laser cavity is recommended where explosions of the lasing medium are possible.

Every laser and laser installation must be designed, installed, operated and maintained to eliminate or reduce any fire hazard.

The only ionizing radiation hazard associated with lasers at present is in the high voltage power supplies and only gross ignorance or carelessness will lead to exposure from this source in this day and age. X-ray or neutron radiation hazard associated with the laser beam or its target interaction lie in the future.

## Future

Scanning Laser Beams. Lasers operating in the scanning mode have assumed commercial importance within the last 3 years. Code Rule 50 stipulates that the laser exposure values can be determined while the beam is in the scanning mode. Exposures therefore are equivalent to a repetitively pulsed laser beam. While it appears logical that the human eye will respond to such exposures in a manner similar to a pulsed laser, no experimental evidence is available on such laser beam exposures to ensure that these are appropriate maximum permissible exposure limits. Further experimental data is needed in this area.

UV and Subnanosecond Laser Pulses. Additional biological data is also urgently needed in the UV region so that maximum permissible limits can be established for corneal and skin exposure.

Pulse width is also a factor in the biological response and data is needed to establish maximum permissible limits for subnanosec laser pulses.

## Use of Lasers

### Construction Industry (SIC #15, 16, 17)

The largest number of individuals potentially exposed to laser radiation are in the construction industry which has approximately 51% of the registrants. The main application is a rather prosaic one involving the establishment of a reference line or plane. Basic surveying tools used by the construction industry have changed little since ancient Egyptian times until the advent of the laser. Stakes, rods and tapes have served for engineering works from Stonehenge to the Empire State building. But now the laser has revolutionized surveying. It can provide a reference line or plane at any desirable angle. In addition distance measurement can be accomplished in seconds with a high degree of accuracy.

In the construction industry the laser is used to (a) provide line and grade in laying of pipe, mainly storm and sewer pipe, (b) provide line and grade in heavy construction projects, such as, tunneling, erection of dams and dredging, (c) provide horizontal plane for installation of floors and ceilings and (d) distance measurement.

All of the lasers which we have encountered in this application are low intensity HeNe lasers. From a sample of 114 lasers the average reported power density was 27 mW/cm<sup>2</sup> with a range from 3 to 160 mW/cm<sup>2</sup>. This classification has 12% of the lasers and 4.1% of the laser workers. (See Figure 1.)

### Instrument, Photographic and Electro-Optical Industries (SIC #38)

This industrial classification has 19% of the registrants but 47% of the lasers and 32% of the laser workers. Lasers are incorporated in instruments, such as, surveying instruments for use by the construction industry as described above. They are used in the holographic studies. High intensity lasers are used in the manufacture of balance wheels and scribing.

### General Manufacturing including Computers (SIC #35)

There are 5% of the laser registrants in this industrial classification with 17% of the lasers and 10% of the laser workers. High intensity lasers, such as, CO<sub>2</sub>, Nd (YAG or glass) and ruby lasers, are used for drilling, welding, melting, burning and micro machining. Low intensity lasers are used for precision measurements, alignment and as a light source.

### Research and Development (SIC #73)

This service category has 3% of the registrations, 11% of the lasers and 5% of the laser workers. The main laser and application are the HeNe low intensity lasers used for alignment.

### Aerospace Industry (SIC #37)

This classification has 2% of the registrations, 8% of the lasers and 5% of the laser workers. High intensity CO<sub>2</sub>, Nd (YAG or glass) and ruby lasers are used for scribing, drilling and metal cutting mainly titanium alloy. Low intensity HeNe lasers are used for alignment of various airplane components onto the airframe.

### Electrical Machinery and Electronics (SIC #36)

There are 10% of the registrations, 3% of the lasers and 4% of the laser workers in this industrial classification. High intensity Nd laser is used for resistor trimming, CO<sub>2</sub> laser for glass cutting and contact cleaner and ruby laser for drilling diamond dies. Low intensity HeNe lasers are used for alignment and smoke detection.

### Miscellaneous Industries

Finally the remaining registrants fall in various industrial classifications, including jewelry, pharmaceutical and mining. The number of registrants is 10% of the total number of registrants, 2% of the lasers and 3% of the laser workers. High intensity Nd lasers are used by the jewelry industry for diamond drilling to improve their quality. High intensity ruby lasers are used in biological research by pharmaceutical industry. HeNe lasers are used by mining industry for grade and alignment and by surveyors for geodetic distance measurements. A high intensity argon laser is used in research on determination of air and water contaminants.

### Recommendations

With several national standard setting bodies establishing maximum permissible exposure limits for laser radiation, many of them different in one way or another, the time is appropriate for an international body (possibly this Association) to take the initiative in formulating Basic Laser Radiation Exposure Guidelines. Expanding interchange of men and goods make it desirable that such guidelines be established to ensure maximum protection of the working population as well as the general public.



References

- <sup>1</sup> Solon, L. R., R. Aronson & G. Gould, Physiological implications of laser beams, *Science* 134, 1506-1508 (1961).
- <sup>2</sup> Ham, W. T., Jr., R. C. Williams, E. A. Mueller, R. S. Ruffin, F. H. Schmidt, A. M. Clarke, J. J. Vos & W. J. Coeraets, Ocular effects of laser radiation, *Acta Ophthalmologica* 43, 390-409 (1965).

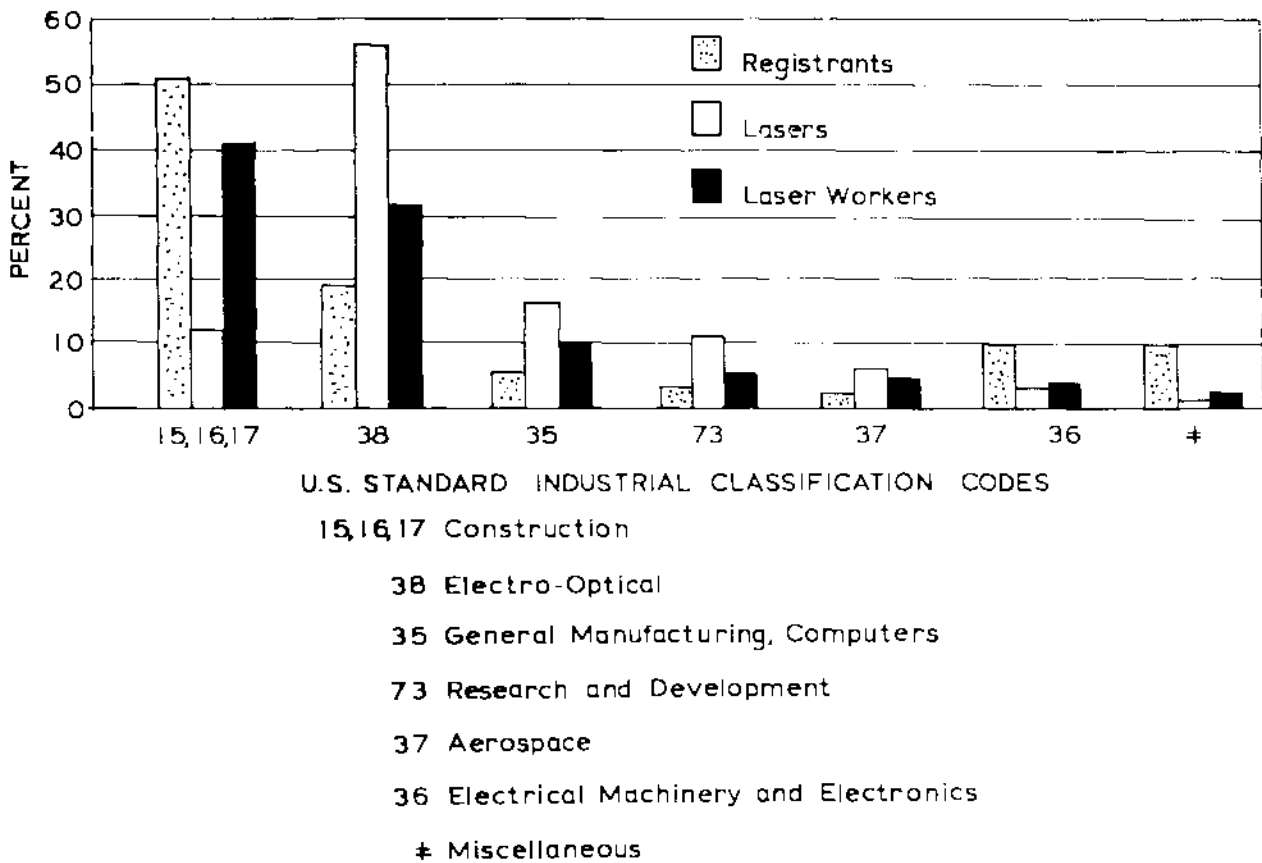


Figure 1- Registration, Laser and Laser Worker Percentages in Selected U.S. Standard Industrial Classifications as of January 1973

COMBINED EFFECT OF IONIZING RADIATION AND A SUPERHIGH-FREQUENCY FIELD ON THE BODY

A.N.Liberman, I.E.Bronstein, V.V.Orobei, M.S.Sakovskaia, A.P.Tchesnokova, V.M.Shubik, R.I.Bikkulov

Radiation Hygiene Institute

The RSFSR Ministry of Public Health

Leningrad

USSR

Abstract

Prolonged investigations of white mice and white rats showed fractional combined X-ray longwave (30 r of single exposure) and SHF (5 mw/cm<sup>2</sup>, 20 min) irradiation to be responsible for more pronounced effects, than those caused by these factors, acting independently. The differences detected were in body and some other organs weight, in lisozimes liters of serum, in productibility of animals and in chromosomal aberrations number in bone marrow.

At combined X-ray, short-wave and SHF-irradiation the intensification of effect was not found.

Observations showed that combined occupational effect of X-rays of different energy and SHF irradiations caused more frequency deviations of organism functional state than SHF field irradiation only.

Introduction

Under conditions of occupation a man is subjected not to single but to several environment factors. Adequate estimation of the possible consequences would account for every factor under consideration. The peculiarities of the combined effect have to be considered also.

The necessity of expansion of investigations in the field of radiation protection, in direction of combined effects study, was pointed out in the I European Congress of Radiation Protection (Menton, 1969) and in the II International Congress of Radiation Protection (Brighton, 1970).

One of the most widespread variants of combined professional effects is combination of roentgen and high frequency (SHF) electromagnetic irradiations<sup>1,2,3</sup>.

However, till recently the investigations of combined action of X-ray and SHF radiations were represented in a few papers<sup>4,5,6</sup>. In these papers the effects of strike momentary combined action were mainly considered.

The results of experiments with the laboratory animals which have been performed during 1967-72 years and the results of clinical-physiology observations of persons during 1962-72 are presented in this paper. The purpose of this work was to study the effects of prolonged combined and isolated X-ray and SHF irradiations.

## I. The Experiments with Animals

### Materials and Methods

The experiments have been carried out with white female mice and white female rats. Mice and rats weighed 13-20 g and 180 - 200 g respectively at the beginning of experiments.

The longwave X-radiation was provided by 60 kV X-ray generator operated at 15.5 kV with 0.17 mm Al and 1.2 mm perspex filters, HVL 0.1 mm Al. The exposure rate was 10.5 r/min. The animals were irradiated from the ventral side of body.

The shortwave X-radiation was provided by 200 kV X-ray generator operating at 137 kV with 0.5 mm Cu and 1.0 mm Al filters, HVL 0.83 mm Cu. The exposure rate was 24.5 r/min.

Nonscattering chamber with SHF radiation generator "Looch-58" (wave length 12.6 cm) was used for continued exposure of animals body ventral side.

The investigations with animals in all series of experiments were performed simultaneously in control groups (unaffected) and in these subjected to X-rays, SHF and combined irradiations. The interval between two consequent exposures was equal to 48 hours. The intervals between X-ray and SHF irradiations were equal 5 - 15 min.

The following tests were used for estimation of the biological action of radiations: survival and average longevity, weight of body and of some organs, stability to the physical loading, productivity and reproduction factors, weight of the month age posterity, the counts of chromosome aberrations in cells of bone marrow the counts of blood cells. Stability of animals to physical loading was determined with our early elaborated method<sup>7</sup>

The complex of immunologic reactions describing a state of nonspecific, specific immunity, allergic and autoallergic reactivity was studied. The factors of nonspecific immunity were bactericidity, titers of lysozyme and complement of serum. Specific immunity was studied after immunization of animals with sheep's erythrocytes. Production of hemagglutinins and of hemolysins were studied. In animals inverse anaphylactic reaction (by Joffe) and complement-fixing autoantibody formation were studied too.

### Results of Experiments

Influence upon the fractional shortwave X-rays at 60 r for a single exposure combined with fractional SHF-radiation at 40 mw/cm<sup>2</sup> delivered during 15 and 18 minutes led to death of half quantity of the animals in more early stages than it occurred from X-ray irradiation only. X-irradiation became a main factor with SHF exposure being reduced up to 11 minutes. In this case the decrease of body weight of mouse was 2.4-2.8 g, intermediate between that of mouse, subjected to each factors taken separately.

In the following series of experiments X-ray single exposure have been reduced up to 30 r and SHF-radiation power density to 6 mw/cm<sup>2</sup> for 30 minutes. Under these conditions decreasing longevity and body-weight of the animals had few differences in groups of X-ray and combined exposure. Most of animals died after 3 months from experiments beginning. SHF irradiation alone caused death of animals not exceeding 15 per cent. After 100 days observation the body weight of the animals was found rather sharply and statistically reliably decreased for the animals exposed to X-rays and in particular to combined radiation. The body weight

of mice exposed to SHF radiation later was close to control level.

Study of the peripheral blood cells composition of rats subjected to SHF irradiation was performed at 50 mw/cm<sup>2</sup> and 10 mw/cm<sup>2</sup> for 20 min and shortwave X-ray irradiation with single exposure 60 r. Whole exposure duration was 45 days. Statistically reliable leucopenia was registered in animals subjected X-ray and comine radiation. Of great interest is great dispersion of leucocytes number of animals exposed to SHF only and combined radiation. However, in the latter case this dispersion was notably decreased with X-ray dose accumulation.

Significant changes of immunologic reactivity of rats exposed to fractionated SHF radiation at 50 mw/cm<sup>2</sup> for 20 min of single exposure have not been resistered. But short-time decrease of lisozyme titer, some intensification of complemental activity of serum, phase variations of antibody formation and excessive creation of complement-fixing autoantibody were found to occur. At the same time shortwave X-ray exposure at 60 r led to conspicuous suppression of all factors studied of nonspecific and specific immunity, decrease of total immunological reactivity and rise of antibody formation. Combined influence was responsible for some decrease of radiation breaking of nonspecific immunity as well as for oppression of serum bacterioidity. Decrease of serum lisozyme titer was less conspicuous than at X-ray irradiation only.

Shortwave X-ray radiation was established to influence mainly upon mice stability to physical loading at the levels of combined short X-ray and SHF irradiation chosen by us. However, decrease of animal stability to the physical loading occurred earlier (on the 41-st day) at combined radiation than at X-ray irradiation only.

Under fractionated irradiation conditions the experimental animals revealed the considerable alterations of spermatogenesis in all stages, in particular, at shortwave X-ray and at combined irradiation. The earliest alterations were noticed in the initial stages of spermatogenesis. So, at some stages of investigations spermatogonia, spermatocytes, spermatids and spermatozoa were absent completely or almost completely and testis bubules were devastated.

Thus, the investigations of the animals reactions to influence of fractionated shortwave X-ray and SHF irradiations did not allow to find clear intensification of effects at combined irradiation. Predominant affect of one of those factors was noted depending on their levels.

When studying the longwave X-ray (30 r dose of single exposure) and SHF (5 mw/cm<sup>2</sup>, 20 min for single exposure) radiation influence, fractional irradiation of mice in different series of tests lasted from 2.5 to 6 months.

The X-ray longwave irradiation (by the dose mentioned) have never resulted in animals' weight decrease during the investigations. As a result of SHF irradiation animals' weight had reliable decrease only on the 110-th day from the beginning of irradiation. Combined irradiation being used, beginning from the 15-th day of irradiation the mice weight have been decreased in comparison with that of unaffected (control) group.

The obtained data of one-factor disperse analysis showed that in spite of negligible influence of isolated X-ray irradiation on the body weight, with combined irradiation it becomes essential.

A tendency have been observed for weight increase of mouse's liver and spleen, when subjected to combined irradiation, pro-

bably because of intense blood filling.

As a result of combination of mentioned irradiation of two kinds decrease of serum lisozime titers in comparison with unaffected group of animals and those under separate irradiation was detected.

In three series of experiments the factors of productivity of mice were studied. In the first two series mice (both males and females) received 31 exposures. In the third series they received 32 seances of exposure. For the last 15 days before the end of irradiation male and female mice were in the same cages (15 female and 5 male mice in each cage). After this male mice were separated from female ones.

The following parameters were determined as the factors of productivity : per cent of delivering females reproducibility factors (per one female under investigation)  $k_1$  - according to the number of mice born,  $k_2$  - according to the number of mice born, which lived up to 30<sup>2</sup> days age.

The series of tests showed some increase in the number of delivering female mice and in the reproduction coefficients in groups of animals exposed to separate irradiation. There were no pronounced differences between these and control groups in the second series of tests (Table 1). On the contrary, the combined irradiation showed the statistically reliable (in comparison with control) decrease of delivering female per cent and marked (30-40 per cent) decrease of reproducibility factors.

Statistical processing using criteria of Wilcoxon showed reliable difference in the values of reproducibility factors for the groups exposed to combined irradiation in comparison with those unirradiated and mostly with the groups exposed to isolated X-ray or SHF irradiation. The chromosome aberration frequency for all groups of animals under investigation exposed for 2-5 months had notable increase in comparison with control. The maximum value of this factor exceeding 2.4 and 10 times the number of spontaneous aberration was obtained for the groups exposed to combined irradiation.

Thus, experimental study made it possible to derive pronounced effect of combined fractional X-ray longwave and SHF irradiations in comparison with isolated irradiation by a number of essential parameters.

## II. Person Observations

### Organization and Method

For examination and observation of the workers the latter were chosen engaged in the field of adjustment and testing of the electronic devices. These people were divided into the following 3 main groups according to forms of occupational factors:

I group - 50 persons working under conditions of combined X-ray longwave and SHF irradiation;

II group - 47 person working under conditions of combined X-ray shortwave and SHF irradiation;

III group - 50 persons working under conditions of SHF irradiation.

The central group (55 persons) included those who work under conditions with no professional irradiation.

The average age of every groupe person examined was 38-39 years for men and 35-38 years for women. Industrial record was

13-18 and 12-15 years respectively. The average age and average term of work of persons dealing with X-ray and SHF sources as well as persons in the control groups were similar.

The program of medical observations and clinical physiological examinations of the workers' groups consisted of investigations: functional conditions of neural and cardiovascular systems; state of blood, state of organs of vision and analysis of illness and complaints.

### Observation Results

Among workers of the first 3 groups complaints of asthenic-vegetative type prevailed, namely: abnormal high tiredness, sleep disturbances, head ache. The number of those complaints in the I and II groups (combined irradiation) reliably exceeded that in the III group (1.16-1.5 times). In the first two groups the number of second medical admices exceeded 2-4 times the same factor for other groups. It was established by means of objective investigation of persons who worked under combined irradiation conditions that deviations in neural system functional state were observed more often than for those who was exposed to SHF irradiation only, and to control group. For example the number of cases of hands fingers and lashes termor in the first two groups notably exceeded the same factor in the 3-rd and control groups.

E.E.G. examination showed that the frequency of deviations in these curves in the I and the II groups exceeded ones in the III-rd and control groups.

Arterial blood pressure levels in all groups under investigation were within the limits of normal values. Only light decrease of maximal arterial pressure was noticed for the I group persons examined in the first stage of observations.

E.C.G. - examinations made it possible to find notable increase of the number of cases with sinus arrhythmia and conduction disturbances for those who worked under combined irradiation.

As a result of capillaroscopy it was found that the frequency of spastic-atonical capillar state detections in the I, II, III and IV groups in equal to 62, 53, 47 and 25 per cent respectively, having statistically reliable differences between the first two groups and the III one as well as unaffected group.

While studying the capillar resistance using Nesterov method, the second (higher) reaction degree appeared reliable more often for persons worked in the conditions of combined irradiation than for ones of the III-rd and control groups.

Those who were exposed to combined irradiation had more frequent occasions of decrease of leucocytes number (less than 4200 in 1 mm<sup>3</sup> of blood) and of trombocytes number (less than 200 thousand in 1 mm<sup>3</sup> of blood) comparing with unirradiated groups; the average is not different from normal.

As a result of investigations of the vision organ state it was found that the frequency of cases of vascular dystonia of eye's bottom in the main groups of persons was not large (of 10-18%) still it had some deviations from the control group (6-10%).

While biomicroscopical studying the transparent eye's medium the lenticular opacity was observed more often in the I and II groups than in the control one. This opacity didn't affect the sharpness of sight in general.

Some immunological deviations were observed in the I group

of persons in comparison with those of the control group, namely, pronounced variation of skin microbes, increasing of C-reactive protein and serum autoantibody formation.

Comparing the disease incidents in the irradiated and in the control groups no essential differences between them were found.

No cases of occupational pathology caused by X-ray or SHF irradiation were observed for all period of investigations.

Thus, according to the most indices of functional state of neural and cardiovascular systems as well as to indices of humoral nonspecific immunity notable effects of X-ray longwave or shortwave irradiation combined with that of SHF were found for workers.

These differences are revealed in the increase of deviation frequency in comparison with the control group as well as with group of workers who were irradiated by SHF only.

Numerous medical observation data showed that the main contribution to functional variations occurrence is the result of occupational factors affects, which took place in the first period of work (before 1960-1961).

In the main groups in 1970 it was found that the degree of the same earlier observed deviations had notably decreased and in general the factors of functional body state did not lie beyond the limits of physiological variations.

### Conclusion

The following effects for animals were found as a result of experiments performed. Pronounced intensification of biological effects were observed for the case of combined SHF and X-ray longwave irradiations. Combined SHF and X-ray shortwave irradiations didn't result in any effects intensification for most of the factors under control.

X-ray irradiation was found to be dominant for changes observed during combined irradiations.

Clinic-physiological observations of the persons occupied in the conditions of combined X-ray and SHF irradiations showed higher frequency of investigated factors deviations than that of those occupied in conditions of SHF irradiation only and for the control (unaffected) group.

### References

1. Yu. A. Osipov, *Gigiena truda i vliyanie na rabotayuschikh elektromagnitnykh voln radiochastot*, Medgiz, Moskva (1965).
2. Yu. A. Osipov et al., *Gigiena i sanitariya*, 6, 35 (1963).
3. A. N. Liberman et al., In: *Trudy po radiatsionnoi gigiene*, 3, (1967)
4. S. M. Michaelson et al., *Aerospace Med.*, 35, 9 (1964).
5. K. N. Klyachina et al., In: *Gigiena truda i biologicheskoe deistvie elektromagnitnykh polei radiochastot*, Ac. med. sci. USSR, Moskva (1963).
6. K. N. Nikonova, In: *Gigiena truda i biologicheskoe deistvie elektromagnitnykh polei radiochastot*, Ac. med. Sci. USSR, Moskva (1968).
7. A. N. Liberman, M. S. Sakovskaya, *Radiobiologia*, 7, 4, 625 (1967).
8. M. A. Nevstrueva et al., *Vliyanie inkorporirovannykh radioizotopov na immunologicheskie protsessy*, Atomizdat, Moskva (1972).
9. V. M. Shubik, *Zhurnal med. i eksp. immunologii*, 9, 12 (1972).

PUBLIC HEALTH AND CONTROL  
OF  
NON-IONIZING RADIATION IN JAPAN

Hiroyoshi Iida and Tokusaburo Koshijima  
National Institute of Radiological Sciences  
Chiba-shi, Japan

Abstract

Some effective measures against a certain laser radiation is reported and the general situation in Japan concerning the protection of people against non-ionizing radiations is described.

Studies made on the possible protective measures against the harmful effect of laser radiation, particularly on the eye and skin, revealed that the following three methods are effective against He-Ne laser radiation of 6,328 Å, 1) absorption by cellophane sheet, 2) absorption by filter and 3) reflection by non-metallic multilayer film.

A number of studies on biological effect of microwave radiation and laser radiation have been conducted in Japan. There are, however, no regulations on the permissible level of these radiations such as those set forth against ionizing radiations on human body. Recently, home microwave ovens are pervading rapidly. In view of these situations, the "Research Committee on Biological Effect of Non-ionizing Radiation from Home Use Instruments" was organized in the Ministry of Health and Welfare last year.

Currently, regulations are applied to the manufacturers of these devices by the Ordinance of Ministry of International Trade and Industry.

Introduction

The results of investigation on protection against laser radiation are described, together with an introduction to the present situation of public health control against non-ionizing radiations in Japan.

The use of laser radiation has been growing in physics research, telecommunication, metal processing, cancer treatment and etc., by which are required higher energy and peak power of laser and hence the increasing problems of protection. Apparently, the laser radiation hazard is caused mainly by heat absorbed, but the true mechanism is not yet revealed. The eye is the tissue of the most interest to be protected against laser radiation followed by the skin.

Radars, which are essential to navigation safety, aerial control, meteorology, naval and military affairs and etc., radiate electromagnetic wave characteristic to their purpose. Recently, home microwave ovens are pervading rapidly, hence



microwave radiation came into contact with the public. The biological effects of microwave radiation are explained mostly by thermal effect.

### Protection against lasers

Since laser beam is directional and its energy can be converged in a very small area, heat of very high intensity is absorbed in a small region of the tissue and burns it.

To protect against laser radiation, following methods are available, i.e., (I) to use material which absorbs laser energy, (II) to use material which reflects laser radiation. Both methods are possibly as far as the materials used do not burn out. (III) Shielding around the laser path is necessary to protect high energy or pulsed laser.

The laser beam used in the present study was the radiation of 6,328 Å wavelength continuously generated from the 10mW He-Ne equipment (Nippon Kagaku Kogyo). Absorption or reflection of the laser beam by a number of substances was examined by the use of photometer and illuminometer together with other spectroscopic analysis. Absorption by commercially available cellophane sheets is shown in Table 1. Spectrophotometric analysis is shown in Fig. 1. Cellophane sheets of green or purple provides effective protection against He-Ne 6,328 Å laser radiation. Absorption by glass filters (Toshiba) is measured as shown in Table 2. From the view-point of absorption coefficient, glass filters are far inferior to cellophane. In general, the absorption by glass filter is effective due to its thickness.

Reflectance was measured of one type of mirror of non-metallic multilayer film (Vacuum Optics Corporation of Japan, Fig. 2) which showed reflection of 97.9 %.

Apparently, protection against other kinds of laser radiations is possible by selecting suitable materials for absorption and reflection.

Table 1. Absorption coefficient of cellophane sheet

Colour	green	purple	blue	red	yellow	colourless
Absorption coefficient (/mm)	98	69	27	4.8	2.4	0.47

(Thickness of one sheet = 20  $\mu$ m = 2.9 mg/cm<sup>2</sup>)

Table 2. Absorption by optical glass filters (Toshiba).

	VG 52	VG 54
Thickness	2.75 mm (0.70 g/cm <sup>2</sup> )	3.17 mm (0.78 g/cm <sup>2</sup> )
Absorption	99.9 %	99.8 %
Absorption coefficient	2.51 /mm	1.93 /mm

### Protection against microwaves

Microwave ovens used by the general public should be designed and manufactured as to leak no radiation from the ovens. Control area should be established around microwave radiator so as to

protect the public from radar radiation. It is well known that copper wire netting is effective to protect researchers and workers from direct microwave radiation (Fig. 3).

According to a research by Dr. K. Ban (Nagoya University), protection clothes or protection box which is made of copper wire netting of 24 mesh is effective against 2,440 MHz microwave radiation. Scattered radiation inside the clothes or box was nearly zero.<sup>4</sup>

#### Regulations on control of non-ionizing radiations in Japan

There are no regulations on the permissible level of these radiations such as those set forth against ionizing radiations on human body. The estimated number of laser equipments is about 30,000 (Table 3) and that of ovens, much rapidly pervading, is about 1,300,000 (Table 4) as of Dec. 31, 1972 in Japan. In view of these situations, the "Research Committee on Biological Effect of Non-ionizing Radiation from Home Use Instruments" (Chairman, Dr. N. Yamagata) was organized in the Ministry of Health and Welfare last year. The Committee collected and examined many domestic and foreign literatures on biological effect of non-ionizing radiation, and also investigated foreign regulations. A report on the above was published by the Committee.

Table 3. Estimated number of laser equipments in Japan  
(as of Dec. 31, 1972)

Total number --- approximately 30,000						
{	Domestic	---	95 %			
	Imported	---	5 %			
{	Gas laser	---	95.2 %	{	He-Ne	92.4 %
		Ar			1.8 %	
		CO <sub>2</sub>			0.7 %	
		Others			0.3 %	
	Solid state laser	4.8 %				
Others	---	few				

(Estimated from the data of Industrial Marketing Consultant Co., IED)

Table 4. Estimated number of microwave ovens in Japan  
(as of Dec. 31, 1972)

Year	Number
1969	311,492
1970	413,901
1971	299,428
1972	approximately 610,000
total produced	" 1,630,000
exported	" 330,000
domestic use	" 1,300,000

(Estimated from the data of Research & Statistics Division, the Ministry of International Trade and Industry)

Currently, regulations are applied to the manufacturers of these devices by the Ordinance of Ministry of International Trade and Industry as follows.

- (1) Frequency of the microwave should be within the range of  $(2,450 \pm 50)$  MHz.
- (2) There should be installed such as additional cut-off device for the oscillator as is motivated by opening the oven door.
- (3) Radiation leakage measured at any 5 cm distance from the oven under the condition that a beaker containing  $(275 \pm 15)$  ml of water is placed at the centre within the oven should be
  - a) less than  $1 \text{ mW/cm}^2$  when the door is shut, and
  - b) less than  $5 \text{ mW/cm}^2$  when the door is ajar to the maximum extent before the cut-off device operates.
- (4) Radiation intensity, except that of the frequency within  $(2,450 \pm 50)$  MHz, should be
  - a) less than  $25 \text{ } \mu\text{W/m}$  at  $300\pi$  from the oven of under 500W, or
  - b) less than  $25\sqrt{P/500} \text{ } \mu\text{W/m}$  (where P is the out-put in W) at  $300\pi$  and less than  $10 \text{ } \mu\text{W/m}$  at  $1,600\pi$  from the oven of above 500 W.

This Committee is expected to examine actively on the maximum permissible energy on human body.

#### Reference

1. Kazutomo Ban: Studies on biological effects of microwave radiation (1st report), *Nippon Acta Radiologica*, 22, (6) 743-749, 1962.

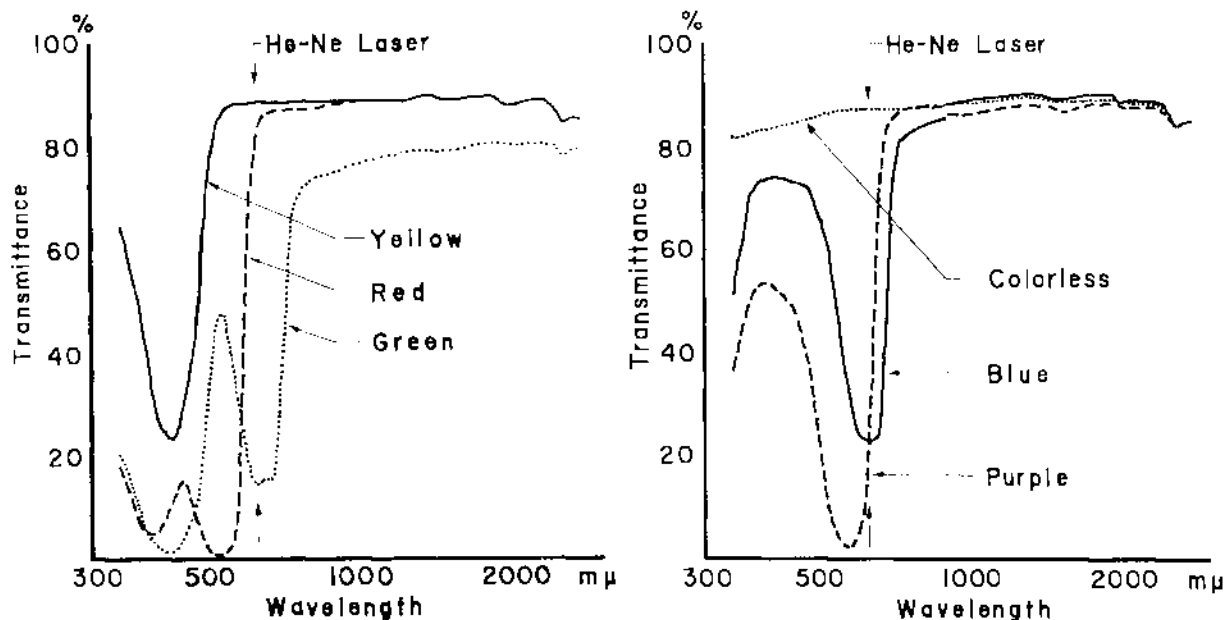


Fig. 1. Spectroscopic transmittance of coloured cellophanes.

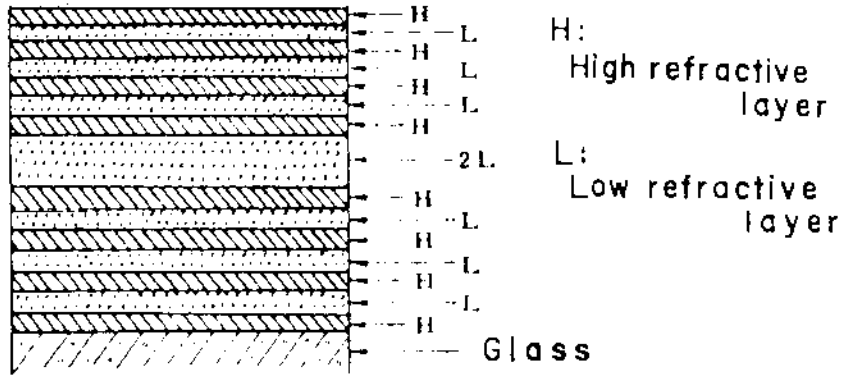


Fig. 2. Mirror of non-metallic multilayer film.



Fig. 3. Garment for protection made with 24 mesh copper nets.

PROGRESS IN THE REDUCTION OF MICROWAVE EXPOSURE FROM MICROWAVE  
OVENS USED IN COMMERCIAL FOOD VENDING OPERATIONS 1/

by

Martin C. Wukasch, P.E. 2/

and

Joseph F. Thiel 3/

Abstract

During the past two years microwave ovens used in commercial food vending operations have been surveyed in cooperation with State participating Local Health Departments and Districts in Texas. These surveys have shown that there is a correlation between general sanitation and maintenance with leakage of microwave radiation. Additionally, since October 6, 1971, the effective date of the microwave oven manufacturing standard (developed under the Radiation Control for Health and Safety Act of 1968, PL 90-602), significant reduction in the percentages of leaking ovens has been noted for ovens manufactured after the effective date.

Introduction

The use of microwave ovens in the commercial food vending operations business over the past several years has been on a steady increase. Among the variables influencing this increase is the fact that extended storage time of refrigerated food is considerably greater than that of food stored hot, and the fact that microwave ovens can quickly heat cold food to serving temperatures.

The biological effects of microwave radiation are generally separated into two divisions, thermal and non-thermal. Present adopted standards relate to the thermal effects of microwave exposure. The ability of microwave ovens to heat food quickly can heat parts of the human body just as quickly. With the thermal sensors just under the skin and the maximum temperature from microwave heating occurring much deeper, the potential exists for significant damage prior to the sensation of pain.

---

1/ Prepared for the 3rd International Radiation Protection Association, Washington, D. C., September 9-13, 1973

2/ Director, Division of Occupational Health and Radiation Control, Texas State Department of Health, Austin, Texas

3/ Environmental Health Specialist IV, Nonionizing Radiation Program, Division of Occupational Health and Radiation Control, Texas State Department of Health, Austin, Texas

Federal regulations found in 21 CFR 278.212 require that all microwave ovens manufactured after October 6, 1971, have microwave radiation leakage of less than a power density of one milliwatt per square centimeter at any point five centimeters or more from the external surface of the oven prior to first sale and, thereafter, less than five milliwatts measured at the same distances.

The American Conference of Governmental Industrial Hygienists' "Threshold Limit Values of Physical Agents", recommends a time weighted microwave power density exposure not to exceed 10 milliwatts per square centimeter, with total exposure time limited to the 8-hour workday. However, this limit may be exceeded under the following conditions: for average power densities between 10 and 25 milliwatts per square centimeter, total exposure time shall be limited to no more than 10 minutes for any 60 minute period during the workday; and, for average power density levels in excess of 25 milliwatts per square centimeter, exposure is not allowed.

The U. S. Department of Labor, Occupational Safety and Health Administration, Occupational Safety and Health Standards are not quite so lenient. In 29 CFR 1910.97 they established a Radiation Protection Guide of 10 milliwatts per square centimeter averaged over any possible 0.1-hour period. This means the following: power density of 10 mW/cm<sup>2</sup> for periods of 0.1-hour or more; and, energy density of 1 mW-hr/cm<sup>2</sup> during any 0.1-hour period.

Beginning early in 1971, microwave ovens were surveyed to determine the rate of failure to meet the above standards. The ovens used in commercial food vending operations were selected for this study as it was felt that they would show the effects of extreme product use and abuse, since they are used by the general public without maintenance on its part. The typical vending installation consists of at least one cold food vending machine, one microwave oven and several drink and candy machines. The installations are generally serviced daily in the morning, with the route man cleaning the oven.

#### Survey Methods

Our surveys were done using a Narda 8100 Electromagnetic Radiation Monitor with a standard five centimeter spacer cone. The ovens were operated at their maximum output using a standard load of 7 Oz. of drinking water in a plastic drinking cup. This load is less than that used in 21 CFR 278.212; however, the results of the tests on defective ovens did not vary significantly with this reduced load, (206 vs 275 ml).

A total of 561 ovens was surveyed in commercial food vending operations with 104 of these being manufactured after the effective date of the manufacturing standard. The use of trained local health department personnel greatly aided the completion of this survey program as well as acquainting the local personnel with the hazards associated with microwave oven radiation emission.

#### Survey Results

Of the 561 microwave ovens used in commercial food vending operations surveyed, 452 or 81% were found to be emitting microwave radiation less than the present federal product standard of 5 mW/cm<sup>2</sup> as measured at 5 centimeters. Six percent of the ovens were in the 5-10 mW/cm<sup>2</sup> range, three percent were in the 10-15 mW/cm<sup>2</sup> range, and 11% were in excess of 15 mW/cm<sup>2</sup>.

The eleven percent in excess of 15 mW/cm<sup>2</sup> were ordered removed from service until corrections could be accomplished by the vending company. This action level was determined as a maximum allowable level to continue operation of the ovens. Ovens in the 10-15 mW/cm<sup>2</sup> range were allowed to operate; however,

the vending company was notified of the defect and requested to repair the oven within 24 hours. The 15 mW/cm<sup>2</sup> level was determined from OSHA regulations using a minimum exposure time of 1 minutes. It is felt that exposure times under one minute are difficult to determine. Under normal use conditions found during the survey, one minute was found to be a typical user exposure/use time.

If we use the ACGIH exposure ceiling value of 25 mW/cm<sup>2</sup>, we find that six percent of the ovens exceeded that value. Also, four percent of the ovens were found to be in excess of 50 mW/cm<sup>2</sup>.

TABLE I

Power Density	% of ovens less than or equal to the power density	% of ovens manufactured prior to October 6, 1971 less than or equal to power density	% of ovens manufactured after October 6, 1971 less than or equal to power density
5 mW/cm <sup>2</sup>	81%	77%	98%
10 mW/cm <sup>2</sup>	87%	84%	100%
15 mW/cm <sup>2</sup>	91%	88%	100%
50 mW/cm <sup>2</sup>	96%	95%	100%

Table I shows the results and distribution of the various surveys broken down into ovens manufactured prior to or after the October 6, 1971, manufacturing standard date. It is of significance to note that only two new ovens out of 104 surveyed failed to meet the new standard. Their emission levels were found to be 6 and 10 mW/cm<sup>2</sup>.

TABLE II

Ovens Manufactured prior to October 6, 1971: Total of 167 in group

Power Density (mW/cm <sup>2</sup> )	# of ovens in Power Density range found dirty	# of ovens in Power Density range found clean	% of ovens found dirty in each range
P < 5	50	79	39%
5 < P < 10	5	6	45%
10 < P < 15	7	3	70%
15 < P < 50	11	4	73%
50 < P	4	2	67%

Ovens Manufactured after October 6, 1971: Total of 72 in group

P < 5	22	48	31%
5 < P < 10	1	1	50%
10 < P	0	0	--

The data in Table II would seem to indicate that the new ovens are being maintained in the same condition and under the same sanitary standard as the older units. However, one should note that in older units there is a correlation between the dirty units and those showing excessive leakage, thereby, showing the importance of proper maintenance.

Swabs were taken on several microwave units to determine whether biological growth could be supported inside the ovens. In the test, swabs soaked in growth media were wiped over an eight square inch area inside the oven. The swabs were stored in a sterile vial and refrigerated until returned

to the Texas State Department of Health laboratory for growth under standard conditions. Colony counts in excess of 10,000 were found; however, this is not a true indication of the total condition of the ovens. More data is still needed to give the true picture of the ovens in operation, since the areas wiped were not food contact surfaces.

If we accept the five milliwatt per square centimeter standard, then one may analyze our results on a vendor basis to determine both microwave emission and sanitary conditions and perhaps determine if the vendor is doing his job. Table III presents our data in this matter. Vending companies doing business under the same name in several different cities have been lumped together.

TABLE III

Vendor Symbol (10 or more ovens)	% less than 5 mW/cm <sup>2</sup>	% of all ovens found dirty when surveyed "U" means undetermined
A (20)	85%	20%
B (22)	100%	U
C (28)	64%	36%
D (113)	74%	27%
E (15)	87%	U
F (85)	87%	33%
G (20)	60%	47%
H (12)	92%	33%
I (51)	84%	36%
J (47)	79%	15%
K (13)	69%	U
L (68)	81%	U
494 ovens	81%	30%

The last figures in the column are presented for statistical purposes only.

The data in Table III when taken together with the data in Tables I and II allows one to analyze a given vendor's maintenance program.

Conclusions

The microwave ovens manufactured after the effective date of the federal product standard, October 6, 1971, have microwave emissions much less than those manufactured prior to the standard date. This lower emission is maintained even though they receive the same maintenance as the older ovens.

The general sanitary condition of microwave ovens in the commercial food vending business leaves much to be desired.



ULTRAVIOLET RADIATION DOSIMETRY  
UTILIZING THERMOLUMINESCENCE

A. Dhar\*, L. A. DeWard and T. G. Stoebe  
Division of Metallurgy  
University of Washington  
Seattle, WA 98195

ABSTRACT

An integrating dosimeter to detect ultraviolet radiation has been developed using the process of "direct" ultraviolet stimulation. This process yields a thermoluminescent signal after an exposure to ultraviolet radiation without prior exposure to ionizing radiation. Various crystals were compared for sensitivity to ultraviolet radiation. The spectral sensitivity, linearity with exposure and long range stability of the stored energy were investigated for MgO crystals which had the most sensitive response. The results of these tests indicate that a sensitive ultraviolet dosimeter can be developed using this phenomenon.

INTRODUCTION

Ultraviolet radiation results from a variety of industrial processes, various kinds of lamps specially designed for ultraviolet emission and natural sources such as sunlight. In addition to the medical therapeutic applications of ultraviolet (u.v.) radiation, there is an increasing application of u.v. radiation for disinfecting and sterilizing<sup>1</sup> materials in industrial and public health areas. The injurious effects of u.v. energy appear to be related to their ability to be absorbed by nucleic acid<sup>2</sup>. Dermatologists and skin photobiologists are mainly concerned with the deleterious effects on man, such as erythema (sunburn), painful inflammation of the membrane of the eye, and the possibilities of skin cancer<sup>3</sup>. The actual effectiveness of sunburn is expressed in terms of the "standard erythema spectrum" covering wavelengths 250 nm to 320 nm. This spectrum and the erythema effectiveness have been widely studied,<sup>4-7</sup> the most sensitive region of erythema being between the wavelengths of 290 and 320 nm. The smallest amount of ultraviolet energy that will produce visible erythema is called a Minimum Erythema Dose (MED) and is defined as the minimum amount of energy required to elicit a just barely visually detected reddening of the skin at one particular wavelength. This MED value, however, is dependent on several factors such as the observation time of the erythema and the anatomical site of testing, as well as the irradiation wavelength and other factors. Thus it would be useful to have a convenient dosimeter which would act as an absolute measurement device.

A number of phosphors utilizing the thermoluminescent phenomenon have been studied as possible dosimeters for the ultraviolet region. Thermoluminescence has received extensive study and use in the past for ionizing radiation dosimetry<sup>8</sup>. The low cost per phosphor, the absence of any associated electronics

\* Associated with the Division of Radiological Sciences

at the site of measurement, and the fact that the thermoluminescent phosphor can operate unattended, are some of the advantages in using thermoluminescence. The small size of a solid phosphor allows the possibilities of performing skin transmission studies in vivo and development as a personnel dosimeter.

One of the first thermoluminescent phosphors receiving attention<sup>9,10</sup> for u.v. detection was  $\text{CaSO}_4:\text{Mn}$ , but it was not sensitive in the erythral region. Wilson et.al.<sup>11</sup> studied a natural  $\text{CaF}_2$  phosphor using the technique of "transferred thermoluminescence (TL)". The same process was utilized by McCullough and Cameron,<sup>12</sup> and Okuno and Watanabe<sup>13</sup> in natural  $\text{CaF}_2$  by Nambi and Higashimura<sup>14</sup> in  $\text{CaSO}_4:\text{Tm}$  and  $\text{CaSO}_4:\text{Dy}$ . "Transferred Thermoluminescence" involves a sequence of operations. The phosphor is first heated to  $700^\circ\text{C}$  to empty deep traps and then is exposed to a "standard" amount of X or  $\gamma$  radiation. The phosphor is then heated to  $400^\circ\text{C}$  for 10 minutes (this empties the traps with glow peak temperatures below  $400^\circ\text{C}$  but does not empty the deeper traps). Upon exposure to ultraviolet radiation, some of the electrons in the deeper traps are "transferred" to the shallower traps corresponding to lower temperature ( $<400^\circ\text{C}$ ) glow peaks. The glow curve of the transferred thermoluminescence is then measured up to  $400^\circ\text{C}$ , and the transferred TL intensity may then be related to the ultraviolet radiation exposure. The transferred TL intensity is also related to the number of traps previously filled by ionizing radiation.

As is evident, this transferred TL process is complex and time consuming. In contrast, the process of "direct" ultraviolet stimulation, with which this work is mainly concerned, yields a thermoluminescent signal after an exposure of ultraviolet radiation, without any prior exposure to ionizing radiation. This simplifies the process and is more convenient for practical use. This paper is concerned with the feasibility of using materials involving the "direct" stimulation process as dosimeters for ultraviolet radiation.

#### EXPERIMENTAL DETAILS AND MATERIALS SURVEY

An xenon arc lamp (PEK:X-151-1453) was used as a u.v. irradiation source. This type of xenon source was used because its spectral distribution closely resembles that of the sun. Other experiments used a monochromator (Jarrell-Ash 1/4 meter) with 3 mm exit slits to select a particular u.v. wavelength. All u.v. intensities were measured with a radiometer (YSI-kettering, model 65) at a standard distance of 5mm from the exit slit of the monochromator. A Harshaw model 2000 thermoluminescence detector was used to record the TL glow curves.

A general survey of the u.v. response of various materials was performed first to find those showing the greatest promise for direct sensitizations. Fifteen different materials were tested by irradiating for 10 minutes with the light directly from the xenon lamp, so that the samples were exposed to a range of wavelengths. The materials did not receive any other radiation prior to u.v. irradiation. The resultant response of the materials tested is shown in Table I, where MgO and magnesium silicate doped with terbium seem to be the most sensitive to direct u.v. stimulation. The results indicate that MgO crystals annealed in an argon atmosphere at  $2000^\circ\text{C}$  for 24 hours have more thermoluminescent sensitivity than the as received or nonannealed MgO crystals; this annealing procedure affects the defect structure of the crystal, which in turn affects the thermoluminescence. The results of this survey also indicated that the total impurity content in MgO plays a significant role in thermoluminescence, since impure MgO has less thermoluminescent sensitivity than the purer MgO.

#### RESULTS AND DISCUSSION

After these initial tests it was decided to experiment further with MgO



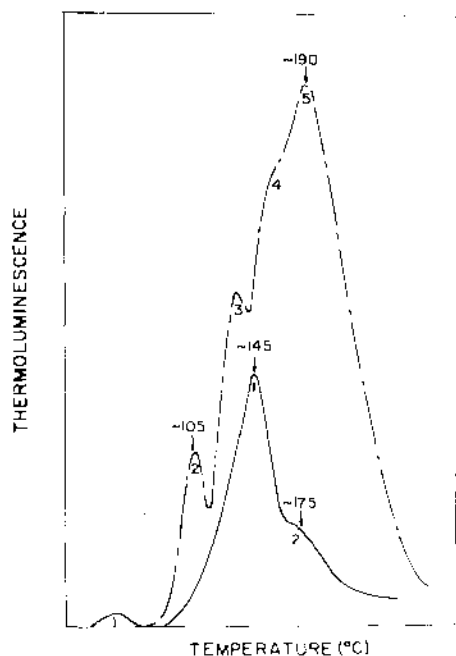


Fig. 1. Thermoluminescent glow curve of MgO-PA type crystal (—) exposed to  $3 \times 10^4 \mu\text{W}\text{-sec}/\text{cm}^2$  at 296.7 nm and LiF (TLD-100) single crystal (---) exposed to 100R of 250 kVp x-rays (thoreas III filter) Peak height of LiF peak 5 is 2.2 times higher than peak 1 in MgO.

which have been previously irradiated with whole u.v. (mixed wavelengths) for 10 minutes before exposure to a particular wavelength. The thermoluminescent intensity of the untreated crystals after such a preliminary exposure followed by an exposure at a particular wavelength was similar to that of the crystals which previously had been treated with whole u.v. exposure. The minimum amount of time required for sensitization for PA crystals was found to lie between 10 and 15 minutes; a 20 minute sensitization period shows a drastic reduction in thermoluminescent sensitivity. This sensitization period of 15 minutes was found to give a maximum response for a number of MgO crystals with different impurity levels. The greatest sensitivity however, was for the purest material (PA crystals).

The sensitivity of PA and NB crystals as a function of wavelength for the u.v. region is shown in Figure 2. All the crystals received the same exposure of  $3 \times 10^4 \mu\text{W}\text{-sec}/\text{cm}^2$  at all wavelengths. This value is the Minimum Erythral Dose (MED) at 296.7 nm based on the standard erythral curve. PA crystals have a completely flat response over the entire wavelength region; thus no wavelength corrections are necessary for this kind of a crystal. NB crystals, on the other hand, show an increase in thermoluminescence at 285 nm followed by a continuous decrease in thermoluminescence to 305 nm and then a uniform response to 325 nm. Thus if NB crystals were to be used for dosimetry purposes, corrections for this wavelength dependence would have to be made. The results shown in Figure 2 are for PA crystals that were sensitized about nine months before this study was done, and for PA and NB crystals sensitized before the experiment. These results show that this sensitization procedure, discussed previously, decreases

crystals since they responded with the greatest sensitivity. Spectral sensitivity, linearity with exposure and the short range stability of the stored energy was studied. Two sets of MgO crystals, designated as PA and NB, were studied. PA crystals have a total impurity content of 255 ppm and NB crystals have total impurities of 815 ppm; more details on these samples are given by Srinivasan et.al<sup>15</sup>.

Figure 1 shows a typical glow curve of a PA crystal, with a main glow peak at about 145°C designated as peak 1. Peak 2 and peak 3 (not shown) are at about 175°C and 325°C respectively. For comparison, a glow curve of X-irradiated LiF (TLD-100) is also shown. In PA crystals, peak 2 is small and not very distinctive, and peak 3 is very broad. Peak 1 and peak 2 were completely absent if there was no prior u.v. irradiation. The most effective peak for dosimetry is peak 1 at 145°C and therefore, all further tests concentrated on this peak. Glow curves of NB crystals show peaks at the same temperature as in PA crystals and an additional peak at 120°C.

The MgO crystals were observed to exhibit a sensitization phenomenon. Untreated crystals irradiated only at one particular wavelength of u.v. radiation show much less TL than similar crystals

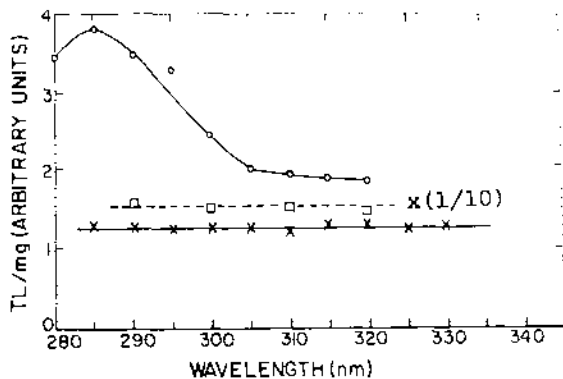


Fig. 2. Thermoluminescent output (TL/mg) for MgO PA and NB crystals as a function of wavelength of irradiation. The circles represent NB immediately after sensitization. The squares connected by the dashed line represent PA immediately after sensitization, whereas the crosses connected by the solid line are for PA sensitized 9 months before the tests.

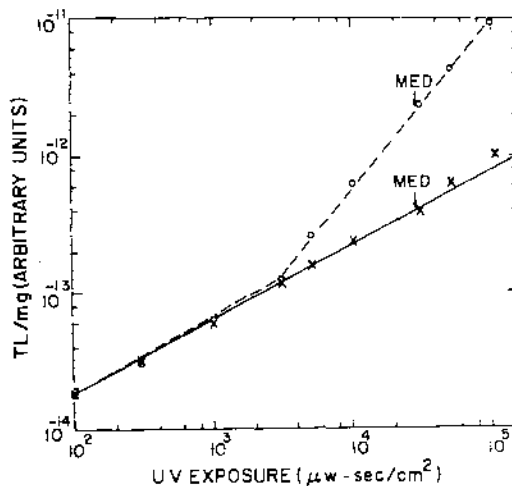


Fig. 3. Thermoluminescent output (TL/mg) for MgO-PA(x) and NB (o) crystals as a function of u.v. exposure at 295 nm. Arrows indicate the MED value at 295 nm.

approximately an order of magnitude in effectiveness in a nine month period. The sensitization decreases by 20% within 15 days. Thus, elapsed time after the sensitization procedure is an important variable requiring attention. Further studies to delineate an optimum time are in progress.

The response of the MgO crystals as a function of ultraviolet exposure at 295 nm is shown in Figure 3. The exposure was determined up to an order of magnitude above the minimum erythral dose (MED indicated in Figure 3). The linear relationship with exposure shown by PA crystals over the entire region would be extremely useful for dosimetry purposes. The relationship between TL/mg and exposure for NB crystals shows two regions of linearity. This kind of MgO crystal could also be used for dosimetry purposes provided the region of operation is roughly known.

Thermoluminescent reproducibility, another important dosimetric consideration, has been checked several times under various conditions. The repeated thermoluminescent measurements yielded a standard deviation of less than  $\pm 5\%$ . This 5% reproducibility in the PA crystals was obtained without any intermediate heat treatments or sensitization procedures; this could be a big advantage of using this system, since no annealing treatments are needed for repeated use of these crystals such as those required for LiF (TLD-100) crystals after each thermoluminescent measurement. The effect of storage at room temperature and of room light, which are other important aspects of the reproducibility of these crystals, was studied for a number of crystals. The crystals were u.v. irradiated to an exposure of  $3 \times 10^4 \mu\text{W-sec/cm}^2$  and the thermoluminescence was measured. The crystals were then cycled through the readout procedure a few times after the first thermoluminescent measurement to assure that there was no thermoluminescence left in these crystals. Then half of the samples were left in room light and the other half was wrapped in a black paper and left in a drawer. After 24 hours, all the crystals were read out again. Crystals which were left in black paper did not show any thermoluminescence whereas those left in room light had a response which was less than 0.5% of the original thermoluminescence, corresponding to an exposure of  $1.5 \times 10^2 \mu\text{W-sec/cm}^2$ . Thus

under normal use, care must be exercised to avoid unnecessary exposure to room light although the response after 24 hours may be considered negligible. If u.v. irradiated crystals are stored at room temperature wrapped in black paper, the response decreases exponentially with time; peak 1 has a half life of 192 hours (8 days).

Two regions of sensitivity and linearity were noted above (Figures 2 and 3) for the impure NB crystals. Since thermoluminescence in MgO may be correlated with the valence changes of Fe and Cr,<sup>16,17</sup> it is probable that these two phenomena might be related to the impurity defect structure within the crystals. The impurity defect structure is affected by annealing, and thus the presence of certain defect structures might yield greater u.v. sensitivity as mentioned previously. Reactions between vacancies and impurities, and the effects of their concentrations in MgO have been studied as a function of temperature using optical methods<sup>18</sup> and ionic conductivity techniques<sup>19</sup>. Irradiation with u.v. light or heating in oxygen or magnesium vapor to change the valence state of the impurities will produce optical absorption bands in MgO crystals which contain no intentionally added foreign ions. Wertz et.al.<sup>20,21</sup> presented evidence for changes in valence of various foreign ions in MgO during irradiation and other treatments. Hansler and Segelken<sup>16</sup> have reported that the amount of Fe<sup>3+</sup> could be changed during ultraviolet irradiation depending on the previous treatment. These changes could be reversed during thermoluminescence. The same effects have been observed with Cr<sup>3+</sup>. Therefore it was suggested that thermoluminescence might be correlated with the valence changes of impurities in MgO. Further investigations are in progress to attempt to correlate changes in the defect structures with maximum thermoluminescent response to ultraviolet irradiation.

#### CONCLUSIONS

On the basis of these results, MgO-PA crystals have been found to be feasible for u.v. dosimetry in the erythemally effective region of 290 to 320 nm. MgO crystals of lesser purity, represented by NB crystals in this paper can also be used but require more care. These MgO crystals yield a thermoluminescent signal after a "direct" u.v. radiation stimulation without any prior exposure to ionizing radiation. For the greatest response, the crystals should be sensitized by irradiating them with whole u.v. (mixed wavelengths) for 15 minutes before using them. MgO-PA crystals are independent of the wavelength of irradiation and have a linear relationship of thermoluminescence as a function of exposure. Thus these crystals can be used without any corrective procedures. The crystals also can be reused immediately with a reproducible response to within 15%. However, these crystals are slightly affected by room light and as such, precaution should be taken to prevent the crystals from receiving unnecessary prolonged exposure to room light.

#### ACKNOWLEDGEMENTS

This work was supported by USPHS, Food and Drug Administration, Bureau of Radiological Health, under research grant RL00124-05. The MgO was supplied by the Oak Ridge National Laboratories through Dr. M. Srinivasan of this University.

#### REFERENCES

1. "Electronic Product Radiation and the Health Physicist"; U.S. Department of H.E.W., BRH/DEP: 70-26; October, 1970, p. 391.
2. S. M. Michaelson, Proc. I.E.E.E. 60, 398 (1972).
3. L. R. Koller, "Ultraviolet Radiation", John Wiley & Sons Inc., N.Y. (1965).

4. H. Blum and W. S. Terus, Am. J. Physiology 146, 97 (1946).
5. H. Blum and W. S. Terus, Am. J. Physiology 146, 107 (1946).
6. M. A. Everett, R. L. Olson and R. M. Sayer, Arch. Dermat. 92, 713 (1965).
7. R. L. Olson, R. M. Sayer and M. A. Everett, Arch. Dermat. 93, 211 (1966).
8. J. R. Cameron, N. Suntharlingam and G. Kenney, "Thermoluminescent Dosimetry," University of Wisconsin Press, Madison, (1968).
9. T. Lyman, Phys. Rev. 48, 149 (1935).
10. K. Watanabe, Phys. Rev. 83, 785 (1951).
11. C. R. Wilson, F. M. Lin and J. R. Cameron, USAEC Progress Report No. C00-1105-136 under contract AT-(11-1)-1105 (1967).
12. E. C. McCullough, G. D. Fullerton and J. R. Cameron, J. Appl. Phys. 43, 77 (1972).
13. E. Okuno and S. Watanabe, Health Phys. 23, 377 (1972).
14. K. S. V. Nambi and T. Higashimura, Proc. III Intl. Conf. Lumn. Dos., Denmark, Risø report No. 249, p. 1107 (1971).
15. M. Srinivasan and T. G. Stoebe, J. Mat. Sci. (in press).
16. R. L. Hansler and W. G. Segelken, J. Phys. Chem. Solids 13, 124 (1960).
17. W. M. Ziniker, J. K. Merrow and J. I. Mueller, J. Phys. Chem. Solids 33, 1619 (1972).
18. A. M. Glass and T. M. Searle, J. Chem. Phys. 46, 2092 (1967).
19. S. P. Mitoff, J. Chem. Phys. 36, 1383 (1962).
20. J. E. Wertz and P. Auzins, Phys. Rev. 106, 484 (1957).
21. J. E. Wertz, P. Auzins, J. H. E. Giffiths and J. W. Orton, Dis. Farad. Soc. 26, 66 (1958).

TABLE I

Thermoluminescent response of various materials after exposure to the entire spectrum of a xenon lamp for 10 minutes.

Sample	Peak Temperature (C°)	Thermoluminescent Peak Height
MgO		
(1) Annealed Pure PA Crystal <sup>a</sup>	145	0.61 x 10 <sup>-8</sup>
	175	0.15 x 10 <sup>-8</sup>
(2) Unannealed Pure Crystal <sup>a</sup>	136	0.22 x 10 <sup>-8</sup>
(3) NB Crystal (Lower Purity) <sup>b</sup>	120	0.26 x 10 <sup>-8</sup>
	145	0.39 x 10 <sup>-8</sup>
Magnesium Silicate: Terbium <sup>c</sup>	190	0.17 x 10 <sup>-8</sup>
Magnesium Borate: Terbium <sup>c</sup>	115	0.12 x 10 <sup>-11</sup>
Calcium Fluoride <sup>d</sup>		
(1) CaF <sub>2</sub> :Mn	153	0.62 x 10 <sup>-11</sup>
	185	0.35 x 10 <sup>-11</sup>
(2) CaF <sub>2</sub> :Dy	170	0.43 x 10 <sup>-11</sup>
	215	0.33 x 10 <sup>-11</sup>
Calcium Sulphate <sup>d</sup>		
(1) CaSO <sub>4</sub> :Mn	120	0.23 x 10 <sup>-12</sup>
(2) CaSO <sub>4</sub> :Dy	190	0.12 x 10 <sup>-12</sup>
Lithium Borate <sup>e</sup>	119	0.76 x 10 <sup>-11</sup>
	165	0.40 x 10 <sup>-11</sup>
LiF (TLD-100)	No Detectable Response	

<sup>a</sup> Oak Ridge

<sup>b</sup> Norton Company

<sup>c</sup> Dai Nippon Toryo Co., Ltd.

<sup>d</sup> Harshaw Chemical Company

<sup>e</sup> Grown in this laboratory

A CALIBRATION FACILITY FOR MICROWAVE MONITORS -  
DESIGN AND OPERATING EXPERIENCE

I.S. Leith

Australian Radiation Laboratory

Melbourne, Australia.

(A broad-band microwave calibration facility has been designed and partly completed. Evaluation of the facility and its progressive design are described and calibration results presented).

---

Since 1963, the Australian Radiation Laboratory has taken an active interest in the hazards of microwave radiation, and has offered advice on request. Following a specific case of inadvertent entry of persons to an area close to a powerful transmitting radar antenna in 1966, a Narda Model B86B3 broadband monitor was purchased to permit measurements to be made in support of the calculations relied on to that date.

It was realised that the calibration of the available monitors was made at several frequencies only, that the variation of calibration factors from one frequency to another was quite large and that "holes" in the frequency response may well occur between calibration points. Thus the need for broad-band calibration was recognised at this early stage. Microwave ovens were subsequently introduced into Australia and Narda Models 8100 and 8200 near-field monitors were purchased in due course. Initial investigations soon revealed marked differences in response of identical probes, hence calibration was first undertaken at 2450 MHz. The system shown in Figure 1 was set up as a first step. At this time no calibration manual for the instruments described above was available from the manufacturer.

The microwave absorber used in this system is Plessey Type AF50 pyramidal absorber twelve inches thick and covering an area 8 feet by 8 feet. The facility was set up on a discontinuous basis in the centre of a large room with a ceiling 11 feet high and having a width of 14 feet. The nearest broad surfaces were thus approximately 13 wavelengths away from receiver and transmitter at 2450 MHz. The radiation source is a diathermy generator (with additional mains power filtering and stabilising circuits) which incorporates a magnetron to produce up to 120 watts of power. A large mismatch at the generator output was successfully tuned out using a two-stub tuner. The circuit was set up using coaxial components because many components were not available in waveguide in LS-band and in order to facilitate extension to broad-band. The power measurement was made with a Hewlett Packard Model 432A thermistor meter and coaxial mount and the manufacturer's calibration was and is relied on as no microwave power or power density standard exists as yet in Australia. The antenna used is one of a pair of identical Narda LS-band

standard gain horns which have been connected by cover flanges to Karda waveguide-to-coaxial adapters. A pair was purchased to undertake measurements of antenna gain, initially to reduce the uncertainty in the gain quoted by the manufacturer.

Power density at distance R was derived using

$$W_R = \frac{P_0 G_R}{4 \pi R^2} \quad (1)$$

where  $G_R$  = Gain of antenna at distance R from antenna

$W_R$  = Power density at distance R from antenna

$P_0$  = Output power at the antenna

$P_0$  is known in terms of the power indicated on the meter

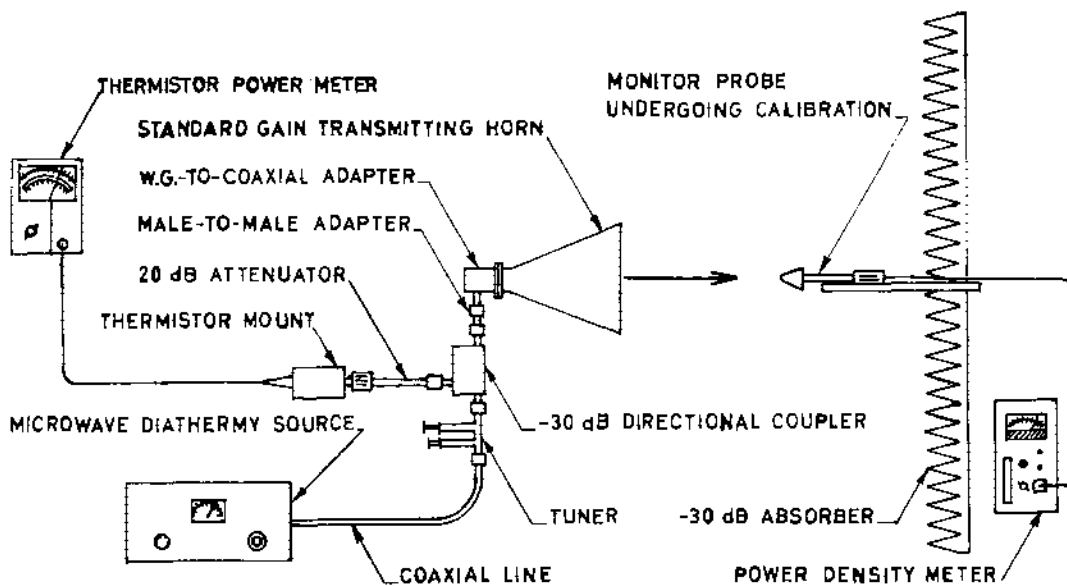


FIG.1 INITIAL SYSTEM FOR CALIBRATION OF MONITORS AT 2450 MHz

The initial calibrations were undertaken at an antenna separation of 3 metres although it was realised that side lobe and main beam reflections would be significant. Calculations show that for the antennas used, the usual far-field criterion of  $2a^2$ , taking  $a$  as the greatest distance between two points on the antenna aperture, is 3.09 metres at 2450 MHz. The total error in calibration could not be estimated as defects in the "anechoic enclosure" could not be determined, however, apart from this uncertainty the total uncertainty, after all possible corrections based on manufacturer's calibrations of components, was estimated to be +16%, -27%. This included an antenna gain uncertainty of  $\pm 7\%$  as given by the manufacturer. A significant part of the total uncertainty was due to conjugate mismatch losses calculated on the basis of manufacturer's specifications of V.S.W.R. The total actual error was thus thought likely to be much less than the maximum uncertainty calculated when tuned for maximum power transfer to the horn antenna.



The maximum reflection from the horn and adapters was calculated to be 7.4% but recent measurements using a dual directional coupler have shown only 0.4% of the forward power being reflected at 2450 MHz when mismatch has been tuned out. The effect of floor reflections has also been investigated with the use of additional absorber (Eastern Microwave Corp. Type FFP-8) recently purchased. This investigation showed received power density to be 2% lower at 3 metres when the floor was covered with absorber. However a 4% decrease was observed at 2 metres and a 4% increase at 96 inches, the calibration distance recommended by Narda for their Model 8100 probes.

An early calibration of a Model 8121 A probe at 3 metres gave a calibration factor of 1.19, corresponding to a response 16% below the expected value but within the limit of uncertainty. This was based on an antenna gain of 50 as given by the monitor manufacturer. In the calibration manual for the 8100 instrument, the manufacturer recommends the use of this gain figure at 2450 MHz at 96" from the front face of the horn. (The same manufacturer provided a gain calibration curve for the horn antennas which gives a gain of 50.13 at 2450 MHz, but it must be assumed that this is intended as a far-field gain as no distance is stated). This calibration was considered satisfactory but later the two horn antennas were used to obtain measurements of received power versus distance over the range 1 to 3 metres.

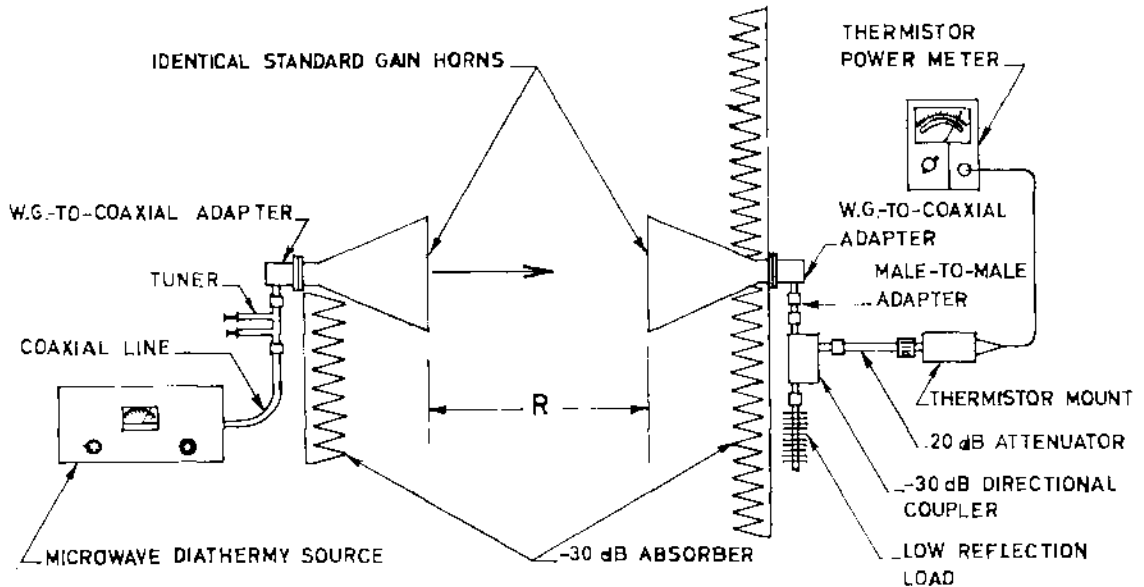


FIG.2 SYSTEM FOR MEASUREMENT OF GAIN OF IDENTICAL HORNS

The circuit shown in Fig. 2 was used. This circuit was acceptable because the microwave generator was shown in other measurements to be remarkably stable, linear and reproducible, especially if care was paid to tuning out the output mismatch. The gain was computed by the far-field power-transmission formula:

$$P_R/P_T = \frac{(\lambda G)^2}{4\pi R^2} \text{ where (2)}$$

- $P_R$  = received power
- $P_T$  = transmitted power
- $G$  = directional gain of both horns
- $\lambda$  = wavelength
- $R$  = antenna separation

As  $P_T$  and  $\lambda$  were kept constant, formula (2) reduces to

$$P_R = \frac{k G^2}{R^2} \quad (3)$$

$$\text{where } k = \frac{P_T \lambda^2}{4 \Pi} \quad (4)$$

and if  $G$  is constant an inverse-square relationship will exist between  $P_R$  and  $R$ .  $G$  was expected to vary because of multiple coupling between the antennas and phase variation across the face of each horn. Computed values of  $G$  varied from 42.6 at 1 metre to 51.7 at 3 metres. The inverse-square relation between  $P_R$  and  $R$  did not hold but could be recovered by the method of Jakes<sup>1</sup> by adding a small distance  $d$  to  $R$  and iterating this procedure until an inverse square curve was obtained. The distance to be added was 32 centimetres, which does not correspond closely to twice the axial height of the horns (approximately 40 centimetres). These are not optimum horns, but the departure from the axial height is rather marked and may indicate errors due to the inadequacy of the "anechoic" enclosure. This was further emphasised when similar range tests using a Narda Model 8121A probe and a single horn required addition of  $d = 20$  centimetres to produce an inverse square fit and constant  $G$ . Approximately one half of the value of  $d$  required for the two rectangular horns was expected. However, the gain computed at 96" was 50.6 which compares well with the gain figure of 50 recommended by the manufacturer in its model 8100 calibration manual. The inverse square fit corresponded to a far field (constant) gain of 57.4.

Recalculating the calibration factor for the initial calibration using the measured gain of 51.7, the response of the probe is 21% low, outside the manufacturer's limits and the uncertainty limit (when reduced by 7% to allow for mismatch removed by use of the tuner).

Although these investigations leave several questions unresolved with regard to absolute calibration, this early facility allowed accurate and useful comparisons of probes and monitors with valuable results. Also the high power of the microwave source allows power density linearity tests on all probes at reasonable separations from the transmitting antenna. These tests have shown all Narda probes for Models 8100 and 8200 to have closely linear response with zero intercept at zero transmitted power despite probe calibration corrections determined at 96" lying within the limits of + 48.7% and - 87.4%. The Narda Model B86B3 broadband monitors checked at 2450 MHz have responses which are fairly linear but have lines of best fit which have a positive intercept at zero power of 2 to 3 mw/cm<sup>2</sup>. At 2 metres, responses of two of these monitors were 66% higher than the estimated value. The manufacturer's recommended distance for calibration is 47". The gain variation measured between 47" and 2 metres was from 45.4 to 49.7 and does not appear to be the source of calibration variation from the manufacturer's setting. The calibration system is presently undergoing refinement. Fig. 3 indicates the system presently assembled and under test in the band 1.7 to 2.6 GHz. The principle of this system is a swept frequency power source employing a solid state sweep oscillator and travelling-wave-tube amplifier. This system is stabilised and levelled on the difference signal between forward and reflected power. The effective V.S.W.R. of the source is expected to be low and at some frequencies extremely small reflected power is expected. However, the horn, adapter combination has been swept frequency analysed recently. The V.S.W.R. is markedly variable and rather large at some frequencies, hence the system is expected to provide a more linear output power as frequency is swept than a system based on forward levelling alone. The output will be frequency dependent nevertheless due to the frequency dependence of horn gain, but this will be low over narrow sweep bands and will be calibrated at single

frequencies in the swept band.

Uncertainty of power measurement seems of minor significance in the total uncertainty but mismatch is of greater consequence due to frequency dependence of the components used in the system. Hence the methods proposed by Engen<sup>2,3,4</sup>, are being adapted at single frequencies to determine the power delivered to the load (the horn, waveguide-to-coaxial adapter combination). This method in one form involves reflecting 100% of the power generated back down the line which would exceed the damage limits of the travelling-wave tube. A more complex form of the method may be used to avoid this,<sup>5</sup> but instead an isolator formed by a three port circulator with matched load is included to protect the travelling-wave tube.

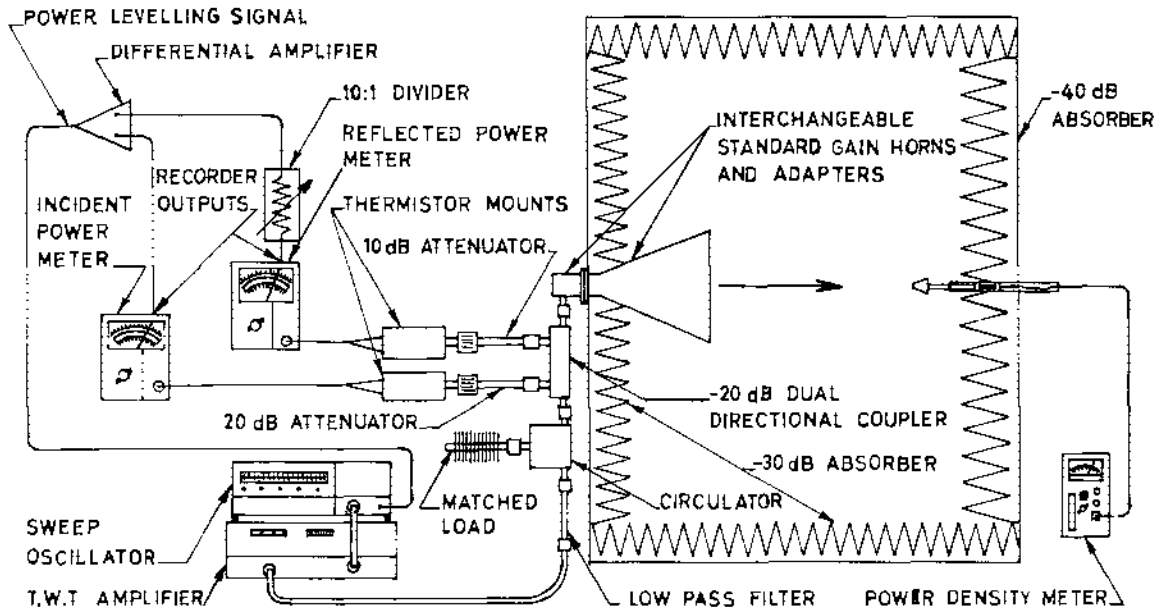


FIG. 3 SWEEPED FREQUENCY CALIBRATION SYSTEM IN L AND S BANDS

The anechoic enclosure is presently being extended in a large room using almost 300 square feet of Eastern Microwave Corp. FFP-8 absorber. Purchase of more absorber is proposed to provide a target wall 12 feet by 12 feet square composed of 18" solid pyramidal absorber and a rectangular enclosure 16 feet long with sides, ceiling and floor made of 8" solid pyramidal absorber. As the frequency band is extended to range from 10 to 20,000 MHz it is proposed to enlarge and lengthen the chamber considerably to counteract the deterioration of absorber performance at the longer wavelengths. True far field measurements are planned in the future, requiring an antenna separation of at least ten metres. It is planned to develop the anechoic chamber as a very wide rectangular chamber with square wedged end walls. A "quiet zone" with - 30 dB reflectivity some ten metres long will be sought. It is proposed to adapt a Hewlett Packard Model 8755 Frequency Response Test Set recently purchased to examine the chamber performance as it is developed. The null-balance technique described by Buckley<sup>6</sup> will be used. Further, the near field gains measured in the chamber when low reflectivity is achieved will be checked by computing the near field (Fresnel zone) power transmission formula.

The calibration system under test and its further extension to wideband is expected to offer several advantages, including accurate swept frequency calibration over broad frequency ranges and over small bands around frequencies of special interest such as 2450 MHz. This is considered important to detect severe frequency response of monitors and allow for frequency variation of sources of microwave hazard.

#### References

1. Jakes, W.C. Gain of electromagnetic horns. Proc. I.R.E. Vol. 39, Feb., 1951, pp. 160 - 162.
2. Engen, G.F. An improved method for microwave power calibration, with application to the evaluation of connectors. J. Res. Nat. Bur. Standards. Vol. 75C, No. 2, April - June, 1971, pp. 89 - 93.
3. Engen, C.F. A method of determining the mismatch correction in microwave power measurements. I.E.E.E. Trans. Inst. Meas. Vol. IM-17, No. 4, Dec., 1968, pp. 392 - 395.
4. Engen, G.F. Power equations: a new concept in the description and evaluation of microwave systems. I.E.E.E. Trans. Inst. Meas. Vol. IM-20, No. 1, Feb., 1971, pp. 49 - 57.
5. Engen, G.F. An introduction to the description and evaluation of microwave systems using terminal invariant parameters. U.S. N.B.S. Monograph 112, Oct. 1969.
6. Buckley, E.F. The design and evaluation of microwave anechoic chambers. Paper presented at Joint Meeting of IRE - PGME, PG MTT and PGAP, Chicago, Illinois, April 8th, 1960, pp. 14 - 16.

COMPUTER-ASSISTED LASER HAZARD CALCULATIONS AND A  
CRITICAL ANALYSIS OF THE PREDICTION EQUATIONS

W.A. Cornelius and I.S. Leitch  
Australian Radiation Laboratory  
Australia

(A computer program for laser hazard calculations with graphical output over the hazardous range is described. The assumptions implicit in the prediction equations in frequent use are examined and anomalies suggested.)

The application of lasers to both industrial and research purposes is becoming a widespread phenomenon. In this paper we shall deal specifically with the hazard evaluation of lasers emitting in the visible region of the spectrum. Gas lasers emitting at visible wavelengths are the most common and their uses include; surveying, rangefinding, alignment of tunnelling machines, pipelaying and holographic testing. Such lasers produce high intensity monochromatic beams of low divergence (i.e. well collimated) and because of these properties an optical system such as the human eye is capable of focusing a laser beam into a very small volume. The retina of the eye is particularly susceptible to damage from the heating effect of a focused beam. Accidental viewing of either a direct or specularly reflected laser beam may result in the production of thermal lesions on the retina. Such lesions may or may not seriously impair vision depending upon the extent of the lesion and its position within the visual field.

To evaluate the potential hazard of a particular laser one must first predict the power density likely to fall on the retina by direct viewing of the laser beam and it is to this matter that we now turn our attention. A number of authors have given methods for the calculation of retinal beam spot size and intensity 1,2,3,4,5,6,7. In summary, the light intensity incident on the retina is dependent upon the power transmitted through the eye and the size of the laser spot on the retina. The latter two quantities are in turn dependent upon; the transmission of the eye at the particular laser wavelength, the output power (or pulse energy) of the laser, the beam diameter at the output aperture, beam divergence (see Fig.1), pupil diameter, the distance of the observer from the laser, atmospheric attenuation and the degree of accommodation (or focusing) of the eye. For the purposes of hazard evaluation the accommodation of the eye will be taken to be that for which the retinal spot diameter will be a minimum i.e. the worst case condition.

Fig. 2 shows the calculated retinal intensity as a function of distance from a helium-neon gas laser operating at a wavelength of 632.8 nanometres with an output power of 3 milliwatts, an output beam diameter of 1 millimetre and beam divergence of 1.5 milliradians. A pupil diameter of 5 millimetres was used. These calculations are largely derived from the method given by Solon et al<sup>1</sup>, however a lower limit of 7 micrometres has been set to the diameter of the beam spot size. This 7 micrometres limit is believed to be that for which the minimum optical aberration can be achieved by the human eye. Atmospheric attenuation has also been taken into account although it is insignificant over the range of Fig. 2.

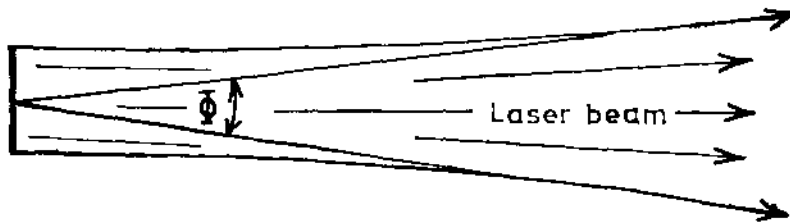


Fig.1. LASER BEAM DIVERGENCE ANGLE  $\phi$

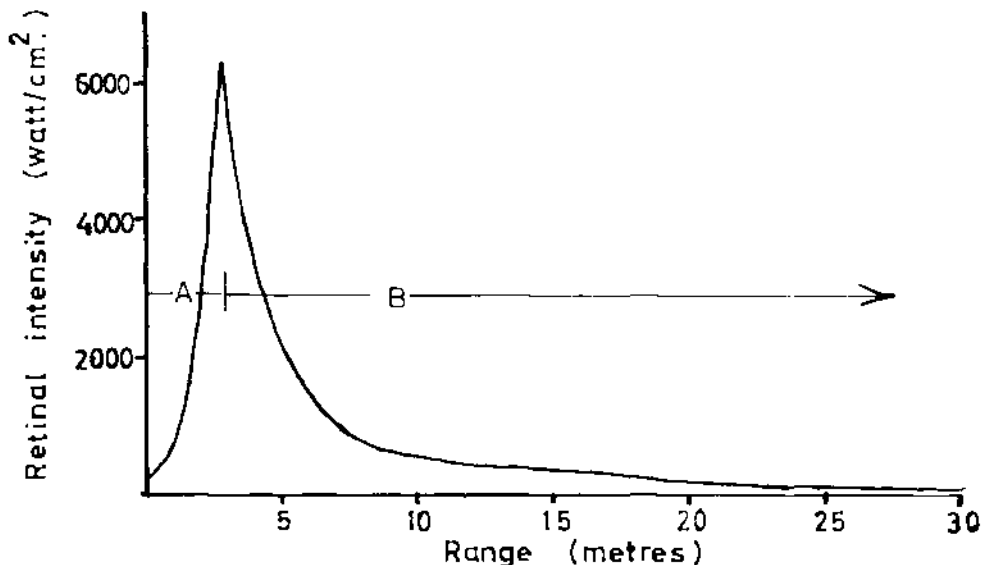


Fig.2. RETINAL INTENSITY VS. DISTANCE FROM LASER

For the sake of analysis three separate cases are considered; two cases close to the laser in what is termed the "near field" and the third case in the "far field" where the laser spot on the retina is no longer resolved and is determined by diffraction at the pupil. In Fig. 2 region A is the near field region and region B is the far field region. The large variation in intensity is due to the fact that the laser beam diverges slightly.

In the far field, although the laser spot on the retina has a constant minimum size and power entering the eye decreases due to beam divergence and falls in proportion to the inverse of the square of the distance (provided the laser beam diameter at the eye is larger than the diameter of the pupil).

The large variation of retinal intensity with distance from the laser close to and within the near field region is a factor which should be taken into account for proper hazard evaluation.

We decided on a graphical approach to hazard evaluation and a computer program was written in Fortran to give graphical output from a normal printer output terminal. Scale factors are selected by the program to give a graph out to, and slightly beyond, the region where the calculated retinal intensity falls below the permitted exposure level. An absolute limit of 2 kilowatts has been placed on the graphical range, because at such distances atmospheric scatter of the laser beam becomes appreciable and intensity predictions will have a large uncertainty factor. The predicted retinal intensity is plotted as a function of distance from a given laser. The "permitted exposure level" for accidental viewing of continuous wave lasers is also plotted. Graphs are plotted for three different pupil diameters corresponding to different ambient lighting conditions. The formula we adopted for the "permitted exposure level" is given by equation 1.

$$I = 1.0/d^2 \quad (\text{see reference 7}) \quad (1)$$

$I$  = permitted retinal intensity for accidental exposure to continuous wave lasers (watts/cm<sup>2</sup>).

$d$  = laser spot diameter on retina (millimetres).

Fig. 3 shows the permitted retinal intensity plotted together with the calculated retinal intensity for 3 different pupil diameters for the same laser as in Fig. 2.

It should be noted that the accuracy of any intensity calculation made is limited by the accuracy of the laser beam divergence which is adopted. We have found that for helium-neon lasers, beam divergence often varies up to 20% of the nominal value stated by the manufacturers. Similar variations can be present in the output power. A graph such as is shown in Fig. 2 is therefore applicable to the particular laser for which the beam parameters have been measured but it does not necessarily apply to other lasers of the same brand and model number.

In many cases the various prediction equations given in literature 1,2,3,4,5,6,7 do not agree with one another in all regions of the laser field, although the shape of the curves obtained is often similar.

Burnett<sup>2</sup> gives an equation for the near field divergence-limited case, which includes a term inversely proportional to distance from the laser, in an expression for the laser image diameter on the retina. When the near-field divergence limited case occurs at small distances from the laser this would make the image diameter calculated from Burnett's equation excessively

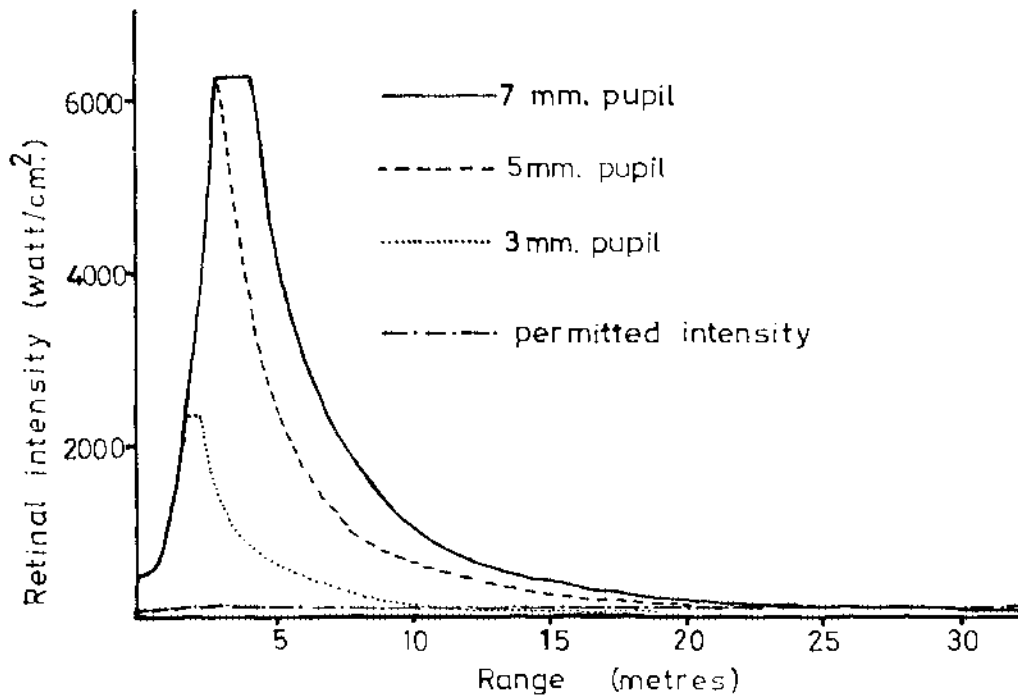


Fig. 3. RETINAL INTENSITIES FOR DIFFERENT PUPIL SIZES

large for realistic hazard analysis. Burnett's equation is correct for calculating the image size of the laser beam aperture on the retina for an accommodated eye, but incorrect as a worst case analysis. The worst case analysis of laser hazards should allow for the minimum possible beam spot size on the retina. In the near field divergence limited case the minimum spot size does not correspond with the image of the laser aperture on the retina, but occurs instead in the focal plane of the lens. Therefore the worst case occurs for an eye focused at infinity and not as Burnett initially assumes for an accommodated eye. In a later publication<sup>3</sup> Burnett gives a simpler equation which is more nearly correct. Burnett<sup>2</sup> gives a beam diameter which is different from that used by Solon et al<sup>1</sup>, although the difference is compensated somewhat by the different equations used to calculate retinal intensity. It would appear however that the method given by Solon et al is more accurate for hazard analysis in all cases and we therefore favour their method.

One factor which has not been allowed for in hazard analysis calculations of this type is that of power or energy profile across the beam. All hazard calculation methods given to date assume an even power distribution out to the beam edge. Many lasers have a power distribution which follows a gaussian pattern in the far field (TEM<sub>00</sub> mode). For these lasers and where the beam diameter is much larger than the diameter of the pupil the worst case retinal intensity may be 50% greater than that calculated when a uniform power distribution is assumed.

The hazard evaluation methods put forward by the various authors are not all in agreement. The investigation of the disagreement is difficult because in some cases assumptions and



approximations made have not been fully stated. Furthermore, the use of the formulae given in the literature may lead to errors of hazard analysis where assumptions and approximations are not fully stated or where the method of application of the formulae proposed is inappropriate.

#### References

1. Solon, Aronson and Gould, "Physiological Implications of Laser Beams" p. 1506, Science, Vol. 134, No. 3489, 10 Nov., 1961.
2. W.D. Burnett, "Laser Eye Hazard Evaluations", Research Report 50-RR-67-563, Sandia Laboratory, Albuquerque, Aug., 1967.
3. W.D. Burnett, "Evaluation of Laser Hazards to the Eye and the Skin", American Industrial Hygiene Association Journal Vol. 30, No. 6, Nov.-Dec., 1969, pp. 582-587.
4. G.W. Flint "Proceedings of the First Conference on Laser Safety", Martin Company, Orlando Florida, May, 1966.
5. J.W. Flood, "The Derivation of Maximum Permissible Exposures to Laser Radiation", Annals of Occupational Hygiene, July, 1967, p. 47.
6. J.F. Reedy, "Effects of High-Power Laser Radiation", Academic Press, New York, 1971.
7. British Ministry of Aviation, "Laser Systems Code of Practice" Shell Mex House, London, 1967.

МАТЕРИАЛЫ К ТОКСИКОЛОГИИ КАЛИФОРНИЯ-252

Заликин Г.А., Москалев Ю.И., Стрельцова В.Н., Жорова Е.С.,  
Семенов А.И., Петрович И.К., Трифонов В.И.

Институт биофизики Министерства здравоохранения.  
Москва, СССР.

Abstract

The report contains data on accumulation, distribution and elimination of various compounds of Cf-252 from the body of animals and characterizes its biological action after a single intravenous injection of acutely, subacutely and chronically effective doses. Symptoms of acute, subacute and chronic injury by Cf-252 are described and characteristics of its blastomogenic efficiency as a function of dose is given.

Введение

Вследствие высокой токсичности и все возрастающего практического применения, в последние годы все большее внимание привлекают вопросы токсикологии трансурановых элементов. Большинство исследований посвящено вопросам распределения и ускорения выведения этих изотопов из организма (1-5). Биологическое действие трансурановых элементов исследовано совершенно недостаточно, и в первую очередь, это касается Cf-252. В настоящей работе приводятся результаты экспериментов на крысах по распределению и биологическому действию различных доз Cf-252.

Материалы и методы

Опыты по распределению Cf-252 выполнены на белых беспородных крысах-самках весом 190±10,0. Изучали кинетику обмена азотно, соляно, лимоннокислой солей и полимерной формы Cf-252. Концентрация цитратного иона в растворе составляла 0,2%. Полимерную форму калифорния получали кипячением невесомых количеств препарата в растворе гидроокиси (NaOH), pH раствора 10,0. Удельная активность изотопов была в пределах 1,2-1,3  $\mu\text{C}$ -мл. Растворы изотопа вводили внутривенно в объеме 0,5 мл и перораль-

но при помощи металлического зонда в объеме 1 мл. Через определенные промежутки времени после введения радионуклида животных забивали декапитацией и определяли содержание активности в органах и тканях параллельно двумя методами по гамма-излучению в пробах не подвергавшихся никакой обработке кроме высушивания и по  $\alpha$ -активности в слое твердого люминофора после озоления и мокрого сжигания биопроб.

Опыты по изучению биологического действия калифорния поставлены на 450 крысах-самках в возрасте 6 месяцев со средним весом  $230 \pm 15$  г.. Азотнокислую соль  $Cf-252$  крысам вводили внутривенно (РН раствор  $\sim 3,0$ ) в дозах от  $0,064$  до  $0,0005 \mu C/g$ . На каждую дозу использовали от 13 до 90 крыс, а в качестве биологического контроля - 75 крыс. Изучали влияние  $Cf-252$  на продолжительность жизни, периферическую кровь, частоту и скорость развития опухолевых и неопухолевых форм отдаленных последствий. Величины ЛД50 для различных сроков определяли методом пробит-анализа. В случае необходимости вносили поправки на смертность и частоту соответствующего патологического процесса в контроле.

### Результаты исследования

Результаты опытов показали, что после внутривенного введения  $Cf-252$  преимущественно накапливается в печени и скелете (табл. I). Максимальный уровень накопления радионуклида в печени при введении растворов нитрата, хлорида и цитрата отмечен через 24 часа после инкорпорации изотопа и составил соответственно  $63,0$ ;  $56,5$  и  $47,3\%$ . Для гидроокиси максимум накопления в печени обнаружен через 1 час ( $75,7\%$ ).

При введении нитрата во все сроки опыта содержание изотопа в печени было выше, чем при введении хлорида и цитрата. Это различие четко выявляется во все сроки опыта, но особенно рельефно выражено на 32 и 64 сутки.

Однако несмотря на некоторые различия в уровнях отложения ритм выведения  $Cf-252$  из печени для растворимых солей тождественен. Растворимые соли  $Cf-252$  из печени выводятся фазно с эффективными периодами полувыведения равными в среднем  $2,47 \pm 0,09$ ;  $8,7 \pm 0,93$ ;  $43,4 \pm 5,0$ . Доли активности, выводимой с каждой фазой, соответственно равны  $0,3$ ;  $0,53$  и  $0,17$ .

Кинетика накопления разных солей  $Cf-252$  в органах имеет также свои особенности.

Через 1 час после введения содержание в скелете растворимых соединений  $Cf-252$  составило  $11-16\%$ , а уровень накопления гидроокиси был значительно ниже ( $3,7\%$ ). Максимальное содержание  $Cf-252$  при введении солей нитрата и хлорида приходится на 32 сутки и составляет  $30\%$  от введенного количества, а при введении изотопа в цитратном комплексе - на 16 сутки и составляет  $38,6\%$ . Максимальный уровень накопления  $Cf-252$  в скелете при введении полимерной формы составляет  $18,1\%$ .

Через час после введения растворимых солей содержание  $Cf-252$  в легких не превышает  $3\%$ , при введении полимерной формы в легких содержится  $13,8\%$ . Через 24 часа это различие исчезает.

В селезенке уровень накопления растворимых солей  $Cf-252$  находится в пределах  $1-1,5\%$ , а при введении полимерной формы оно достигает  $5,4\%$ . Высокое содержание полимерной формы  $Cf-252$  в селезенке отмечали до 32 суток опыта. В почках наблюдается обратная картина: содержание  $Cf-252$ , введенного в форме растворимых солей, составляет  $5,3-2\%$ , а гидроокиси  $0,6-1,2\%$ .

Обнаружено относительно более высокое содержание цитратного комплекса  $Cf-252$  в мышцах (до 10%), коже (до 11%) через 24 часа после введения изотопа. Самая высокая концентрация  $Cf-252$  отмечена в печени через час после введения изотопа. Для растворимых солей она достигала 7,8%, а для полимерной формы 10% от введенной активности. Во все сроки опыта концентрация  $Cf-252$  в I г костной ткани находилась на уровне двух процентов. Максимальная концентрация активности в скелете отмечена на 16 сутки опыта при введении цитрата  $Cf-252$ .

Высока концентрация  $Cf-252$  в цитовидной железе, через час после введения она составила 1,82%. В мышцах концентрация изотопа не превышала во все сроки опыта 0,08%.

Опыт с пероральным введением проведен с азотнокислым раствором  $Cf-252$ . В опыте использованы крысы равного возраста (табл.2)

Обнаружена отчетливая зависимость величины всасывания  $Cf-252$  от возраста. У крысят 7-дневного возраста величина всасывания составляет 1,1-1,6%, в возрасте месяца 0,21-0,25%, у взрослых 0,14-0,17%.

Резорбированная доля  $Cf-252$  в относительно больших количествах откладывается в скелете. Доля изотопа в скелете тем выше, чем моложе животные. Так у недельных крыс в скелете содержалось от 80 до 90% резорбированного  $Cf-252$ , у месячных от 64 до 86%, а у взрослых от 48 до 64%.

Результаты экспериментов по биологическому действию свидетельствуют о высокой токсичности  $Cf-252$ .

Средняя продолжительность жизни крыс при введении  $Cf-252$  в дозе 0,0005  $\mu\text{C}/\text{г}$  не отличается от контроля. В костной ткани и печени этих животных аккумулируются дозы излучения равные, соответственно, 257 и 30 рад.

Величины остро (LD50/30 - 0,012  $\mu\text{C}/\text{г}$ ) и подостро (LD50/60 - 0,011  $\mu\text{C}/\text{г}$ ) эффективных доз при инъекции  $Cf-252$  одинаковы, а хронически эффективные (LD50/360 - 0,006  $\mu\text{C}/\text{г}$ ) ниже их. По величинам остро-эффективных доз  $Cf-252$  токсичнее других трансураниевых элементов:  $Pu-239$ ,  $Am-241$  и  $Cm-244$  величины LD50/30 для которых равны, соответственно, 0,06; 0,11 и 0,11  $\mu\text{C}/\text{г}$  (1, 4, 5). Указанные различия в токсичности трансураниевых элементов наиболее отчетливо выявляются при сравнении величин остроэффективных доз. Хронически эффективные дозы (LD50/360) для  $Pu-239$  (0,011  $\mu\text{C}/\text{г}$ ) и  $Am-241$  (0,01  $\mu\text{C}/\text{г}$ ) в два раза ниже, а для  $Cm-244$  (0,0044  $\mu\text{C}/\text{г}$ ) практически такие же как для  $Cf-252$  (0,0055  $\mu\text{C}/\text{г}$ ). Причины большей эффективности  $Cf-252$  при введении больших доз не ясны и подлежат дальнейшему изучению.

Динамические наблюдения за составом периферической крови показали, что при введении  $Cf-252$  в дозах равных и выше 0,004  $\mu\text{C}/\text{г}$  у животных развивается типичное для лучевого заболевания угнетение органов кроветворения, наиболее отчетливо выраженное на 7-28 сутки (доза в костях ~ 35-125, печени ~ 90-177, селезенке ~ 18-65 рад) после инкорпорации изотопа. Степень угнетения кроветворения зависит от количества введенной активности и срока наблюдения. По сравнению с действием рентгеновских и гамма-лучей (5) лейкопения, вызванная  $Cf-252$ , характеризуется более медленным развитием, относительно большим угнетением миелопоэза и меньшим подавлением лимфопоэза.

По результатам гематологических исследований оценены уровни доз, вызывающих уменьшение количества лейкоцитов, лимфоцитов, нейтрофилов и эритроцитов на 50% на 3, 7, 14 и 30 сутки.

Величины ED 50% на 7-30 сутки для лейкоцитов, лимфоцитов и нейтрофилов оказались практически одинаковыми и в среднем равны 0,015; 0,017 и 0,018  $\mu\text{C}/\text{г}$ , а для эритроцитов 0,048  $\mu\text{C}/\text{г}$ .

По этому показателю эритроциты приблизительно в 3 раза более устойчивы. Изменения в количественном составе лейкоцитов, лимфоцитов и эритроцитов в течение первых двух недель опыта отсутствуют при введении  $Cf-252$  в дозах равных и ниже  $0,002 \mu C/g$  (доза в костях 37; в селезенке 15; печени 15 рад, а в случае нейтрофилов при дозах равных и ниже  $0,004 \mu C/g$ ).

При дозах, равных  $0,0005-0,002 \mu C/g$ , изменения в составе периферической крови отсутствуют на протяжении 16 месяцев наблюдения. При введении указанных количеств изотопа в костной ткани, печени, почках и селезенке за 16 месяцев аккумулируются дозы излучения, равные, соответственно, 129-1030; 15-119; 14-58 и 13-107 рад.

В поздние сроки у самок крыс развиваются опухоли молочных желез. У крыс, затравленных  $Cf-252$ , опухоли молочных желез обнаруживаются в более ранние сроки и в большем проценте случаев, чем у контрольных животных. При введении  $Cf-252$  в дозе  $0,008 \mu C/g$  первые опухоли молочных желез обнаружили через 4,7 месяца (у 2 из 19 живых крыс), при дозе  $0,004 \mu C/g$  через 5,1 месяца (у 2 из 50 крыс), при дозе  $0,002 \mu C/g$  через 4,7 месяца (у 1 из 82 крыс), при дозе  $0,001 \mu C/g$  через 7,5 месяцев (у 2 из 74 крыс), при дозе  $0,0005 \mu C/g$  через 10 месяцев (у 1 из 61 крысы) и в контроле через 7,5 месяцев (у 1 из 56 крыс).

На рис. 1 приведены данные о сроках обнаружения опухолей молочных желез у 10% крыс. В контроле указанную частоту опухолей молочных желез наблюдали через 16,6 месяцев, при введении  $Cf-252$  в дозах  $0,0005$ ;  $0,001$ ;  $0,002$ ;  $0,004$  и  $0,008 \mu C-g$ , соответственно, через 11; 9; 8 и 4,7 месяца. Зависимость срока появления опухолей молочных желез у 10% крыс от дозы  $Cf-252$  в полном логарифмическом масштабе имеет линейный характер. Время обнаружения опухолей молочных желез у 10% контрольных крыс (16,6 месяцев) соответствует средней продолжительности жизни этих животных (16,8 мес.). Экстраполированная из рис. 1 доза  $Cf-252$ , которая не повлияет на спонтанную частоту опухолей молочных желез и среднюю продолжительность жизни крыс равна  $0,0002 \mu C/g$ . При поражении животных этой дозой  $Cf-252$  за время жизни крысы в скелете аккумулируется поглощенная тканевая доза равная  $\sim 100$  рад, а в печени 12 рад.

Результаты аутопсий показали, что гибель животных в острой стадии интоксикации (до 30 суток) наступает в результате аплазии органов кроветворения, развития геморрагического синдрома и некробиотических нарушений в паренхиматозных органах.

Ведущими в картине острого лучевого поражения  $Cf-252$  являются нарушения костномозгового гемопоэза. У всех крыс, получивших  $Cf-252$  в количестве  $0,064-0,008 \mu C/g$  и павших на 9-11 сутки (доза в скелете 915-520 рад соответственно) костный мозг опустошен. В имбибированной эритроцитами строме костного мозга у этих животных располагались единичные кроветворные клетки необычного вида - более крупные, чем обычно, базофильные и хроматофильные эритробласты содержали избыточное количество цитоплазмы. Во многих эритроблестах наблюдалась плазматизация цитоплазмы (рис. 2а).

Необычно крупные клетки миелоидного ряда полигональной или неправильной формы имели неоднородную цитоплазму, где среди облаковидных базофильных комплексов располагались слабосифильные участки. Крупные ядра этих клеток были близки к ядрам гемопитобластов и миелобластов. Отдельные ядра имели бобовидные или палочкообразное строение. Встречались митозы с мостами, трехплоскостные митозы. Общим для всех этих новообразованных клеток был

диссонанс в степени зрелости ядра и цитоплазмы и крупные размеры.

У крыс получивших  $0,016 \mu\text{C}/\text{г}$   $\text{Cf-252}$  с течением времени клеточность костного мозга возрастала за счет формирования вышеописанных миелобластов. К 15 дню в костном мозге появлялись метамиелоциты и мегакарициты, но многие из них распадались. Все вышеизложенное свидетельствует не только о крайней напряженности восстановления костномозгового миело и эритропоэза у животных с острым поражением  $\text{Cf-252}$  близким по типу к реактивному ретикулезу, но и о дефективности и неполноценности репарации миелопоэза и в меньшей степени эритропоэза.

Костномозговая недостаточность в острой фазе поражения  $\text{Cf-252}$  не купировалась селезеночным кроветворением, так как селезеночный эритро и миелопоэз у животных-носителей остроэффективных количеств  $\text{Cf-252}$  в течение первых 30 дней отсутствовал. Лимфоидные фолликулы селезенки у этих животных были опустошены, в них сохранялись лишь центральные артерии и небольшое количество малых лимфоцитов, концентрически расположенных вокруг артерии на ретикулярном каркасе фолликула. Многие лимфоциты распадались. Клеточная популяция красной пульпы была представлена ретикулярными клетками, набухшим эндотелием синусов, гистиоцитами и множеством макрофагов с гемосидерином.

Реакция паренхиматозных органов в острой фазе поражения  $\text{Cf-252}$  характеризовалась некробиотическими изменениями в сочетании с проявлениями регенераторных процессов. У 9 из 27 подопытных крыс (33%) в течение первых 10-15 суток наряду с диффузным лизисом отдельных печеночных клеток развились очаги микронекрозов в центральных и интермедиарных частях долек и интенсивное крупнокапельное ожирение гепатоцитов в периферических отделах долек. Развитию некрозов гипоксической природы предшествовали циркулярные нарушения в виде резчайшего расширения центральных вен и капилляров центральной части дольки и спазма ветвей печеночной артерии в перипортальных прослойках. В эти же сроки проявлялся полиморфизм печеночных клеток — появлялись клетки крупнее обычных гепатоцитов с большим ядром.

В почках, наряду с явлениями повреждения структурных элементов органа: пикноз ядер эндотелия артерий, разрыхление стенок, вакуолизация мышечных клеток артерий, периваскулярные отеки, резкая гиперемия вен, спадение петель капилляров клубочков, увеличение просветов боуменовых капсул, зернистое перерождение эпителия извитых канальцев, к 9-22 дню наблюдались признаки восстановления эпителия извитых канальцев, о чем свидетельствует появление многоядерных клеток в извитых канальцах и клеток с необычно крупными ядрами.

Реакция эндокринных органов (щитовидной и паращитовидной желез, надпочечников, яичников) характеризовалась нарушениями кровообращения — расширением и полнокровием венозно-капиллярной сети, кровоизлияниями, отеками. В паренхиме эндокринных органов в острой фазе поражения наблюдалась утилизация гормонов без их воспроизведения. Так, например, щитовидные железы этих животных состояли в основном из мелких фолликулов, выстланных низким кубическим эпителием, не содержащим коллоида. В надпочечниках гипертрофированный корковый слой состоял преимущественно из однородных клеток с мелкопенистой цитоплазмой.

Хроническая фаза поражения  $\text{Cf-252}$  (дозы  $0,016-0,008 \mu\text{C}/\text{г}$ ) характеризовалась склеротическими изменениями внутриорганных артериальных ветвей, прецирротическими и цирротическими изменениями печени, сосудистым нефросклерозом разной степени выраженности, гипопластическим состоянием костномозгового кроветворения

с атипизмом регенерации и образованием опухолей костной и кроветворной ткани, опухолей печени, почек, молочных желез и эндокринных органов.

Для костномозгового гемопоэза животных с хроническим поражением  $Cf-252$  характерна меньшая, чем в норме, клеточность гемопоэза. В костном мозге чередовались гипопластические очаги с незрелым миелопоэзом с фокусами более зрелого миелопоэза и комплексами эозинофильных клеток (рис. 2б, в). Очаговые пролифераты из эозинофильных клеток встречались не у всех животных. Эритропоэз, представленный оксифильными эритробластами и нормобластами, несколько превышал миелопоэз.

Атипизм костномозгового миелопоэза у крыс с хроническим лучевым поражением  $Cf-252$  характеризовался сочетанием необычно крупных (особенно ядра) и необычно мелких клеток, диссоансом между степенью зрелости ядра и цитоплазмы (по цитоплазме клетки соответствовали метамиелоцитам и лейкоцитам, а по ядру промиелоцитам, миелоцитам или миелобластам и наоборот), разрывом между зрелыми и незрелыми клетками с отсутствием промежуточных форм (рис. 2г), уменьшением количества мегакариоцитов и обилием клеток с гиперсегментированными ядрами.

На вышеописанном фоне костномозгового гемопоэза при введении  $Cf-252$  в дозах 0,016 и 0,008  $\mu\text{C}/\text{г}$  у 3 из 12 крыс (25%) развился фиброз костного мозга, у 42% животных сформировалось предлейкемическое состояние с наличием *lygatus leucemicus* и обилием эозинофильных клеток с ядрами миелобластов (рис. 2в). У 2 из 12 крыс (17%) развился миелолейкоз, у 1 (8%) — эритробластов и у одной крысы (8%) хронический лимфолейкоз.

Приведенные выше данные свидетельствуют о нарушении процессов дифференцировки миелоидных клеток костного мозга при сохранности их пролиферативных потенций. В основе особенностей костномозгового кроветворения в хронической фазе поражений  $Cf-252$  помимо непрерывного прямого действия изотопа на популяции кроветворных клеток лежали и сосудистые изменения.

У 50% крыс с хроническим лучевым поражением имел место вкотопический гемопоэз в селезенке, печени и шейных лимфоузлах. У крыс с хроническим лучевым поражением лимфопоэз в лимфатических узлах осуществлялся с некоторым напряжением, о чем свидетельствовало превалирование больших лимфоцитов и лимфобластов в фолликулах.

Селезенка, за исключением случаев лейкоза и случаев с интенсивным незрелым компенсаторным гемопоэзом, как правило, была атрофирована. Строма ее во всех случаях содержала массу макрофагов с гемосидерином, что свидетельствовало о повышенной гибели эритроцитов у описываемых животных.

В хронической фазе поражения  $Cf-252$  наблюдались либо прецирротические изменения печени с гибелью комплексов печеночных клеток вследствие резчайшего ожирения или некроз, с разрастанием соединительной ткани, различными формами регенерации гепатоцитов, перипортальным склерозом, либо выраженный постнекротический цирроз печени (у 4 из 12 крыс) с формированием регенераторных аденом (рис. 3а) и холангиом. Особенностью регенерации печеночной паренхимы в этих случаях является отсутствие долькового строения и мозаичность новообразованной ткани, состоящей из комплексов клеток различного калибра и образование гепатоцитов с несколькими ядрами (рис. 3б), а также гепатоцеллюлярных симпластов. В генезе цирроза печени определенную роль играли вначале спазмы, а затем склеротические изменения ветвей печеночной артерии.

Аналогичная реакция имела место и в почках, где у всех крыс к двухсотому дню и позже возникал нефросклероз сосудистого генеза различной степени выраженности (рис. 3в). Необходимо подчеркнуть, что на всех этапах формирования нефросклероза в канальцах почек осуществлялась регенерация эпителия с формированием крупноядерных и многоядерных клеток (рис. 3г).

В щитовидной железе, паращитовидной железе, надпочечниках и гипофизе в хронической фазе поражения на фоне угнетения функциональной активности (отсутствие коллоида в щитовидной железе, паренхиматозная перестройка ее и т.д.) шла пролиферация новообразованного эпителия, формирующего очаговые гиперплазии и аденоматозные узелки.

Таким образом, в хронической стадии поражения  $Cf-252$  для всех изученных систем общим является снижение и искажение дифференцировочных потенциалов клеток с сохранением их пролиферативной способности, осуществляющейся на фоне недостаточности и дефектности сосудистой сети и склерозирования соединительной ткани. Осуществление регенераторных процессов в этих условиях реализуется всеми способами, присущими физиологической регенерации с доминированием явлений полиплоидизации.

Гиперпластические процессы наблюдали в молочной и паращитовидной железах. У части крыс, павших в отдаленные сроки, обнаружены различные воспалительные процессы: бронхопневмония, бронхоэктатическая болезнь, гнойный отит.

Опухолевые формы отдаленных последствий были представлены новообразованиями молочных желез, костей, легких, печени, почек, надпочечников и гипофиза.

При введении  $Cf-252$  в дозах 0,008; 0,004; 0,002; 0,001 и 0,0005  $\mu C/g$  суммарная частота опухолей у крыс, павших после 200 суток, составляла соответственно 54,5% (у 6 из 11 крыс), 36,0% (у 13 из 36 крыс), 51,7% (у 28 из 54 крыс), 53% (у 19 из 36 крыс) и 28% (у 9 из 32 крыс), а в контроле 8,3% (у 1 из 12 крыс). Чаще всего развивались опухоли молочных желез и костей. Остеосаркомы локализовались в различных костях скелета, но преимущественно в бедре. При введении  $Cf-252$  в дозах от 0,001 до 0,008  $\mu C/g$  у 127 крыс, павших после 200 дня, остеосаркомы развились у 18 крыс (14,1%) после аккумуляции в скелете к моменту гибели поглощенных тканевых доз от 494 до 3.240 рад. Средняя продолжительность жизни крыс с остеосаркомами зависела от введенной активности и при введении изотопа в дозах 0,008; 0,004; 0,002 и 0,001  $\mu C/g$  составляла, соответственно, 360; 373; 441 и 463 дня, а первые случаи гибели крыс с остеосаркомами при введении указанных доз изотопа наблюдали на 322, 329, 386 и 392 сутки.

Результаты работы позволяют оценить уровни доз  $Cf-252$ , не влияющие на среднюю продолжительность жизни ( $\leq 0,0005 \mu C/g$ ), количественный состав эритроцитов ( $\leq 0,004 \mu C/g$ ), лейкоцитов, лимфоцитов и нейтрофилов ( $\leq 0,002 \mu C/g$ ), появление опухолей молочных желез ( $\leq 0,0002 \mu C/g$ ) и остеосарком.

В заключение следует подчеркнуть, что  $Cf-252$  избирательно откладывается в печень и костную ткань. В отличие от других трансурановых элементов он обладает большей токсичностью и обуславливает более выраженное и длительное нарушение восстановительных процессов.



Таблица 1.  
Содержание калифорния-252 в органах и тканях крыс после внутривенного введения раствора различных бозов изотопов (% от введенного количества)

Органы, ткани	Время введения, сутки							
	1 час	1	4	8	16	32	64	
Кровь	1 <sup>1</sup>	22,3±5,9	1,4±0,1	0,7±0,0				
	2	12,6±0,6	1,6±0,2	0,4±0,1				
	3	9,8±2,9	0,4±0,1	-				
	4	2,2±0,5	0,5±0,1	0,5±0,1				
Печень	1	43,8±3,3	63,0±3,1	44,2±5,0	32,2±2,1	11,3±3,9	8,9±1,2	5,3±1,2
	2	38,0±1,4	56,5±1,2	34,2±7,7	34,5±2,8	20,2±3,6	3,5±0,2	2,3±0,0
	3	35,4±3,5	47,3±0,6	25,3±3,2	15,1±1,6	12,3±1,4	3,7±0,8	2,0±0,5
	4	75,7±1,3	54,0±3,1	43,2±3,7	11,4±3,1	15,5±3,7	6,4±2,2	2,6±0,4
Почки	1	2,3±0,3	2,6±0,1	3,6±0,8	1,8±0,2	1,0±0,3	1,2±0,1	1,9±0,1
	2	3,2±0,7	3,5±0,2	2,7±0,2	1,7±0,4	1,3±0,1	0,7±0,2	0,4±0,0
	3	5,3±1,2	4,0±0,3	3,9±0,3	2,3±0,2	1,6±0,1	1,2±0,2	0,8±0,0
	4	0,6±0,0	1,3±0,1	1,1±0,1	1,0±0,3	1,1±0,4	0,7±0,2	0,4±0,1
Селезенка	1	1,2±0,1	1,7±0,2	1,3±0,0	1,7±0,1	0,2±0,0	1,5±0,3	1,2±0,2
	2	1,0±0,2	1,4±0,1	0,9±0,2	1,7±0,4	1,0±0,1	0,7±0,2	0,7±0,2
	3	1,9±0,2	0,7±0,1	0,6±0,2	0,4±0,0	0,4±0,1	0,5±0,0	0,3±0,1
	4	4,6±1,1	3,6±0,4	5,4±0,5	2,9±1,2	2,7±0,7	0,7±0,3	1,0±0,4
Легкие	1	3,0±0,2	0,4±0,0	0,3±0,0	0,2±0,0	0,1±0,0	0,2±0,0	0,1±0,0
	2	3,2±0,7	0,5±0,2	0,5±0,0	0,6±0,1	0,2±0,0	0,1±0,0	0,1±0,0
	3	1,2±0,2	0,4±0,1	1,1±0,1	0,2±0,0	0,2±0,0	0,2±0,0	0,1±0,0
	4	13,8±1,3	0,9±0,1	0,9±0,1	0,3±0,0	0,3±0,1	0,2±0,1	0,1±0,1
Скелет	1	11,1±0,7	19,0±0,5	20,9±0,5	26,1±1,3	17,9±3,3	29,6±3,7	28,1±2,2
	2	12,6±2,4	22,3±2,3	21,3±2,0	25,1±1,3	28,5±1,6	30,4±2,4	27,0±1,7
	3	16,1±1,9	24,6±1,5	19,6±1,8	29,4±3,6	16,6±2,1	34,7±2,3	26,6±1,8
	4	3,7±0,3	10,4±0,4	18,1±2,4	16,6±1,0	16,0±1,0	14,6±2,1	15,6±3,0
Мозги	1	8,3±0,3	3,7±0,4	2,8±0,0	1,8±0,1	1,4±0,6	2,0±0,6	3,0±0,7
	2	3,5±0,6	2,7±0,4	4,1±1,0	2,1±0,5	3,4±0,3	1,9±0,5	1,3±0,5
	3	10,0±1,8	9,6±1,0	6,3±0,5	5,4±0,6	5,5±1,2	4,0±0,1	2,7±0,3
	4	3,6±0,5	3,4±0,4	1,3±0,4	3,8±1,9	3,0±0,6	-	-
Хвост	1	4,7±0,9	3,7±0,9	2,8±0,0	4,1±0,4	1,9±0,0	2,4±0,6	-
	2	5,2±0,8	4,7±0,5	3,5±0,8	2,7±0,6	2,2±0,1	-	0,31
	3	8,2±0,4	11,0±0,7	10,2±2,4	5,4±1,2	1,00	3,7±0,7	4,1±0,7
	4	3,1±0,0	4,8±0,7	-	5,0±0,0	-	-	-

1) I - альфа, 2 - дельта, 3 - гамма, 4 - гидроокись.

Таблица 2

Содержание калифорния-252 в органах крыс различного возраста после перорального введения раствора азотно-кислотной соли изотопа (% введенной активности)

Орган	Время после введения		
	1 сутки	4 сутки	8 сутки
<b>Возраст крысят 7 дней</b>			
1. Легкие	0,015 ± 0,003	0,005 ± 0,0007	0,0034 ± 0,0013
2. Селезенка	0,0053 ± 0,0012	0,0046 ± 0,0008	0,0038 ± 0,0003
3. Печень	0,070 ± 0,020	0,045 ± 0,0034	0,021 ± 0,002
4. Почки	0,043 ± 0,0121	0,027 ± 0,0012	0,033 ± 0,005
5. Скелет	0,538 ± 0,130	0,710 ± 0,018	0,460 ± 0,062
<b>Возраст крысят 30 дней</b>			
1. Легкие	0,0037 ± 0,0014	0,0018 ± 0,0002	0,0022 ± 0,0003
2. Селезенка	0,0028 ± 0,0006	0,0015 ± 0,0001	0,0016 ± 0,0001
3. Печень	0,027 ± 0,0013	0,013 ± 0,0014	0,0067 ± 0,0008
4. Почки	0,012 ± 0,004	0,0053 ± 0,0001	0,0023 ± 0,0002
5. Скелет	0,079 ± 0,015	0,088 ± 0,0074	0,072 ± 0,012
<b>Взрослые крысы</b>			
1. Легкие	0,0013 ± 0,0002	0,0012 ± 0,0005	
2. Селезенка	0,0010 ± 0,0001	0,0007 ± 0,0001	
3. Печень	0,045 ± 0,003	0,017 ± 0,001	
4. Почки	0,0062 ± 0,0003	0,0049 ± 0,0003	
5. Скелет	0,048 ± 0,006	0,043 ± 0,006	

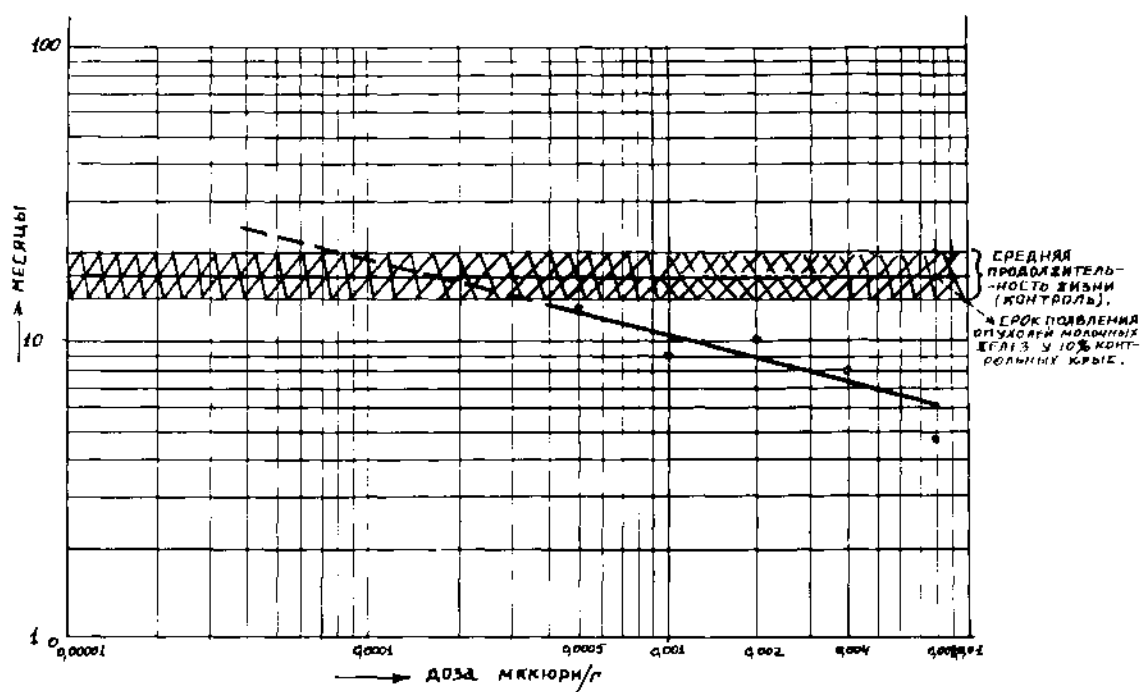


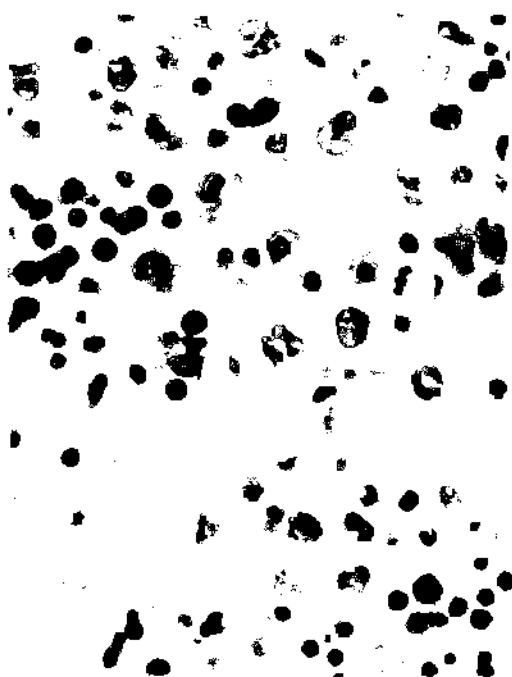
Рис. I. Зависимость срока обнаружения опухолей молочных желез у 10% живых крыс от дозы Cf-252.



Рис. 2. Состояние гемопоэза у крыс с поражением Cf-252. Гематоксилин-эозин.

- а) Гемопоэз в опустошенном костном мозге. Крупные ретикулярные клетки, атипичные эритробласты. 0,064  $\mu\text{C}/\text{г}$ . 9 сутки.  $\times 1000$ .
- б) Гинопластический костный мозг. Плазматизация эритробластов. Необычно крупные атипичные миелобласты и лейкоциты. 0,008  $\mu\text{C}/\text{г}$ . 322 сутки.  $\times 900$ .
- в) Очаг из атипичных эозинофилов с ядрами типа миелобластов и миелопитов. 0,016  $\mu\text{C}/\text{г}$ . 178 сутки.  $\times 900$ .

а



б



в

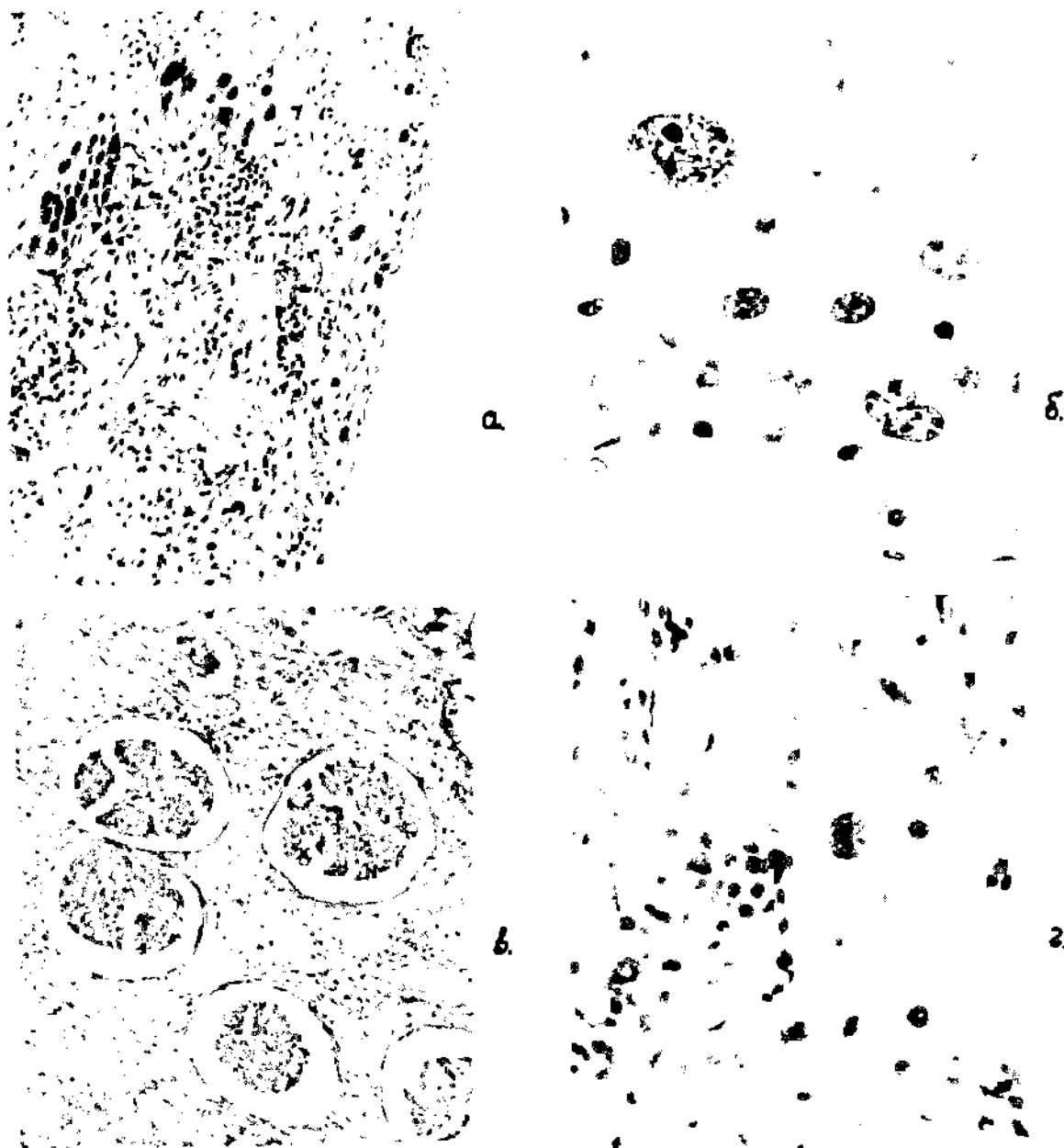


Рис. 3. Склеротические и восстановительные процессы в органах крыс с хроническим лучевым поражением Cf-252.

- а) Постнекротический цирроз печени с узловатыми регенератами.  
0,008  $\mu\text{C}/\text{г}$ . 500 суток. х 400. Гематоксилин-эозин.
- б) Регенерация в цирротической печени. Полиплоидные гепатоциты.  
0,016  $\mu\text{C}/\text{г}$ . 500 суток. х 400. Гематоксилин-эозин.
- в) Нефросклероз. 0,008  $\mu\text{C}/\text{г}$ . 178 суток. х 200. Моллери.
- г) Формирование необычно крупных эпителиальных клеток с некробиозом эпителия в склеротической почке.  
0,016  $\mu\text{C}/\text{г}$ . 178 сутки. х 500. Гематоксилин-эозин.

## Литература

1. Булдаков Л.А., Любчанский Э.Р., Москалев Ю.И., Нифатов А.П.  
Проблемы токсикологии плутония-239.  
Атомиздат, Москва, 1969.
2. Заликин Г.А., Москалев Ю.И., Петрович И.К.  
Распределение и биологическое действие америция-242.  
Радиобиология 8(1), 65, 1968.
3. Заликин Г.А., Москалев Ю.И., Петрович И.К., Рудницкая Э.И.  
Биологическое действие америция-241.  
Радиобиология 9(4), 599, 1969.
4. Москалев Ю.И., Рудницкая Э.И., Заликин Г.А., Петрович И.К.  
Биологическое действие азотнокислого нептуния-237.  
Гигиена и санитария 36 (2), 42, 1971.
5. Москалев Ю.И., Петрович И.К.  
Реакция периферической крови в зависимости от условий  
лучевого воздействия.  
Радиобиология 01, № 6, 881, 1971.

## ВЛИЯНИЕ ФТОРИРОВАННОЙ ПИТЬЕВОЙ ВОДЫ И КАЛЬЦИНИРОВАННОЙ ДИЕТЫ НА РАДИОРЕЗИСТЕНТНОСТЬ ЖИВОТНЫХ ПРИ ВНЕШНЕМ ГАММА-ОБЛУЧЕНИИ

В. А. Книжников, В. А. Грозовская  
Институт биофизики Министерства здравоохранения СССР  
Москва, Союз Советских Социалистических Республик

### Аннотация

In experiments on 2,016 albino rats it has been found that continuous high levels in the intake of fluorine with drinking water and calcium with the diet increases the animal survival to 1.5 - 2 times after single or fractional total gamma or x-ray irradiation in the doses of LD<sub>10/30</sub>-LD<sub>70/30</sub>. In experiments on 205 rats it has been found that fractional irradiation in the summary doses of 200 R doesn't cause radiation disease but shortens the animal life by 10%. The addition of calcium and fluorine to the diet considerably neutralizes the negative influence of the irradiation on duration of life.

Ранее было показано, что уровень поступления кальция с диетой и концентрация фтора в питьевой воде способны влиять на радиорезистентность экспериментальных животных при общем внешнем гамма- и рентгеновском облучении /1, 2/.

Целью исследований, описываемых в настоящем сообщении, было получение дополнительных материалов, характеризующих влияние кальция и фтора на выживаемость облученных животных. Полученные материалы сопоставляются с данными ранее проведенных исследований /1, 2/ с целью выведения некоторых закономерностей влияния кальция и фтора на радиорезистентность при однократном или дробном облучении в различном диапазоне доз.

### Влияние кальция и фтора на выживаемость крыс после облучения в остропоражающих дозах

Эксперимент с профилактическим добавлением кальция и фтора. Исследование включало в себя два опыта, поставленных на 346 крысах-самцах линии Вистар, которые до облучения в течение 60 дней получали виварийный рацион с добавлением углекислого кальция к кормовым брикетам и фтористого натрия к питьевой воде. Исходное содержание фтора в воде контрольных животных (из московского водопровода) составляло 0,1-0,2 мг/л; содержание кальция в кормовых брикетах - 40 мг на животное в сутки. Содержание кальция и фтора в пище и воде опытных групп животных довели до величин, несколько превышающих общепринятый физиологический оптимум (160 мг/сутки и 3,5 мг/л соответственно).

Материалы по выживаемости животных после облучения в дозе 700 р (первый опыт) и 600 р (второй опыт) представлены в табл. I.

Таблица I

Влияние на выживаемость однократно облученных крыс профилактического добавления кальция и фтора к диете

№ опы- тов	Доза, рент- ген	Группа животных	Содержание в диете		Коли- чество крыс в группе	Вес животных перед облуче- нием, г	Пало за 30 дней после облуче- ния, %
			Фтор, мг/л	Кальций, мг			
1	700	Кальциевая	0,2	160	29	241	17,2
		Фторная	3,5	40	33	233	33,3
		Контрольная	0,2	40	33	226	43,8
2	600	Кальциевая	0,2	160	80	232	17,5
		Фторно- кальциевая	3,5	160	67	227	16,0
		Контрольная	0,2	40	104	225	37,5

ПРИМЕЧАНИЕ: 1. После облучения добавление кальция и фтора к диете было прекращено. 2. Срок диеты до облучения - 60 дней. 3. Вид воздействия - однократное общее гамма-облучение на установке ЭГО-2 при мощности дозы 400 р/мин.

Добавление кальция к диете в первом опыте повысило выживаемость примерно на 27%, во втором - на 20% (по сравнению с контролем, выживаемость в котором принята за 100%). Обогащение воды фтором в первом опыте также повысило выживаемость (на 10%), однако эффект от фтора был менее выраженным, чем от кальция. Обогащение во втором опыте диеты одновременно фтором и кальцием дало наилучший в данном опыте эффект, однако, говорить о суммации защитных эффектов обоих элементов сами по себе материалы данного опыта не позволяют. Можно отметить также, что профилактическое назначение кальция и фтора (т. е. только до момента облучения) оказало почти такой же защитный эффект, как и длительное применение данных элементов до и после облучения, о котором сообщалось ранее /1, 2/.

Эксперимент с добавлением кальция и фтора к физиологически полноценной диете. Опыт был поставлен на 219 беспородных белых крысах-самцах, которых в течение двух месяцев до облучения и затем до конца опыта содержали на экспериментальной диете. Поступление кальция контрольным животным составляло около 100 мг/сутки, "кальциевым" и "фторно-кальциевым" - около 200 мг/сутки. Содержание фтора в питьевой воде фторно-кальциевой группы поддерживалось на уровне 4 мг/л; в воде двух других групп - 0,45 мг/л. Результаты опыта представлены в табл. 2.

Материалы табл. 2 указывают на небольшое, однако, статистически достоверное повышение радиорезистентности у животных, содержащихся на кальциевой и фторно-кальциевой диетах. Величина защитного эффекта в данном опыте была заметно ниже, чем в проведенных ранее опытах с аналогичной дозой облучения. При этом особенно снизился эффект от дополнительного введения фтора. Очевидно, подобное снижение эффекта связано с тем, что в исходном рационе контрольных животных содержание кальция и фтора было более

высоким, чем в ранее проводившихся опытах /1, 2/. При этом, по-видимому, для повышения радиорезистентности достаточно добавления к воде нескольких десятых долей миллиграмма фтора на литр, а дальнейшее обогащение воды фтором сверх 0,5 мг/л дает мало эффекта. Что касается кальция, то при исходном рациона, содержащем достаточное количество этого элемента, дальнейшее обогащение рациона оказывает меньше эффекта, чем в случаях, когда исходный рацион содержит кальций на уровне нижней границы физиологической нормы. Практически важно, что и при полноценной по кальцию диете его дополнительное введение в рацион оказывает статистически достоверный радиозащитный эффект.

Таблица 2

Влияние на выживаемость однократно облученных крыс дополнительного внесения кальция и фтора в диету, содержащую эти элементы в количествах, близких к физиологическому оптимуму

Группа животных	Содержание в диете		Количество крыс в группе	Вес животных, г		Пало за 30 дней после облучения, %	Достоверность разницы по выживаемости, P
	Фтор, мг/л	Кальций, мг		В начале опыта	После облучения		
Кальциевая	0,45	220	71	176,3±2,3	281±4,1	42,3	0,01
Фторно-кальциевая	4,0	220	74	176,3±1,4	284±4,4	40,5	0,01
Контрольная	0,45	100	74	176±1,4	282±2,7	51,3	-

ПРИМЕЧАНИЕ: Доза облучения - 750 р при мощности 29,3 р/мин.

Некоторые общие закономерности влияния кальция и фтора на радиорезистентность в зависимости от величины остропоражающей дозы. В табл. 3 приведены в обобщенном виде частично ранее опубликованные материалы всех экспериментов по изучению влияния дополнительного введения кальция и фтора на течение острого лучевого поражения. Опыты, вошедшие в табл. 3, проведены по аналогичной схеме с длительным содержанием животных до облучения на диете. Поступление кальция с суточным рационом опытным животным - 160 мг; концентрация фтора в воде - 3,5 мг/л. Особенности отдельных экспериментов видны из таблицы и примечаний к ней.

Материалы табл. 3 позволяют отметить, что повышение радиорезистентности имело место как при добавлении одного кальция к рациону, так и кальция со фтором. В последнем случае эффект, как правило был наиболее выраженным. При дозах облучения выше СД70/30 эффект не был четким и практически отсутствовал. Во всем диапазоне доз ниже СД70/30 эффект был существенным и статистически достоверным. Практически важно, что диета с повышенным содержанием кальция, а также кальция и фтора повышала выживаемость животных не только в первые 30 дней после облучения, но и в течение длительного последующего периода. Важно также отметить, что влияние кальция и фтора на радиорезистентность имело место при относительно небольших вариациях уровня поступления этих элементов с диетой, которые не выходят за рамки колебаний, имеющих место в реальной действительности /3, 4/.



Таблица 3

Влияние уровней кальция и фтора в диете  
на радиорезистентность крыс  
в остропоражающих дозах

	Доза облучения		Группа животных	Число животных в группе	Пало за 30 дней после облучения, %	Срок выживания 50% облученных крыс, дней
	По биологическому эффекту	Экспозиционная, рентген				
1	Менее СД <sub>20/30</sub>	670-920 дробно	Фторно-кальциевая	90	4	202
			Контрольная	105	10	164
2	СД <sub>30/30</sub> СД <sub>50/30</sub>	600-700	Кальциевая	60	33	88
			Фторно-кальциевая	74	24	119
			Контрольная	73	49	31
3	СД <sub>51/30</sub> СД <sub>70/30</sub>	600-850	Кальциевая	246	43	48
			Фторно-кальциевая	216	37	70
			Контрольная	246	58	20
4	Более СД <sub>70/30</sub>	750-900	Кальциевая	79	76	9
			Фторно-кальциевая	155	81	13
			Контрольная	107	87	10

ПРИМЕЧАНИЕ: 1. По 2-4 опыта в каждой серии. 2. При дробном облучении животные получали дозу за 5 сеансов в течение одной недели. 3. Мощность источника облучения - от 156 до 717 р/мин. 4. Содержание кальция в суточном рационе контрольных животных - 40 мг, концентрация фтора в их воде и воде животных кальциевых групп - 0,2 мг/л.

Влияние кальция и фтора на продолжительность жизни крыс,  
дробно облученных в подострых дозах

На практике интерес представляют дозы, которые значительно меньше использованных в изложенных выше опытах, т. е. дозы, укладывающиеся в нормативы радиационной безопасности и встречающиеся в реальных условиях работы персонала (рентгенологов и радиологов, работников атомных электростанций и т. д.) Для определения возможного влияния кальция и фтора на эффекты от облучения в указанных дозах учитывалась продолжительность жизни животных, а также возникновение у них в отдаленном периоде злокачественных новообразований. Индуцирование последних в опытах на животных, как известно, требует или больших доз облучения, намного превосходящих профессиональные уровни, или постановки опытов на очень большом числе животных. Вследствие этого в качестве основного показателя благополучия животных и отдаленных последствий облучения мы использовали данные по продолжительности жизни. Сокращение продолжительности жизни при воздействиях на уровня 100-400 бэр удавалось регистрировать даже в опытах на небольшом числе животных /5, 6/.

В опыте использовали 205 беспородных белых крыс-самцов с начальным весом 228±2,0 г. Животные были разбиты на 4 группы и помещены на обычную виверную диету, отличающуюся в группах содержанием кальция и фтора, как это было в описанных выше опытах (т. е. содержание кальция в опытных группах 160, в контрольных - 40 мг; фтора - 3,5 и 0,2 мг/л соответственно).

Уровень кальция и диете варьировали за счет внесения в бри-

каты очищенного углекислого кальция, а фтора — путем добавления в воду фтористого натрия. Крысы получали указанный рацион в течение 2 месяцев до облучения и далее до конца жизни.

Облучение производили дробно в суммарной дозе 200 рентген (8 сеансов с интервалом по 24 часа). Одна из групп животных не облучалась и служила биологическим контролем. В течение опыта животных регулярно взвешивали, осматривали, пальпировали, павших вскрывали, и измененные органы подвергали гистологическому исследованию. Для выявления лейкозов делали отпечатки селезенки и костного мозга.

Облучение не вызвало задержки в прибавке веса ни в начале, ни в конце опыта. По данным осмотра, животные после облучения признаков лучевой болезни не имели: сохранили обычную подвижность, хороший аппетит, блестящую шерстку. Наблюдение показало также, что длительное, пожизненное, содержание животных на диете с повышенной концентрацией кальция и фтора не повлияло отрицательно на их вес и состояние, определяемое при внешнем осмотре. Исключением явилась лишь специфическая для легких форм флюороза исчерченность эмали нижних зубов у части крыс, получавших воду с повышенным содержанием фтора.

Средняя продолжительность жизни животных иллюстрируется данными табл. 4.

Таблица 4

Средняя продолжительность жизни животных  
(в днях после облучения)

Группы животных	Кальциевая	Фторно-кальциевая	Контрольная	Биоконтрольная
Все животные	453±17	458±20	418±21	458±34
Все, пережившие 200 дней после облучения	457±16	487±15	448±17	497±29
Пережившие 200 дней; но времени гибели 90% в группе	443±13	472±13	431±14	494±28
Те же; достоверность разницы между группами, P	-	< 0,05	< 0,05	

Наибольшего внимания в табл. 4 заслуживают данные по последней категории животных, т. е. по крысам, пережившим 200 дней и составившим 90% погибших. Эта категория представляется нам наиболее важной, поскольку оставшиеся в каждой группе единичные экземпляры "долгожителей" не характеризуют устойчивость основной массы животных. Анализ материалов по этой категории животных позволяет констатировать, что облучение вызвало существенное сокращение продолжительности жизни (разница между биоконтрольной и контрольной группами статистически достоверна).

Обогащение диеты кальцием и фтором увеличило жизнеспособность животных (разница между контрольной и фторно-кальциевой группами также статистически достоверна).

Основными причинами гибели животных в отдаленном периоде были нарушения сосудистой проницаемости (кровоизлияния во

внутренних органах, экссудаты в грудной и брюшной полостях), а также пневмонии и абсцессы, развившиеся, по-видимому, на почве сниженной облучением иммунореактивности.

Среди животных зарегистрированы единичные спонтанные опухоли различных органов и тканей, включая молочные железы. Однако небольшое количество опухолей и возникновение их у 12% необлученных животных не позволяет, как это и предполагалось, связать их возникновение с лучевым воздействием или диетой. Несколько более закономерные данные получены по лейкозам (табл. 5).

Таблица 5  
Лейкозы среди животных, переживших 200 дней

Показатели	Группы животных	Кальциевая	Фторно-кальциевая	Контрольная	Биоконтрольная
Процент животных с лейкозами		1,7	0	5,8	0
Время от облучения до гибели (дней)		797	-	350	-

Ввиду небольшого числа наблюдений закономерность проявления лейкозов по группам не представляется полностью убедительной.

Возможные механизмы защитного действия фтора и кальция. Защитное действие фтора при острой лучевой болезни мы считаем возможным объяснить способностью фтора стимулировать кроветворение /7/. Благоприятное влияние кальция при остром лучевом поражении, очевидно, может быть обусловлено известным свойством кальция нормализовать сосудистую проницаемость.

Что касается защитных эффектов, проявившихся в отделенном периоде после облучения малыми дозами, оно, возможно, связано со свойством фтора как микроэлемента повышать иммунореактивность /8/. Влияние кальция, по-видимому, может иметь двойной характер. Во-первых, здесь представляется вероятным специфическое защитное влияние этого элемента на лучевой мутагенез, который был обнаружен в опытах на культуре тканей /9/. Во-вторых, весьма реальным представляется неспецифическое действие кальция, которое можно ожидать на основании сообщений в литературе о положительном влиянии на продолжительность жизни животных дополнительных количеств кальция /10/, а также о более благоприятных показателях смертности среди населения, потребляющего жесткие питьевые воды, отличающиеся повышенной концентрацией кальция /11/.

#### Выводы

1. Длительное повышенное поступление фтора и кальция с диетой существенно повысило радиорезистентность и выживаемость экспериментальных животных после облучения в диапазоне остропоражающих доз от СД<sub>20/30</sub> до СД<sub>70/30</sub>.

2. Содержание животных на диете с повышенными количествами кальция и фтора способствовало существенному уменьшению отрицательного влияния малых доз облучения на продолжительность жизни.

3. Влияние кальция пищи и фтора питьевой воды на радиорезистентность проявлялось при относительно небольших колебаниях в уровне их поступления, встречающихся в естественных условиях и не выходящих за границы физиологической нормы.

### Литература

1. Книжников В.А., Грозовская В.А. Радиобиология, 1968, т.8, № 3, 429.
2. Книжников В.А., Грозовская В.А. Радиобиология, 1970, т. 10, № 4, 617.
3. Габович Р.Д. Фтор и его гигиеническое значение. М., Медгиз, 1957.
4. Книжников В.А., Степанов Ю.С. и др. Поступление продуктов испытаний ядерного оружия населению Советского Союза с пищевым рационом и водой в 1963-64 гг. М., Атомиздат, 1965. Документ ООН А/АС 82/С, 1083.
5. Орешкин И.Г. и др. В кн. "Отделенные последствия лучевых поражений". М., Атомиздат, 1971, стр. 92.
6. Москалев Ю. И., Петрович И.К. Там же, стр.517.
7. Книжников В.А., Белоусова О.И., Грозовская В.А. Гигиена и санитария, 1965, № 10, 12.
8. Книжников В.А. Журнал микробиологии, эпидемиологии и иммунологии, 1958, 5.
9. Рукавишников Ю.М. В кн. "Тез. докл. симпозиума по проблемам радиочувствительности на молекулярном, клеточном и организменном уровнях". Новосибирск, 1966.
10. Campbell H. L., Sherman H. C. Am. J. Physiol., 1945, 144, 717
11. Schroeder H. A. J. Amer. med. Ass., 1960, No 178, v. 17, 1902-1908.

POTENTIATION OF  $^{90}\text{Sr}$  STRONTIUM BIOLOGICAL ACTION  
ON AN ORGANISM BY AGRICULTURAL CHEMICALS

Mukhin, I.J.; Borovikova, N.M.;

Nagovitsina, L.I.

Kiev (Ukr. SSR)

Analysis of investigations on determining the mode of binary mixture ( $^{90}\text{Sr}$  and DDT) action at its long-term administration daily to an organism showed that for this purpose quantitative data only are insufficient, especially in those cases when integral findings and possible effects are criteria. Accordingly, to elicit difference between components and binary mixture effect rate on the weight increase, duration of life and intensity of tumour output by quantitative evaluation only is of great difficulty. The quantitative evaluation with due regard for qualitative peculiarities of occurring effects and time of their appearance permits to elicit the  $^{90}\text{Sr}$  biological action potentiation by the chemical (DDT) at their combinative long-term administration to an organism.

Presence of  $^{90}\text{Sr}$  and DDT in the environment, their high migratory capacity and the possibility of combined human intake caused the necessity for evaluation of consequences not only isolated  $^{90}\text{Sr}$  and DDT effect on an organism but combinative disturbance forms. The purpose of the given study was to define the mode of binary mixture ( $^{90}\text{Sr}$  and DDT) action when it was administered orally on a daily schedule to animals. 200 white rats inbred with the primary weight of  $140 \pm 15$  g were used in experiments. Compounds were cannulated in rats i.p. in such dosages: 0.3  $\mu\text{Ci}$  of  $^{90}\text{Sr}$  (group I), 0.2 mg of DDT (group II), 0.8 ml of binary mixture containing 0.3  $\mu\text{Ci}$  of  $^{90}\text{Sr}$  and

0.2 mg of DDT (group III) and 0.3 ml of vegetable oil (group IV).

During 186 days 55.8  $\mu$  Ci of  $^{90}\text{Sr}$  (groups I and III), 37.2mg of DDT (groups II and III) and 55.8 g of vegetable oil were administered to animals.

#### Materials and Methods

$^{90}\text{Sr}$  storage in rat skeleton for groups I and III resulted in increasing not only dose rate from 4.0 up to 12.8 rads per day (by the 30th day and the 186th day after radioactive label injection, respectively) but the adsorbed dose per animal skeleton from 71.5 rads (by the 30th day after injection) up to 1482 rads by the last injection day.

In the further study (up to the death of animals for groups I and III) the dose rate reduced gradually. The dose by the above date was 6.6-6.9 rads/day.

By the end of study the adsorbed dose was 4,488 (group III) and 4,953 rads (group I).

During the study period the

rats weight for groups I and III was lower than for controls. At the same time rats of group III had invariably the minimum weight. Annual increase of animals weight for this group was  $100 \pm 10$  g and for groups I and II was  $130 \pm 10$  g.

For the last study weeks the rats weight for group III was  $180 \pm 15$  g, and for group I and for group II was  $250 \pm 20$ g. It is characteristic that the rats weight increase for group III was stopped by the 220th day, for group I and II by the 360th day, for controls only by the 450th day.

Duration of study rat life was 580 days for group I, 462 days for group II, 330 days for group III, and 720 days for controls.

Over a two-year study period there was only one hypophysis innocent tumour in the control group. The tumour output was diagnosed by the 760th day of the study.

The primary tumour for group I was observed by the 488th day. In the following 145 days 4 tumours of internal organs, one osteosarcoma, and a milk gland fibroadenoma have also been diagnosed. Additionally, at morphological studies of marrow and thigh bones 2 presarcomas (by the 575th day) and 3 leucosis (by the 488th day) were found. The adsorbed dose per skeleton was 3,900 rads by the time of the primary tumour and 2 leucosis revealed. Eighteen rats survived before the primary tumour determined. The primary tumour for group III was diagnosed by the 300th day when the adsorbed dose per skeleton was 3,000 rads.

In the following 333 study days 5 tumours of internal organs, 2 osteosarcomas, 4 presarcomas, and 5 leucosis for group III were observed. There was no tumour for group II during the study period.

Quantitative erythrocytes changes for group I and III were ob-

served during the study period. At the same time thrombocytes and reticulocytes increase were primarily observed. By the end of the study second month marked reticulocytopenia and thrombocytopenia were developed.

Quantitative leucocytes shifts were revealed by the 30th day after  $^{90}\text{Sr}$  and binary mixture injection.

Leucopenia instead of leucocytosis was developed by the 100th day. Changes of the number of morphous blood elements for group I were 15-20% higher than for group III

During the first 3 study months peripheral blood picture did not differ when compared with controls.

In the following months the number of white and red blood cells for group III was 15-18% lower than for controls.

Activity of serum alkaline phosphatase, osseous tissue

and internal organs for groups I-III was changed homogenously having difference only in quantitative relation.

During the study period (the 30th, 100th, 187th, and 450th day) activity of serum alkaline phosphatase, osseous tissue, and organs for group III was higher than for group I in spite of DDT (group II) constantly caused the intensive enzyme inhibition.

#### Results

It is impossible to determine the mode of binary mixture action due to only quantitative evaluation of integral findings (animal weight and duration of life). At the same time it is possible to speak about the chemical effect on  $^{90}\text{Sr}$  potentiating action taking into consideration temporary evaluation on each of the findings. Thus, discontinuance of weight increase for group III has become 230 days earlier than for the control group and 50 days earlier than for both groups I and II.

The death 50% of rats for group III has become 440 days earlier than for the control group and 10 days earlier than for both groups I and II.

It is impossible to demonstrate that the chemical potentiates the  $^{90}\text{Sr}$  action on hematosi state by evaluating only the quantitative shifts in morphous blood elements for groups I, II, and III. However, leucosis frequency for group III (5 cases; 3 of them are in combination with internal organs tumours) and for group I (3 cases; 2 of them are in combination with tumours) can be evidence for the assertion of intensification  $^{90}\text{Sr}$  action on hematosi organs.

The number of malignant tumours of internal organs and flexible tissues for group I and III was identical. Meanwhile, 4 numerous tumours of 6 (group III) and 1 from 6 (group I) were occurred in rats.

Additionally, 5 rats of 6



(group III) and 3 rats of 6 (group I) had tumours in combination with other effects.

In a year after  $^{90}\text{Sr}$  and DDT injection was stopped, 3 rats (group I) and 6 (group III) were died from malignant neoplasms and leucosis. Rats for group I and III compared showed no significant differences in the number of osteosarcomas (one for group I, two for group III). However, 2 presarcomas were found for group I, 4 for group III.

Thus, it is insufficient to have available information about the quantitative evaluation of arising effects to determine the mode of binary mixture action and specification of its components activity.

As the results obtained, temporary and qualitative evaluation, which should be used as criteria for determining the mode of such a combinative action, may be significant additional data.

The data presented here permit to specify simultaneously the ac-

tivated mixture component. In the case reported the presence of the chemical in mixture produced activation of  $^{90}\text{Sr}$  effect on some functions of living beings including the hematosis function and enzyme activity.

Additionally, the DDT presence stimulates the  $^{90}\text{Sr}$  blastogenic activity that resulted in the greater frequency of appearance of numerous tumours and combinations with other effects.

## ВЛИЯНИЕ ИНКОРПОРИРОВАННЫХ РАДИОНУКЛИДОВ НА ИММУНИТЕТ

М.А. Невструева, В.А. Колотвин, Р.Е. Лившиц, В.М. Шубик

Институт радиационной гигиены, Ленинград, СССР

**ABSTRACT** - In this report the data of the study of wide immunologic reaction complexes describing a state of nonspecific and specific immunity, allergic and autoallergic reactivity at a single and chronic administration of radioisotopes (Sr-90, I-131, Cs-137, Ce-144, Ra-226, Pu-239) in animals are presented. The differences of some radioisotopes and total irradiation effect on immunity are revealed. Dependence of immunological action upon biophysical properties of radioisotopes is exposed.

Настоящее сообщение обобщает результаты многолетних исследований авторов, сопоставленных с данными литературы (1,2,3) по действию на иммунологическую реактивность некоторых наиболее важных в практическом отношении радиоизотопов (Cs -137, Ce-144, I-131, Sr -90, Pu -239, Ra-226) при их однократном и хроническом введении экспериментальным животным. Целью этих исследований являлось:

1. Характеристика влияния радиоизотопов с различными биофизическими свойствами на комплекс иммунологических реакций, характеризующих состояние неспецифического, специфического иммунитета, аллергической и аутоаллергической реактивности.

2. Выявление возможных различий в изменениях иммунологических показателей при действии внешнего и внутреннего облучения, обусловленного введением различных радиоизотопов.

3. Изучение роли нарушений иммунологической реактивности в патогенезе поражений, вызванных действием внутреннего облучения.

4. Получение информации о "радиочувствительности" различных факторов иммунитета с тем, чтобы рекомендовать наиболее чувствительные иммунологические реакции для выявления ранних признаков лучевых поражений.

Экспериментальные исследования были проведены на белых беспородных крысах и кроликах. Данные о концентрациях введенных изотопов и формирующихся поглощенных дозах представлены на таблице 1.

В качестве факторов неспецифического иммунитета были изучены фагоцитарная реакция нейтрофилов крови, бактерицидность, титры лизоцима и комплемента сыворотки крови. Для характеристики состояния специфического иммунитета проводилась иммунизация животных корпускулярными и вирусными антигенами с последующим определением титров различного рода антител. Была изучена реакция обратной анафилаксии (т.н. реакция на общую иммунологическую реактивность по Иоффе), продукция полных и неполных аутоантител. Более подробно использованные методы исследования описаны в ряде наших публикаций (2,4,5).

Проведенные исследования выявили угнетение всех изученных показателей неспецифического иммунитета. Изменения их имели фазовый характер, зависели от количества введенных радионуклидов. Так, при однократной инкорпорации изотопов в концентрациях 100-400 мккюри/крыса, когда начальная мощность дозы составляла несколько десятков рад/сутки, а суммарная поглощенная доза несколько сот рад, подавление иммунологических реакций было наиболее глубоким, а периоды нормализации редкими. При введении же концентраций на 2 порядка меньше, когда начальная мощность дозы исчислялась десятками рада/сутки, а суммарная поглощенная доза составляла несколько рад, было найдено неглубокое преходящее снижение лишь некоторых иммунологических реакций.

При изучении влияния внутреннего облучения на образование антител были выявлены те же закономерности: изменение их продукции имело фазовый характер, и наблюдалась известная зависимость между количеством введенных радионуклидов и выраженностью колебаний иммуногенеза. Данное положение иллюстрируется на примере изменений антителообразования при однократной инкорпорации  $Sr-90$  (таблица 2).

Как можно видеть, угнетение антителогенеза наступало раньше и отмечалось чаще при введении 100 мккюри радиостронция по сравнению с инкорпорацией 10 мккюри.

Аналогичная зависимость выявлялась при действии и других изученных радионуклидов.

Изучение в реакции Эрне-Нордина образования иммунологически активных клеток селезенки позволило получить некоторые представления о механизме нарушения иммуногенеза при внутреннем облучении. Было выявлено отчетливое изменение образования этих клеток соответственно количеству введенного изотопа. В большинстве опытов найден известный параллелизм между изменениями количества иммунологически активных клеток и титрами антител. Можно полагать, что одной из причин изменений образования антител является нарушение продукции иммунологически активных клеток.

Обратная анафилактическая реакция, т.н. проба на общую иммунологическую реактивность по Иoffee оказалась весьма чувствительной к действию внутреннего облучения. Эта реакция снижалась уже в первые дни после инкорпорации больших концентраций радионуклидов (100-400 мккюри при однократном введении)  $Cs-137$ ,  $Se-144$ ,  $Sr-90$ . Снижение ее регистрировалось при сравнительно небольших поглощенных дозах порядка 30-115 рад.

Что касается исследования аутоантител, то появление их отмечалось в крови не только облученных, но и практически здоровых крыс и кроликов. По-видимому, речь идет о так называемых нормальных аутоантителах, нейтрализующих токсические продукты жизнедеятельности организма. При действии внутреннего облучения число положительных реакций на аутоантитела значительно увеличивалось и было тем больше, чем выше концентрации использованных в опытах радионуклидов. Появление аутоантител имело фазовый характер, что, возможно, было связано с периодической их фиксацией на соответствующих тканях.

Зависимость иммунологических изменений от концентрации введенных радионуклидов и тканевой дозы, фазовый характер этих изменений были выявлены при действии всех изученных радиоизотопов. Однако различие их биофизических свойств находит отражение в особенностях иммунологических сдвигов. Так, образование аутоантител к тем или иным тканям зависело от характера распределения изотопов. Повышенное образование аутоантител регистрировалось преимущественно к тканям, подвергавшимся наиболее значительному

радиационному воздействию. Так, при действии  $\text{Sr-90}$  и  $\text{Ra-226}$  наиболее существенные различия с контролем были получены при определении аутоантител к тканям костного мозга, при внутрибрюшинном введении  $\text{Ce-144}$  и внутривенном введении  $\text{Pc-239}$  - к печени, при поступлении  $\text{I-131}$  к щитовидной железе. При введении же такого равномерно распределяющегося изотопа как  $\text{Cs-137}$  повышенное образование аутоантител обнаруживалось к тканям печени, костного мозга, семенника и даже мышечной ткани, то есть ко всем использованным в наших опытах антигенам.

Различия в характере распределения и ритме выведения радионуклидов находят отражение в особенностях изменения и других иммунологических реакций. Так, при введении  $\text{I-131}$  поглощенная доза в щитовидной железе достигала 35 крад, доза на экстрагиреодные ткани была в 1000 раз меньше. Видимо, вследствие этого и были найдены сравнительно слабые иммунологические изменения на ранних стадиях после инкорпорации этого изотопа.

Снижение иммунологических показателей даже при однократной инкорпорации радионуклидов всегда было длительным, но особенно длительное подавление иммунологической реактивности без тенденции к восстановлению в течение всего периода наблюдения отмечалось при инкорпорации радиостронция, что, вероятно, было связано с весьма длительным периодом полувыведения этого радионуклида, с прочной его фиксацией в скелете.

Снижение иммунологических реакций при внешнем облучении было более глубоким, но менее продолжительным, чем при инкорпорации радионуклидов. Внешнее облучение оказывало более выраженное действие на первичный иммунологический ответ иммунизированных животных, в то время как при поражении продуктами ядерного деления большей чувствительностью обладала вторичная иммунологическая реакция. Выявленные различия, видимо, обусловлены более медленным формированием дозы, непрерывным длительным облучением и замедлением темпа восстановительных процессов даже при однократной инкорпорации радиоизотопов.

Существенный интерес представляет сопоставление динамики иммунологических процессов при хроническом внешнем гамма-облучении и хроническом введении радиоизотопов  $\text{Cs-137}$ ,  $\text{Sr-90}$ ,  $\text{I-131}$ . Изменения иммунологических показателей сравнивались при сопоставимых дозах внешнего и внутреннего облучения. При этом не было выявлено сколько-нибудь выраженных различий при действии адекватных доз внешнего и внутреннего облучения, обусловленного инкорпорацией  $\text{Cs-137}$ . Однако локализация других двух изотопов  $\text{I-131}$  и  $\text{Sr-90}$  в щитовидной железе или скелете обусловило наличие ряда особенностей их действия на иммунологические реакции, характер которых принципиально не отличался от особенностей, выявленных при однократном введении этих радионуклидов.

Переходя к вопросу о роли нарушений иммунитета в патогенезе поражений, вызванных действием внутреннего облучения, следует отметить, что выявленное в наших исследованиях угнетение иммунологической реактивности, по-видимому, могло обусловить снижение устойчивости организма к экзогенной и эндогенной инфекции, что и было обнаружено рядом авторов при действии инкорпорированных радиоизотопов (6, 7). Это угнетение является одной из основных причин гибели облученного организма.

Были проведены специальные опыты по сопоставлению появления аутоантител с показателями, характеризующими функционально-морфологическое состояние органов (активность трансаминаз, С-реактивный белок, гистологические данные). Показано, что изменения функции органов и появление С-реактивного белка обычно предшествует по-

вышению образования аутоантител. Вслед за появлением последних развиваются выраженные функционально-морфологические изменения соответствующих органов. Полученные данные, не свидетельствуя о причинно-следственных отношениях отмеченных явлений, говорят скорее в пользу участия аутоантител в патогенезе лучевых поражений при внутреннем облучении. В пользу последнего говорят и данные о возможном участии аутоантител в формировании постлучевого асперматогенеза. При этом роль аутоантител, определяющихся в разных иммунологических реакциях — преципитинов и комплементсвязывающих аутоантител может быть, по-видимому, различна.

Результаты изучения чувствительности различных факторов иммунитета к действию инкорпорированных радионуклидов отражены на таблице 3.

Приведенные данные показывают наличие известных различий в "радиочувствительности" иммунологических реакций. Однако большинство показателей начинало снижаться (фазовые изменения) при небольших уровнях поглощенных доз, порядка нескольких десятков рад. Мы полагаем, что наиболее "радиочувствительные" и простые в постановке иммунологические реакции, которые дают выраженное снижение при действии различных радиоизотопов, можно использовать для выявления ранних признаков радиационных поражений и для целей гигиенического нормирования радиоактивных поступлений. К таким реакциям по нашим данным можно отнести определение общей иммунологической реактивности, аутоантител, С-реактивного белка, иммунологического ответа на ревакцинацию.

Таким образом, при воздействии инкорпорированных радионуклидов отмечаются выраженные изменения иммунологической реактивности, которые имеют весьма важное значение для течения и исхода лучевых поражений.

#### Литература

1. КЛЕМПАРСКАЯ Н. Н. с сотр.. Радиоактивные изотопы и иммунитет. Атомиздат. М., 1969.
2. НЕВСТРУЕВА М. А. с сотр.. Влияние инкорпорированных изотопов на иммунологические процессы. Атомиздат. М., 1972.
3. ШУБИК В. М., ЖМЭИ, 1970, 5, 52.
4. Он же Гигиена и санитария, 1969, 5, 103.
5. Он же ЖМЭИ, 1972, 9.
6. ЖИГАЛЕВ И. А. Материалы международной конференции по мирному использованию атомной энергии, т. II. Биологическое действие излучений. М., 1958, 102.
7. ТРОИЦКИЙ В. Л., ТУМАНИН М. А.. Влияние ионизирующих излучений на иммунитет. Медгиз, М., 1958.

Таблица 2

Изменения образования антител у крыс, пораженных Sr-90 х)

Количество введенного изотопа	Вид определявшихся антител	Время после введения изотопа, месяцы									
		1		3		6		9		12	
		В	Р	В	Р	В	Р	В	Р	В	Р
100	Агглютинины	У	У	С	С	У	У	У	У	У	С
	Тормозящие гемагглютинацию	У	Н	У	У	С	С	Н	Н	С	Н
10	Агглютинины	Н	У	С	Н	Н	Н	У	У	У	У
	Тормозящие гемагглютинацию	С	С	Н	У	С	С	Н	Н	У	Н
	Поглощенная доза, рады	87	150	206	260	405	450	530	570	642	

х) В-вакцинация; Р-ревакцинация; У-угнетение образования антител в 2-10 раз; С-стимуляция образования антител в 2-10 раз; Н-титры антител на уровне контроля.

Таблица 3

Суммарные поглощенные дозы (в радах), при которых отмечались изменения иммунологических показателей х)

Изотопы Иммунологические реакции	Cs - 137		Ce-144		Sr - 90				I-131	Ra-226
	I	А	Б	I	А	I	А	Б	Б	Б
Фагоцитарная реакция	30	300				45		30		
	30	30				14		30		
Бактерицидность сывротки	45		270	40	430	90	870	100	680	780
	120		270	120	150	30	30	60	210	120
Титр лизоцима сывротки	30	120	1780	10	110	250	2060	1540		
	30	7	390	14	14	120	90	360		36
Титр компонента	45		30	3		45		20		
	120		270	60		120		30		
Образование антител вакцинация	300				110	530				
	30				14	270				
То же ревакцинация						150				
						60				
Обратная анафилаксия	40	60	60	20	110	90	115	30		370
	60	3	30	30	14	30	3	30		60
Сх-реактивный белок						120				
						60				
Образование аутоантител	30		30	30		90				14
	30		300	60		30				60

х) I. А.-однократное введение изотопа; Б-хроническое введение изотопа. I-фазовые изменения реакции; 2-стойкие изменения. П.- в числителе-поглощенная доза, в знаменателе-время ее формирования, дни.

Таблица I

Концентрация радиоизотопов и поглощенные дозы в критических органах  
(Опыты на белых крысах)

Ритм по- ступле- ния	Изотопы Показатель	Cs -137		Se-144		I - 131		Sr -90		Pu-239		Ra-226		
		Критичес- кие органы	Мышцы	Печень	Щитовидная железа		Скелет	Пе- чень	Ки- шеч- ник	Скелет				
Одно- крат- ное вве- де- ние	Способ введения	Через рот		Внутри- брюшинно		Через рот				Внут- ривен- но	Через рот	Через рот		
	Концентра- ция изото- па мккюри/ крыса	400	40 4	200	20 2	480	364	115	18	100	10	1	2	20
Хро- ни- чес- кое вве- дение	Поглощен- ная доза за год (в радах)	46		46		35000	29000	15000	3000	64		180	7	
	Способ введения	Через рот												
Хро- ни- чес- кое вве- дение	Концентра- ция изото- па, мккюри в день на крысу	Cs - 137		I - 131		Sr - 90		Ra-226						
	5	1	0,1	0,01	0,001	0,015	0,0015	0,4	0,1	0,01	0,003	0,0003		
Хро- ни- чес- кое вве- дение	Поглощен- ная доза за год	1650	370	35	0,35	0,035	2350	245	1540	38		26	2,5	
	Поглощен- ная доза за 1,5года	2400	595	51			3520	370	2840	72		50	5	

Примечание: При однократном введении доза для Pu -239 дана за неделю; доза для I-131 - за 14 дней; доза для Se- 144 - за 6 месяцев.

## COMPARISON OF THE BIOLOGICAL EFFECT OF STRONTIUM-90, CESIUM-137, IODINE-131 AND EXTERNAL IRRADIATION

N.A.Zapol'skaya, V.V.Borisova, L.Ya.Zhorno, L.N.Lavrent'ev,  
E.D.Pavlitskaya, A.V.Fedorova and N.G.Yakovleva  
Institute of Radiation Hygiene  
Leningrad, U.S.S.R.

### Abstract

In the paper, a comparative estimate of radiotoxic effect of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{131}\text{I}$  and external irradiation is given. The investigations were carried out on albino rats under the conditions of chronic exposure. In the range of the commensurate doses and exposure times, the relative radiotoxicity of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external irradiation according to haematological and biochemical indices and lifespan was found to be close to unity. The effectiveness of  $^{131}\text{I}$  according to the same indices was considerably lower. As to the incidences of malignant tumours - the efficiency of  $^{90}\text{Sr}$  proved to be in excess of those for  $^{137}\text{Cs}$ ,  $^{131}\text{I}$  and external irradiation.

### Introduction

In the recent years, the attention of researchers has been attracted to the problems of comparative assessing a toxic action of nuclides and external irradiation. However, most of these works concerns blastomogenic efficiency of bone-seeking nuclides<sup>1-4</sup>.

This report treats the problem of chronic exposure to  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ , and external irradiation in its comparative aspect.

### Materials and Methods

The experiments were done on albino rats, males and females. A total of 864 test rats was divided into 11 treated groups and a control one. The animals were fed a daily diet containing the nuclides throughout lifespan in the following doses: 2.0, 0.5, 0.05  $\mu\text{Ci}$   $^{90}\text{Sr}$ , 25.0, 5.0, 0.5  $\mu\text{Ci}$   $^{137}\text{Cs}$ , 0.075 and 0.0075  $\mu\text{Ci}$   $^{131}\text{I}$  per kg of body weight per day. The external exposure was delivered at levels of 5.0, 1.0 and 0.2 R/d. These dosages, for all the above types of exposure, were conditionally denoted as "large", "medium" and "small" doses, the absorbed doses at the end of the year being 1500-2300, 240-360 and 40-70 rads, respectively.

The haematological, biochemical, morphological indices, the animal lifespans and the tumour incidences were studied.



## Results and Discussion

In the case of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  or external irradiation at "large" doses, the peripheral blood studies have evidenced the short-period initial leukocytosis and the prolonged leukopenia advanced gradually. When exposed externally, the leukopenia occurred somewhat earlier than in administering  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . No essential differences in the extent of depression in leukopoiesis induced by  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  or external radiation have been displayed at the late periods of time (540-660 days). The leukocyte number in these groups was 35-45% lower than in the control one. During the first months, the leukopenia is due to lymphopenia mostly. To the end of exposure, however, the neutrophil number decreased, too.

For the  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external exposure at "large" doses, the thrombocyte response was generally similar to the leukocyte one, but the reduction in the thrombocyte number took place later and was shown to a lesser degree. For a long time, no quantitative changes in red cells were found. The moderate anaemia has developed in the animals only to the end of the experiment. The reticulocyte number increased markedly at this period.

In the range of "medium" doses, the persistent leukopenia at 7 months postintake occurred only in administering  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . To the end of the second year, the leukocyte number in these groups was 20-23% lower than in the control one. When exposed chronically to the same doses of external radiation, the animal response of leukopoiesis depression was unstable. The administration of "large" and "medium"  $^{131}\text{I}$  dosages didn't lead to the marked reduction in the formed elements of peripheral blood.

The cytological investigations have given evidence that for all exposures the number of leukocytes with structural disturbances of nucleus and cytoplasm (binucleated lymphocytes, hypersegmented neutrophils etc.) increased in the peripheral blood. When exposed chronically to the "large" and "medium" doses of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $\gamma$ -irradiation, the number of degenerative leukocytes exceeded that in controls to the end of the second year by a factor of 3 and 1.5, respectively. In case of administering similar doses of  $^{131}\text{I}$ , the cytological changes in the leukocytes were less intensive.

The presented data give evidence that the modes of changes in peripheral blood at an early and late stages are identical, when exposed to  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external radiation. To determine the dose-effect relationship, the response of leukopoiesis depression was used as the most specific one for the effect of radiation exposure (Fig. I).

As one can see from Fig. I, the dose-effect relationship can be represented by an exponential function. The course of the curve is apparently influenced by the process of repair.

The analysis of findings shows that the relative toxicity of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external irradiation is close to unity. The minimal dose that induces leukopenia is equal to 150-200 rads.

The action of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external irradiation of "large" do-

ses has led at early stages to increasing activity of some blood ferments. However, the stimulation phenomena were rapidly replaced by prolonged inhibition of ferment activity. From the 60th day to the end of the observations, the activity of cholinesterase in erythrocytes and serum was 25-35% lower than in control data. In administering  $^{131}\text{I}$ , no marked inhibition of ferment activity was observed.

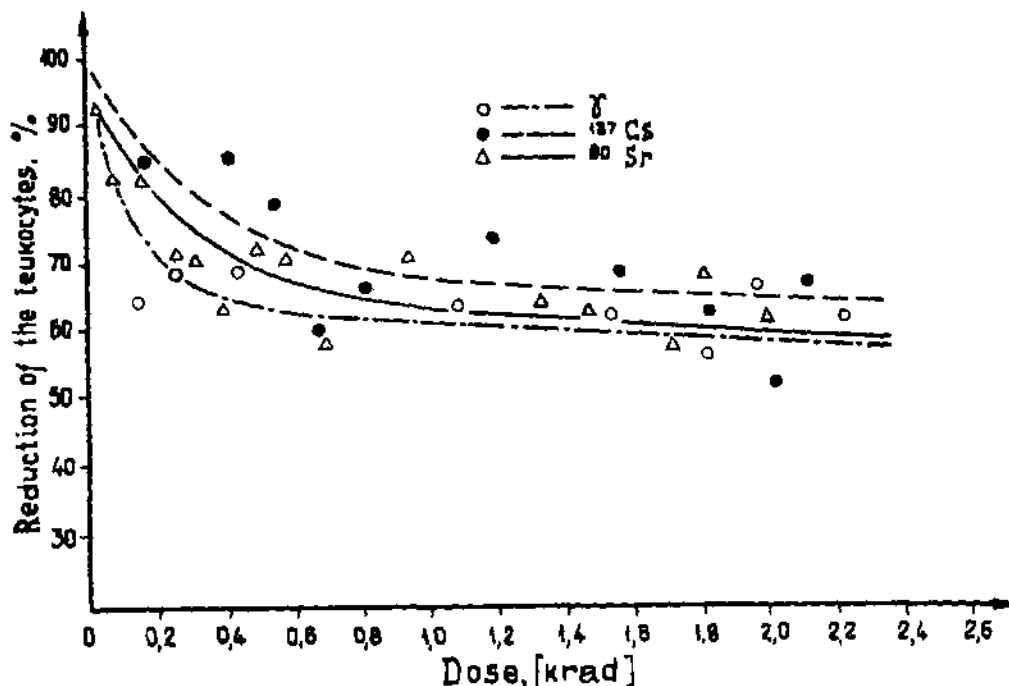


Fig. 1. The response of leukopoiesis as a function of the absorbed dose.

When exposed to  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external irradiation with the same doses, the concentration of residual nitrogen in blood on day 90 exceeded that of control by 45-60% and was kept at this level to the end of the observation. No significant differences in the intensity of reaction, when exposed to  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external  $\gamma$ -irradiation, have been revealed.

To compare the action of nuclides and external irradiation, times of animal deaths and tumour incidences were studied. The median survival times and its confidence intervals of exposed animals have been determined. The results of calculation are given in Table I.

The significant lifespan shortening was observed among the animals exposed to "large" doses of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external irradiation. A slight lifespan shortening has been revealed also for the animals who received  $0.5 \mu\text{Ci } ^{90}\text{Sr}/\text{kg}$  per day. Lifespan shortening was on the average 0.09 day per rad. The mortality curve plotted against absorbed dose was S-shaped (Fig. 2).

In exposing to  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external irradiation, the maximum mortality of animals, when plateaued, was 40, 30 and 34%, respectively, the absorbed dose being 4000 rads for bone tissue and 3000-

Table 1.  
The average lifespan of animals for various exposures

Exposure	The number of animals	Median (life) /days/	95% confidence limits of Me
$^{90}\text{Sr}$	0.40 $\mu\text{Ci/d}$	75	420
	0.10 $\mu\text{Ci/d}$	75	497
	0.01 $\mu\text{Ci/d}$	75	570
$^{137}\text{Cs}$	5.0 $\mu\text{Ci/d}$	74	452
	1.0 $\mu\text{Ci/d}$	74	567
	0.1 $\mu\text{Ci/d}$	74	603
$^{131}\text{I}$	0.0150 Ci/d	75	567
	0.0015 Ci/d	75	587
$\gamma$ -irradiation	5.0 R/d	42	446
	1.0 R/d	75	543
	0.2 R/d	75	620
control	75	608	540 - more than 660

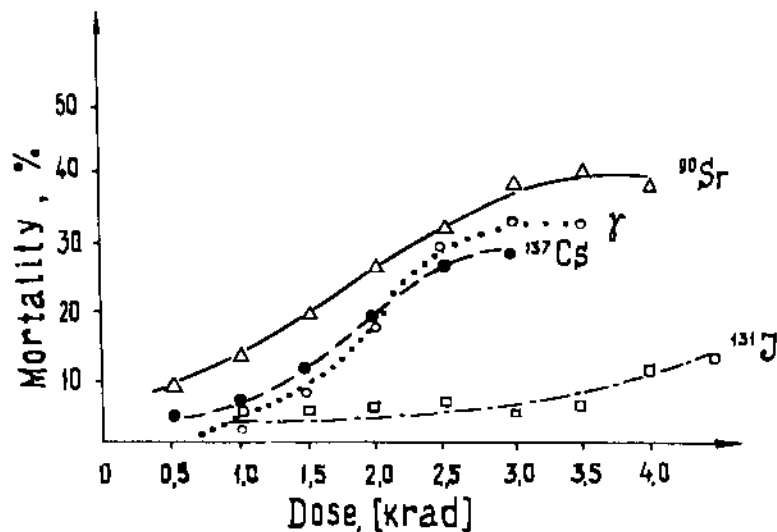


Fig.2. Mortality of animals versus absorbed dose with taking into account the death of control animals.

-3500 rads for whole body. The maximum mortality of animals per 1 rad, when exposed to  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external irradiation, was the same, being equal to 0.01%. Hence, it follows that the toxicity of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and external irradiation, when estimated on such criterion as the mortality of animals, is close to unity. Administering  $^{131}\text{I}$  has not led to the significant mortality of animals as compared with controls.

The pattern of diseases and the causes of death of the exposed and control animals were identical on the whole. The death of rats was caused mostly by the chronic inflammatory processes in lungs, intestinal and peritoneum. On the background of the above phenomena the individual animals have shown the malignant and benign tumours. The location and incidence of tumours depended upon the types of exposure and the dose (Table 2).

In administering 2  $\mu\text{Ci}$   $^{90}\text{Sr}$ /kg per day, the incidence of malig-

Table 2.

The tumour incidence following exposure to "large" doses of radionuclides and external irradiation

Tumours	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{131}\text{I}$	$\gamma$ -irradiation	Control
osteosarcomas	5.0 (6.7%)				
leukosis	3.0 (4.0%)	4.0 (5.4%)	-	2.0 (4.7%)	-
lymphosarcomas	6.0 (8.0%)	-	-	1.0 (2.4%)	-
thyroid adenomas	3.0 (4.0%)	2.0 (2.7%)	5.0 (6.7%)	1.0 (2.4%)	-
thyroid cancer	-	-	2.0 (2.7%)	-	-
parathyroid adenomas	1.0 (1.3%)	1.0 (1.4%)	-	-	-
mammary fibroadenomas	-	4.0 (5.4%)	1.0 (1.3%)	2.0 (4.7%)	1.0 (1.3%)
soft tissue fibromas	-	3.0 (4.0%)	-	1.0 (2.4%)	-
tumours in adrenal glands		-	-	1.0 (1.3%)	1.0 (2.4%)
total of rats with tumours	18.0 (24%)	14.0 (18.9%)	9.0 (12%)	8.0 (19%)	1.0 (1.3%)
total of rats in the group	75 (100%)	74 (100%)	75 (100%)	42 (100%)	75 (100%)

nant tumours was 18.7%. The latent period (the time from onset of exposure to death of animal) was 300-540 days for lymphosarcomas and 450-660 days for leukosis and osteosarcomas. The absorbed doses, just before lymphosarcoma appearance, reached 1350 rads, those for leukosis and osteosarcomas did 2200 and 2400 rads, respectively. Lymphosarcomas developed from lymph nodes of mesentery or lymphatic tissue of blind gut. Osteosarcomas localized in the femur, usually in its lower third. In addition to the malignant neoplasms, the benign tumours were found in the form of adenomas in thyroid and parathyroid glands.

In administering 25  $\mu\text{Ci}$   $^{137}\text{Cs}/\text{kg}$ , the incidence of malignant neoplasms was considerably lower and was equal to 5.4%. All the tumours were attributed to the haemopoietic tissue and were developed during the period from 410 to 600 days with the absorbed doses of 1840-2900 rads. At the same time,  $^{137}\text{Cs}$  has induced the relatively high percentage of benign tumours.

Administering  $^{131}\text{I}$  resulted in the thyroid tumour incidence of 9.4%. The malignant tumours arose with the absorbed doses of 4000-4500 rads.

In the continuous  $\gamma$ -irradiation of animals to the dose of 5 R/d, the incidence of malignant tumours was equal to 7.1% (leukosis, lymphosarcoma). The tumours have appeared for the period of time ranged from 320 to 600 days, when the absorbed doses reached 1500-3000 rads.

In administering  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{131}\text{I}$  at concentration of 0.5, 5.0 and 0.0075  $\mu\text{Ci}/\text{kg}$  respectively and exposing externally to the dose of 1 R/d, the total incidence of tumours was 3-6 times lower than in case of the exposure to "large" doses. However, the differences in location and in mode of tumour growth among the individual test groups of animals were kept the same.

The analysis of the total findings gives evidence that according to the haematological and biochemical values and lifespans of animals, the relative toxicity of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $\gamma$ -irradiation is close to unity under the conditions of chronic exposure. The general toxic effect of  $^{131}\text{I}$  for the same doses is less pronounced.

Close values of effectiveness of  $^{137}\text{Cs}$  and  $\gamma$ -irradiation seem to be determined by the similar spatial and temporal distribution of absorbed doses. In concerning with the  $^{90}\text{Sr}$  effect, it should be borne in mind that the latter is not restricted to the critical organ alone. When  $^{90}\text{Sr}$  ingested daily, the gastrointestinal tract is exposed considerably together with bone tissue and marrow. In addition, the high energy  $\beta$ -particles escape beyond the bone and expose the hypophysis, the thyroid, as well as the liver, kidney and other organs.

Blastomogenic effectiveness of  $^{90}\text{Sr}$  with regard to the induction of malignant neoplasms was found to be higher than those of  $^{137}\text{Cs}$ ,  $^{131}\text{I}$  and external irradiation. Perhaps, it is due to the effect which was compared with the average dose to skeleton without taking the nonuniformity of radionuclide distribution into account. However, this point requires a further experimental investigation.

#### References

1. Howard E.B., Clarke W.J., Karagianes M.T., and Palmer R.F., "Sr-90 induced bone tumours in miniature swine", Rad.Res., Sept.1969, 39, No.3, 594.
2. Mays C.W., Taylor G.N., Jee W.S.S., and Dougherty T.F., "Speculations on the risk to adult man from a "permissible" body burden of  $^{239}\text{Pu}$  (0.04  $\mu\text{Ci}$ ) equally divided between bone and liver". Rad.Res., July 1970, 43, No.1, 264.
3. Nilsson A., "Pathologic effects of different doses of radiostrontium in mice - Dose effect relationship in  $^{90}\text{Sr}$  - induced bone tumours". Acta Radiologica, April 1970, 9, Fasc. 2, 155.
4. Owen M., Sissons H.A., and Vaughan J., "The effect of a single injection of high dose of Sr-90 in rabbits". Brit.J.Cancer, 1957, 11, 229.

ОТДАЛЕННЫЕ ПОСЛЕДСТВИЯ ПРИ ВНЕШНЕМ БЕТА ОБЛУЧЕНИИ  
Ю.И. Москалев, В.И. Шелеснова, И.А. Миловидова  
Институт биофизики Министерства здравоохранения  
Москва Союз Советских Социалистических Республик

Abstract

The effect of various doses (from 600 to 7200 rad) from  $^{90}\text{Sr}$ - $^{90}\text{Y}$  source of external beta irradiation on life duration, frequency and rate of skin and mammary tumour development and cataracts (whole-body irradiation and irradiation of the front and back parts of the body) has been studied in the experiment on white strainless rats. The effective dose values of acute ( $\text{LD}_{50/30}$ ), subacute ( $\text{LD}_{50/120}$ ) and chronic ( $\text{LD}_{50/360}$ ) radiation exposure were lower after whole-body irradiation (4750, 4680 and 3240 rad respectively) than after irradiation of the front (5960, 5120 and 4310 rad) or back (6910, 5830 and 5380 rad) parts of the body.

Benign (adenoma of sebaceous glands, fibroma) and malignant (basalioma, flat-cell cancer, sebaceous gland cancer, sarcoma) skin tumours, benign (adenoma, adenofibroma, fibroadenoma, fibroma) and malignant (cancer, sarcoma) mammary tumours and cataracts developed at late periods in the irradiated parts of the body after the use of all above doses. Frequency and rate of skin and mammary tumour development and cataracts were proportional to the irradiation dose.

Введение

Особенности клинических проявлений и количественные закономерности поражающего действия внешнего бета-излучения в зависимости от дозы и условий лучевого воздействия были предметом многих исследований<sup>1-3</sup>.

В этих работах достаточно подробно описана клиника поражающего действия бета-излучения, установлены уровни доз вызывающих ранние, а в ряде случаев и поздние реакции кожи, описана зависимость этих реакций от глубины проникновения излучения в кожу и объема облучаемой поверхности. Меньше внимания было уделено изучению развития отдаленных последствий в зависимости от дозы и локализации лучевого воздействия. Данные об уровнях оптимальных и минимальных blastogenic доз для кожи противоречивы, отсутствуют сведения о возможности развития опухолей молочных желез под влиянием внешнего бета излучения, мало данных о катарактогенном действии бета-лучей.

Настоящая работа посвящена анализу влияния общего внешнего бета-облучения, облучения передней или задней половины тела

крысы на продолжительность жизни, частоту и скорость развития опухолей кожи, молочных желез и катаракт.

#### Материалы и методы исследования

Для тотальных и парциальных облучений крыс использовали специально сконструированный для облучения мелких лабораторных животных бета-облучатель, детально описанный в работе<sup>4</sup>. Всего в опытах использовано 760 самок белых беспородных крыс в возрасте 3-4 месяцев, весом 180-220 г. Животных облучали в широком диапазоне доз от 600 до 7200 рад. В опытах применяли общее облучение, облучение передней или задней половины тела. При облучении половины тела необлученную часть тела защищали экраном из свинца толщиной 5 мм. При облучении в дозах 600, 1200 и 2400 рад в каждом варианте опыта было по 30, при облучении в дозах 3600, 4800, 6000 и 7200 рад по 15 крыс, а в контроле 25 крыс. Кроме того в офтальмологических исследованиях имелась дополнительная группа животных, состоявшая из 285 крыс облучавшихся тотально в дозах 800, 1600, 3200 и 4500 рад. На каждую дозу было взято по 60 крыс а в качестве контроля 45 животных.

При оценке биологического действия бета излучения применяли клинические, офтальмологические и морфологические методы исследования. У животных выявляли сроки развития эпиляции, эпидермита и изъязвлений. Локализацию, частоту и скорость развития опухолей молочных желез и кожи учитывали ежемесячно путем визуального осмотра и пальпации. Частоту опухолей молочных желез выражали в процентах от числа живых крыс в момент обследования. По данным динамики вымирания крыс при помощи метода пробит-анализа определяли среднюю продолжительность жизни животного и величины LD50% для различных сроков. Секционный материал фиксировали в 10% формалине, парафиновые срезы толщиной 5 $\mu$  окрашивали гематоксилин-эозином. Глаза животных обследовали клинически с помощью электрического офтальмоскопа и лупы +15Д. Степень помутнения хрусталика определяли по классификации американских авторов<sup>5</sup>.

#### Результаты исследований

Продолжительность жизни. Динамика вымирания облученных крыс зависит от дозы и условий лучевого воздействия. При облучении животных в дозах равных и меньше 3600 рад изменения в средней продолжительности жизни крыс отсутствуют. При общем облучении в дозе 4800 рад крысы начинают погибать через 19 дней, при облучении передней половины - через 23, а при облучении задней половины тела - через 98 дней. При общем облучении все крысы пали в промежутки времени между 16-30 днем, при облучении передней половины тела в течение месяца погибло 60% (9 из 15 животных), а при облучении задней половины тела - 33% крыс (5 из 15 животных).

При дозе 7200 рад гибель животных при общем бета-облучении, облучении передней или задней половины тела начинается практически в одинаковые сроки на 15, 17 и 14 день, соответственно. Однако, при общем облучении все крысы пали в течение 15-22 дней, при облучении передней и задней половины тела в течение месяца пало соответственно 73% (11 из 15 животных) и 60% (9 из 15 животных) крыс.

LD50/30 при общем бета-облучении оказалась равной 4750 рад, при облучении передней и задней половины тела, соответственно, 5960 и 6910 рад. Большая биологическая эффективность общего внешнего облучения выявляется при сравнении не только острых, но также подостро и хронически эффективных доз. Так, величины LD50/360 (3240 рад) и LD50/480 (2660 рад) при общем бета облучении оказа-

лись в 1,3-1,2 и 1,5-1,7 раза ниже, чем при облучении, соответственно, передней (4310 и 3200 рад) и задней (5380 и 3940 рад) половины тела. Облучение задней половины тела менее эффективно не только по сравнению с облучением всего тела, но и с облучением передней половины его. Табл. 1.

Клиническая картина поражения. Одним из наиболее тяжелых последствий воздействия  $\beta$ -лучей являются поражения кожи, степень выраженности которых зависит от дозы. При дозах 600-1200 рад, как при общем, так и при местном воздействии изменения кожи выражены слабо. На 21-25 сутки после облучения наблюдается незначительная эпилляция, которая полностью восстанавливается через 1-2 месяца после облучения. При общем облучении в дозе 2400 рад на 14-20 день развивается эпилляция, за которой следует дерматит и изъязвления кожи. Через 1,5 месяца у 2-х крыс развилась лучевая язва. Изменения кожи при тотальном облучении животных в дозе 3600 рад характеризовались развитием эпилляции (на 10-12 день), эпидермита (на 15-20 день) и лучевых язв (на 30-35 день). Наиболее выраженная картина поражений кожи развивается при дозе 4800 рад.

Опухоли кожи. У крыс, павших в отдаленные сроки (через 8-10 месяцев) после облучения развиваются опухоли кожи как эпителиального, так и соединительно-тканного происхождения. У контрольных крыс опухолей кожи не наблюдали. При общем бета-облучении опухоли кожи развиваются при дозах равных и выше 600 рад, при облучении передней половины тела - при дозах 1200 рад, а при облучении задней половины тела - при дозах 2400 рад. При общем облучении опухоли кожи возникают примерно с одинаковой частотой в передней и задней половинах тела, при облучении половины тела только в облученных частях, что свидетельствует о ведущей роли прямого действия радиации в генезе этого типа новообразований. Максимальную частоту и мультицентрическое развитие опухолей независимо от условий лучевого воздействия наблюдали при облучении животных в дозе 3600 рад и выше. Гистологические данные представлены в табл. 2. Плоскоклеточный рак, как правило, развивался на фоне выраженных изменений кожи, таких как дерматит и лучевая язва, базалиомы - напротив, формировались при отсутствии изменений со стороны кожного покрова. У одного и того же животного нередко возникали опухоли различного гистологического строения. Плоскоклеточный рак может сочетаться с формирующейся базалиомой, фибромой с саркомой и т.д.

Опухоли молочных желез. Частота, скорость возникновения и локализация опухолей молочных желез зависят от дозы и условий облучения. При общем бета-облучении опухоли молочных желез возникают примерно с одинаковой частотой в передней и задней половинах тела, в то время как при облучении половины тела почти исключительно в облученных частях. Первые опухоли молочных желез у контрольных животных обнаружили в среднем через  $13,8 \pm 2,4$  месяца после начала эксперимента. При общем облучении в дозах 600-2400 рад они появлялись значительно раньше (через 6,0-9,6 месяцев), чем у контрольных животных. При облучении в дозах 1200 и 2400 рад опухоли молочных желез были обнаружены раньше в задней половине тела (через 6,0-7,6 месяцев); в передней половине тела они были выявлены, соответственно, через 9,4 и 11,2 месяцев. При дозе 3600 рад опухоли молочных желез в передней и задней половинах тела развивались примерно в те же сроки (через 13,3 и 12,2 месяца), что и у контрольных крыс. Следовательно при больших дозах общего бета-облучения опухолей молочных желез появляются позже, чем при меньших уровнях лучевого воздействия.



При общем бета-облучении максимальную частоту опухолей молочных желез в передней (40%) и задней (43%) половинах тела обнаружили через 16-18 месяцев при дозе 1200 рад. У контрольных животных в это время частота опухолей составляет 12,6%. В опыте с облучением половины тела существенно различаются сроки появления первых опухолей молочных желез в облученных и необлученных частях тела. Так, при облучении передней половины тела в дозах 600, 1200, 2400 и 3600 рад первые опухоли молочных желез в облученной части тела были обнаружены, соответственно, через 7,9; 5,7; 9,4 и 9,2 месяца, а в необлученной задней половине - через 18,9; 12,7; 17,9 и 20 месяцев, т.е. в те же сроки или даже несколько позже, чем у контрольных крыс. При облучении задней половины тела в указанных выше дозах первые опухоли в облученных железах были обнаружены, соответственно, через 7,9; 6,8; 7,6 и 7,4 месяца, а в передней необлученной половине - через 12,7; 11,2; 11,2 и 9,2 месяца. Морфологическому обследованию было подвергнуто 92 опухоли молочных желез. Табл. 3. Доброкачественные опухоли были представлены фиброаденомами, аденомами, фибромами, аденофибромами. Злокачественные - раками (криброзный, аденокарцинома) и саркомами. Доброкачественные опухоли молочных желез часто формировались на фоне кистозно-фиброзной мастопатии. В нескольких случаях они находились в стадии малигнизации. Опухолям молочных желез часто сопутствуют кистозные изменения яичников и увеличение гипофиза. Указанные изменения наряду с прямым действием радиации, вероятно, также играет определенную роль в генезе опухолей молочных желез.

**Катаракты.** Изменения в хрусталике крыс, характерные для так называемой лучевой катаракты, возникают и развиваются в прямой зависимости от дозы как при облучении всего тела, так и передней половины его. При облучении задней половины тела типичных для лучевого воздействия изменений в хрусталике обнаружено не было. Начальные изменения (I-я степень помутнения) в хрусталике в диапазоне от 800 до 4500 рад были выявлены в пределах от 11,5 до 41 недели. Скрытый период появления начальных катарактальных изменений зависит от дозы: при дозе 800 рад I-я степень была зафиксирована через 41 неделю от момента облучения, а при дозе 4500 рад - через 11,5 недель. Через 46 недель после начала облучения в дозах 1600, 3200 и 4500 рад катаракты обнаружили в 100% случаев, в то время как при дозе 800 рад только в 27%. Даже через 63 недели при этой дозе катаракты выявили только в 44% случаев. Динамика развития помутнения в хрусталике при воздействии в дозах от 800 до 4500 рад представлена графически (рис. 1). Материалы настоящего исследования находятся в хорошем соответствии с данными ряда авторов.

### Заключение

Отчетливое увеличение новообразований кожи и молочных желез обнаружено при облучении бета-лучами в дозах, равных и выше 600 рад, а катаракт при дозах равных и выше 800 рад. Катаракты, опухоли кожи и молочных желез возникают в результате прямого действия радиации на радиочувствительные структуры ткани, о чем свидетельствует возникновение указанной патологии только в облученных частях тела (опухоли кожи, катаракты) или более раннее развитие и появление в большем проценте случаев (опухоли молочных желез) по сравнению с адекватным контролем.

Таблица №1

Величина LD<sub>50</sub>% для различных сроков в зависимости от условий облучения крысы бета-лучами Sr<sup>90</sup> - Y<sup>90</sup> (с доверительными интервалами для p=0,05)

LD 50%	Общее облучение	Облучение половины тела	
		передняя	задняя
30	4750 (4350 - 5155)	5950 (5455 - 6465)	6910 (6297 - 7523)
60	4680 (4257 - 5103)	5340 (4969 - 5711)	6250 (5713 - 6787)
120	4680 (4228 - 5132)	5125 (4741 - 5509)	5830 (5326 - 6334)
240	3310 (3110 - 3510)	3800 (3416 - 4184)	5500 (4933 - 6067)
360	3240 (2800 - 3510)	4310 (3645 - 4975)	5380 (5023 - 5767)
480	2660 (2272 - 3048)	3270 (2837 - 3563)	3940 (3405 - 4475)

Таблица №2

Микроскопическая характеристика опухолей кожи

Доза облучения (рад)	кол-во вскрытых животных	кол-во животных с опухолями	Общее число опухолей	Доброкачественные опухоли		Злокачественные опухоли			
				аденома салivных желез	фиброма	базалиома	плоско-клеточный рак	рак салivных желез	саркома
600	34	1	2	—	—	2	—	—	—
1200	33	2	3	—	1	2	—	—	—
2400	43	6	10	—	—	2	5	—	3
3600	33	18	24	1	1	3	13	1	5
4800	8	5	10	—	—	—	6	2	2
6000	3	2	4	—	—	—	1	—	3
7200	3	2	3	—	—	—	—	—	3
Контроль	9	—	—	—	—	—	—	—	—
Всего	186	36	56	1(1,8%)	2(3,6%)	9(16,1%)	25(44,6%)	3(5,3%)	16(28,6%)

Таблица №3

Микроскопическая характеристика опухолей молочных желез

Доза облучения (рад)	кол-во вскрытых животных	кол-во животных с опухолями	Общее кол-во опухолей	Доброкачественные опухоли				Злокачественные опухоли	
				аденома	адено-фиброма	фибро-аденома	фиброма	рак	саркома
600	34	7	10	1	2	7	—	—	—
1200	33	22	30	2	6	15	2	4	1
2400	43	22	28	1	5	17	—	5	1
3600	33	14	17	—	2	7	1	5	2
4800	8	2	2	—	—	1	—	1	—
7200	3	1	1	—	—	—	—	—	1
600-7200	154	68	89	4(4,5%)	15(16,9%)	47(52,7%)	3(3,4%)	15(16,9%)	5(5,6%)
Контроль	9	3	3	—	1	2	—	—	—
Всего	183	71	92						

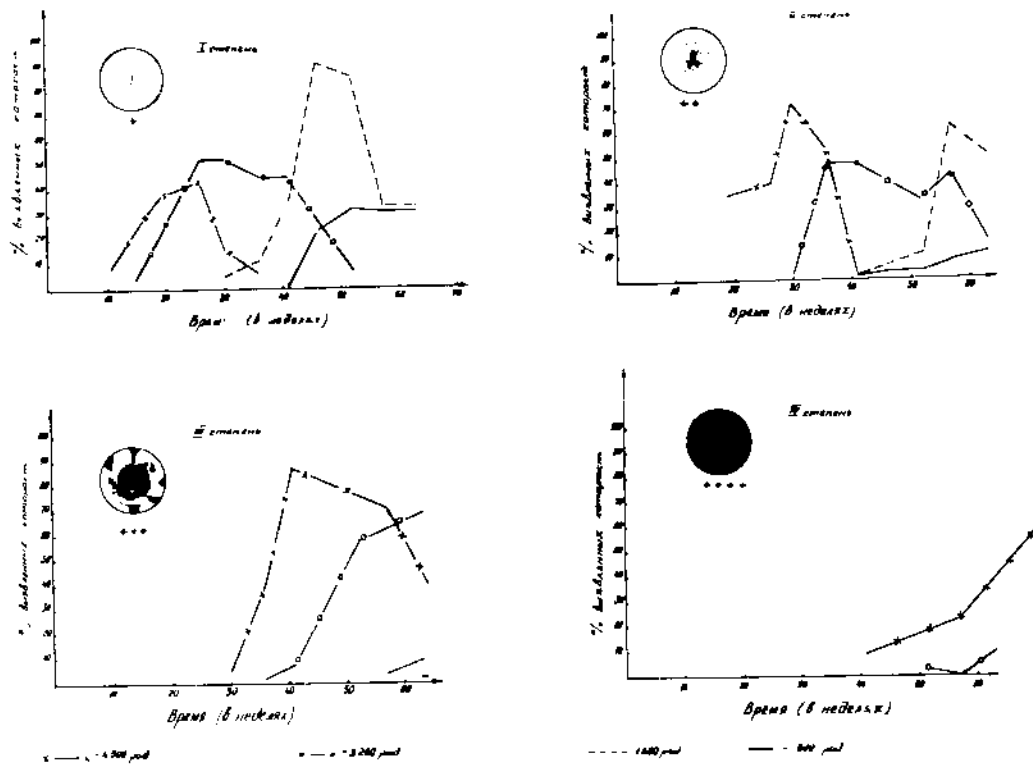


Рис. 1. Динамика развития помутнения в хрусталике при облучении в дозах 800-4500 рад.

#### Библиография

1. J. Raper, P. Henshaw, R. Smider (1951). In Effects of external beta radiation (by ed R. Zirkle New York Chap.3, p.62).
2. Ю.В. Себрант (1970). Биологическое действие внешнего бета-облучения. Москва, Атомиздат.
3. Н.Г. Даренская, Т.М. Правда, И.К. Рейтаровский (1971). Зависимость клинических проявлений поражения крыс от величины дозы тотального внешнего  $\beta$ -облучения. В сб. "Вопросы общей радиобиологии". Москва, Атомиздат, стр. 79-89.
4. В.С. Грамматикати, В.К. Мостинская, Е.С. Страшненко (1971). Бета-установка для тотального облучения мелких лабораторных животных. В сб. "Вопросы общей радиобиологии". Москва, Атомиздат, стр. 14-18.
5. K.W. Christenberry, J. Furth (1951). Induction of Cataracts in Mice by slow Neutrons and x-rays "Proc. Soc. Exp. Biol. Med." 77, 3, 559-560.
6. A.C. Upton, K.W. Christenberry, J. Furth (1953). Comparison of Local and Systemic Exposures in Production of Radiation Cataract "A.M.A. Arch. Opth." 49, 2, 164-167.
7. D. Cogan, J. Goff, E. Graves (1952). Experimental Radiation Cataract "A.M.A. Arch. Opth." 47, 5, 584-592.
8. G. Merriam (1956). The Effects of Beta Radiation on the Eye "Radiology" 66, 2, 240-245.
9. И.А. Миловидова, Ю.И. Москалев (1972). Изменения в хрусталике крыс при действии общего внешнего  $\beta$ -облучения. Р радиобиология, т. 12, 2, стр. 301-303.

ИЗМЕНЕНИЕ СРЕДНЕЙ ПРОДОЛЖИТЕЛЬНОСТИ ЖИЗНИ  
КРЫС В ЗАВИСИМОСТИ ОТ ЧАСТОТЫ РАЗВИТИЯ ОСТЕО-  
САРКОМ, ИНДУЦИРОВАННЫХ СТРОНЦИЕМ-90

В. Л. Шведов

Министерство здравоохранения СССР  
Институт биофизики  
Москва  
СССР

The experiments were carried out on white rats at the age 3.5-4 months at the beginning of the experiment. The animals were divided into ten groups of 100 rats each. The animals of nine experimental groups received Sr-90 orally every day within the entire duration of their life, the dose being 0.00005; 0.0005; 0.005; 0.05; 0.5; 1.0; 2.0; 4.0 and 5.0 daily per animal.

Consequently the minimal Sr-90 daily intake which evoked the development of osteosarcoma was 0.5 daily. The dose rate to the critical organ was not more than 25 rads daily, the absorbed dose being 5,000 rads.

Thus the performed investigations show that osteosarcomas are not the main cause of the average lifespan decrease of the population.

В настоящее время радиобиологическая литература располагает большим фактическим материалом <sup>1,2,3,4</sup> относительно развития остеосарком у лабораторных животных в зависимости от дозы инкорпорированных остеотропных изотопов, в частности стронция-90.

Большинство авторов считает, что закономерность частоты развития костных опухолей имеет линейный вид, т.е. чем выше тканевая доза в критическом органе, тем чаще появляются остеосаркомы и тем короче время, необходимое для их развития. Однако, в последнее время <sup>5,6,7</sup> появляются данные, которые свидетельствуют о том, что линейный характер частоты остеосарком проявляется лишь в определенном интервале доз.

Учитывая важность этого показателя при оценке риска поражения стронцием-90 человека представляется необходимым изучить закономерность развития остеосарком в зависимости от поглощенных доз в критическом органе и влияния их частоты на величину средней продолжительности жизни в условиях хронического поступления в организм этого изотопа.

#### МАТЕРИАЛ И МЕТОДИКА

Эксперименты проведены на белых крысах-самцах в возрасте 3,5-4 месяцев к началу опыта. Все животные были разделены на 10 групп, девять из которых ежедневно, через рот, в течение всей жизни получали стронций-90 в широком интервале концентраций (табл. I).

Животные всех подопытных и контрольных групп находились в совершенно равных условиях вивария на обычном рационе.

Дозиметрические исследования проводили в динамике через 30, 90, 180, 270, 360 и 540 дней от начала опыта. На каждую точку забивали по 3-5 крыс из каждой группы. Расчет тканевых доз производили на основании фактического определения концентрации стронция-90 в бедренной кости с учетом 50% поглощения энергии.

Таблица I

Суточные концентрации стронция-90 и число животных в подопытных и контрольной группах

Стронций-90 (мккюри/сутки)	Число животных	Стронций-90 (мккюри/сутки)	Число животных
0,00005 *	100	1,0	100
0,0005	100	2,0	100
0,005	100	4,0	100
0,05	100	5,0	100
0,5	100	Контроль	100

\* — активность стронция-90.

Для изучения продолжительности жизни крыс и частоты развития у них остеосарком в каждой подопытной и контрольной группах находилось по 50 животных. Наличие остеосарком определяли при помощи пальпации трубчатых костей. Всех погибших животных вскрывали для определения у них причин смерти, используя для этой цели гистологические методы исследования.

### РЕЗУЛЬТАТЫ

Общее состояние подопытных животных, получавших ежедневно стронций-90 в количествах от 0,00005 мккюри до 0,05 мккюри в сутки, ничем не отличалось от контрольных крыс. Начальное снижение веса тела зарегистрировано к концу первого года у животных, которые подвергались воздействию радиостронция в концентрации 0,5 мккюри в сутки.

Ежедневное поступление в организм более высоких концентраций стронция-90 (1,0; 2,0; 4,0 и 5,0 мккюри) приводило к более выраженному изменению этого показателя. Так, например, у крыс, получавших по 5,0 мккюри изотопа в сутки, уже через 3 месяца от начала опыта вес тела был достоверно ниже веса контрольных животных и в последующем до конца их жизни не восстанавливался.

На рис. 1 приведены результаты формирования в бедренной кости крыс поглощенных доз в зависимости от концентрации стронция-90 и времени поступления его в организм животных.

Как видно из графика, эксперимент охватывал широкий диапазон доз. Минимальные величины лишь в несколько раз превышали фоновые значения, тогда как максимальные — приводили к развитию острых и подострых форм лучевых поражений, которые резко сокращали продолжительность жизни подопытных животных.

На рис. 2 представлены данные о смертности крыс относительно времени поступления в организм радиостронция.

Как видно из графика, кривые смертности подопытных и контрольной группы животных имеют S-образный вид. По времени начала гибели крыс все кривые можно разделить на 2 группы. Через 50-100 дней от начала опыта начинали погибать крысы, которые ежедневно получали стронций-90 от 1,0 до 5,0 мккюри в сутки. Поступление изотопа в организм от 0,5 мккюри и ниже приводило к начальной гибели животных не ранее чем через 200 дней. В этом же интервале времени обнаружено начало гибели контрольных крыс.

Интересно отметить, что с увеличением времени поступления в организм стронция-90 кривые смертности явно разделяются на 3 группы. Резко сокращалось время средней продолжительности жизни

крыс при ежедневном поступлении в организм по 2,0; 4,0 и 5,0 мккюри радиостронция. Затем следуют группы животных, получавших по 1,0 и 0,5 мккюри изотопа. Все остальные кривые, характеризующие смертность крыс, тесно переплетались между собой и мало чем отличались от контроля.

Результаты изменения средней продолжительности жизни подопытных животных в зависимости от дозы приведены в табл. 2.

Таблица 2

Продолжительность жизни контрольной и подопытных групп животных

№ №	Стронций-90 (мккюри/сут.)	Д о з а ( ЛВ - 50 )		Средняя продолжительность жизни (ЛВ-50)
		рад	рад/сутки	
1	Контроль	-	-	557,2 <sub>+21,0</sub>
2	0,00005	2,4	0,003	543,5 <sub>+18,8</sub>
3	0,0005	12,0	0,03	524,9 <sub>+28,5</sub>
4	0,005	118,0	0,25	535,6 <sub>+19,2</sub>
5	0,05	1130	2,5	524,9 <sub>+29,2</sub>
6	0,5	7680	25,0	378,7 <sub>+15,2</sub>
7	1,0	12400	50,0	328,6 <sub>+11,3</sub>
8	2,0	13300	100	219,2 <sub>+10,8</sub>
9	4,0	19000	200	189,1 <sub>+ 9,2</sub>
10	5,0	18500	220	151,8 <sub>+ 7,3</sub>

Начальное снижение средней продолжительности жизни крыс (табл. 2) отмечено в подопытной группе животных, которые ежедневно получали по 0,5 мккюри стронция-90. К этому времени поглощенная доза в их критическом органе составила 7680 рад.

С целью выяснения величины пороговой дозы, которая не приводила к сокращению продолжительности жизни животных, материал был подвергнут статистической обработке при помощи корреляционного и регрессионного анализа. Результаты анализа ( $r = 0,74$ ) показали, что максимальной величиной поглощенной дозы в критическом органе, которая еще не оказывает влияние на продолжительность жизни крыс, является 4000 рад.

Анализируя зависимость этого показателя от мощности дозы (табл. 2) видно, что не на всех уровнях она играет ведущую роль. Так, например, в группах животных, которые ежедневно получали стронций-90 от 0,00005 мккюри до 0,05 мккюри в сутки не обнаружено влияния как поглощенной, так и мощностей доз. Не проявляет также своего влияния мощность дозы в диапазоне величин от 25 до 50 рад/сутки (суточное поступление изотопа от 0,5 до 1,0 мккюри). В этом случае, как видно из таблицы, время продолжительности жизни подопытных животных зависит от величины поглощенной дозы. Мощность дозы, как ведущий фактор в снижении средней продолжительности жизни, прослеживается лишь на уровнях выше 50 рад в сутки.

Причины смерти подопытных крыс, за исключением остеосарком, мало чем отличались от контрольных животных. Однако, течение патологических процессов у крыс, получавших стронций-90 выше 0,5 мккюри в сутки существенно отличалось от контрольных.

Любое воспаление сопровождалось нагноением, которое в последующем носило генерализованный характер.

Спонтанных случаев остеосарком у контрольных крыс и у животных, получавших стронций-90 в количествах от 0,00005 мккюри до 0,05 мккюри в сутки в течение всей их жизни не отмечено, хотя под наблюдением было около 12000 крыс.

Первая остеосаркома, относительно времени, появилась через 175 дней от начала опыта у крыс, получавших стронций-90 ежедневно по 5,0 мккюри в сутки (табл. 3).

Частота образования остеосарком у подопытных животных

Таблица 3

Стронций-90 (мккюри в сутки)	Число животных	Выход остеосарком		Латентный период (дни)	Минимальная доза (рад)	Средняя продолжительность жизни (дни)	
		количество	%			при наличии остеосарком	без остеосарком
0,5	50	13	26,0	275	5000	375,7 $\pm$ 18,0	384,8 $\pm$ 32,1
1,0	50	21	42,0	220	7000	351,6 $\pm$ 21,3	293,9 $\pm$ 28,4
2,0	50	8	16,0	260	18000	300,0 $\pm$ 19,8	273,4 $\pm$ 31,2
4,0	50	2	4,0	250	30000	287,5 $\pm$ 24,1	181,9
5,0	50	1	2,0	175	22000	175,0	151,8

Однако, минимальная доза в костной ткани, при которой зарегистрирована первая опухоль, составляла 5000 рад. Такая доза была сформирована в критическом органе крыс через 275 дней от начала поступления в организм стронция-90 по 0,5 мккюри в сутки.

Анализируя результаты, приведенные в табл. 3 видно, что время, необходимое для развития остеосарком (латентный период) не зависит от дозы. Следует отметить, что по этому вопросу в литературе единого мнения нет. Так, например, М.Финкель (1955)<sup>8</sup> не обнаружила зависимости времени латентного периода от дозы. Тогда как другие авторы<sup>9,10</sup> нашли определенную зависимость — чем выше доза, тем короче время, необходимое для развития опухоли.

Наиболее важным в этом вопросе являются такие параметры как частота появления опухолей у животных в зависимости от дозы, влияние остеосарком на изменение средней продолжительности жизни, а также наличие или отсутствие пороговых доз для развития бластомогенного процесса.

Литературные данные относительно частоты развития опухолей в зависимости от дозы колеблются в очень широких пределах. Это объясняется тем, что большинство авторов, определяя эту величину у мелких лабораторных животных, относят количество остеосарком к числу животных, переживших 200 дней. В этом случае получается искусственное завышение частоты опухолей, которое характеризует лишь случайно оставшуюся в живых часть животных, и никакого отношения не имеет к общей оценке этого явления.

Зависимость частоты появления остеосарком, как видно из табл. 3 и рис. 3, является прямой лишь в определенном интервале доз. При этом, минимальной канцерогенной концентрацией стронция-90 при ежедневном его поступлении в организм является 0,5 мккюри в сутки на крысу, оптимальное количество изотопа не превышало 1,0 мккюри в сутки, поглощенные дозы составляли 5000 и 7000 рад, соответственно. Последующее повышение поглощенной дозы в скелете животных сопровождалось резким сокращением выхода опухолей. Такое положение

часто объясняется тем, что мелкие лабораторные животные, в связи с их коротким периодом жизни, не доживают до полной реализации опухолей. Однако, сравнение времени латентного периода развития бластомогенного процесса по отношению к средней продолжительности жизни разных видов животных и человека показало, что у мышей, крыс, собак и человека время, необходимое для развития остеосарком составляет от 10 до 30% их средней продолжительности жизни. Следовательно, эти данные могут свидетельствовать о том, что реализация лучевого канцерогенеза у разных видов животных происходит с такой же закономерностью как и у человека. Подтверждением этого является тот факт, что у крыс, получавших ежедневно по 4,0 мкюри стронция-90, средняя продолжительность жизни по сравнению с предыдущей группой животных была короче лишь на 15%, тогда как выход опухолей у них сокращался в 4 раза. Об этом же свидетельствует и другой пример. На кривых смертности (рис. 2) точками обозначено время появления опухолей относительно величины суточного поступления стронция-90. Из графика видно, что минимально необходимое время для развития остеосарком переживали 90, 80, 60 и 30% животных, соответственно получавших радиостронций по 1, 2, 4 и 5 мкюри в сутки, тогда как частота появления у них опухолей составляла 42, 16, 4 и 2%.

Следовательно, есть все основания считать, что развитие остеосарком определяет не продолжительность жизни животных, а величина дозы. Уменьшение выхода опухолей в подопытных группах крыс, которые ежедневно получали стронций-90 по 2 мкюри и выше, обусловлено величиной мощности дозы в критическом органе, которые были настолько велики, что приводили к преобладанию в кости процессов разрушения над восстановлением. Бластомогенный же процесс, как известно, всегда идет на базе восстановления.

Определенный интерес в практическом отношении представляет оценка влияния частоты появления остеосарком на среднюю продолжительность жизни подопытных животных.

В табл. 3 приведены данные, характеризующие среднюю продолжительность жизни животных, погибших от остеосарком и крыс, смерть которых наступила от различных других причин. Сравнивая эти результаты видно, что частота появления у животных остеосарком не оказывает влияния на величину средней продолжительности жизни. По-видимому, ведущими в сокращении продолжительности жизни являются другие причины, обусловленные радиационным поражением различных систем организма, и в первую очередь, кроветворной системы. Для наглядности на рис. 4 приведены кривые распределения смертности животных от остеосарком и других причин.

Как видно из графика, удельный вес остеосарком, по сравнению с другими причинами, приводящими к смерти животных, относительно не велик. Это может свидетельствовать о том, что частота появления остеосарком самостоятельного значения в сокращении продолжительности жизни не имеет, а её по-видимому, следует рассматривать как любую другую причину, приводящую к гибели животных. Если это так, то становится очевидным, что для развития остеосарком требуются определенные условия в критическом органе.

С целью определения пороговой дозы, на рис. 4 (в) сделана попытка экстраполяции частоты появления остеосарком в область малых доз. Как видно из графика, для начального развития бластомогенного процесса в костных тканях требуется поглощенная доза не менее 3,6 крад, тогда как пороговая доза по критерию продолжительности жизни равна 0,4.

Следует отметить, что вопрос о наличии или отсутствии пороговых доз для развития бластомогенного процесса до настоящего времени остаётся спорным, хотя все экспериментальные исследования однозначно относят его к пороговым реакциям. Вместе с тем, МКРЗ,



на основании теоретических и генетических исследований, принято считать остеосаркомы лучевой этиологии непороговыми реакциями. Исходя из этого, остеосаркомы следует рассматривать наиболее чувствительным критерием оценки опасности поражения стронцием-90 популяции людей.

Учитывая важность этого показателя мы попытались оценить его чувствительность относительно других критериев, характеризующих биологическое действие радиостронция (табл. 4).

Таблица 4

Зависимость доза-эффект

Э ф ф е к т	Пороговая доза (рад)	Э ф ф е к т	Пороговая доза (рад)
Лейкопения	300	Поражение глаз	2000
Нейтрофилопения	600	Цитопения миелоидн. ростка	3000
Цитопения костного мозга	1000	Сокращение продолжит. жизни	4000
Нарушение эстральн. цикла	1000	Л е й к о з н	4500
Нарушение остеогенеза	2000	Остеосаркомы	5000
Цитопения эритроидн. ростка	2000		

Из приведенных в табл. 4 результатов видно, что частоту появления остеосарком, при всех прочих равных условиях, отнести по чувствительности на первое место не представляется возможным. Подтверждением этому могут также служить данные различных авторов <sup>7, 11</sup>, приведенные на рис. 3, которые свидетельствуют о том, что появление остеосарком у крыс и собак при однократном и хроническом поступлении в организм стронция-90 происходит в диапазоне доз, равных 5-7 килорад. Эксперименты, проведенные <sup>3, 12, 7, 13, 14</sup> на собаках и миниатюрных свиньях в условиях хронического поражения их стронцием-90, также показывают, что начало проявления бластомогенного процесса лежит в пределах больших доз (7-15 крад).

Для сравнения на рис. 3 приведены сведения <sup>15</sup> продолжительного наблюдения за людьми, в скелете которых был аккумулирован радий-226. Эти данные убедительно показывают, что остеосаркомы для своего развития требуют определенной дозы воздействия. Следовательно, оценивая опасность поражения стронцием-90 человеческой популяции по критерию частоты появления остеосарком, по видимому, следует вводить некоторый коэффициент запаса, который бы этот эффект переносил на первое место. В противном случае, тем более с точки зрения беспороговости этого процесса, оценка риска появления остеосарком может искусственно скрыть все другие эффекты, которые играют важную роль в продолжительности жизни животных и человека.

Таким образом, анализ собственных и литературных данных показал, что частота появления остеосарком, индуцированных стронцием-90, носит линейный характер в диапазоне от минимальных до оптимальных бластомогенных доз. Дальнейшее увеличение мощности и поглощенной дозы характеризуется обратной зависимостью.

Остеосаркомы не снижают среднюю продолжительность жизни популяции и по значимости находятся на одном уровне с любой другой причиной, приводящей к гибели животного. Для развития бластомогенного процесса требуется определенная доза воздействия.

Оценка опасности поражения стронцием-90 человека должна учитывать место расположения критерия появления остеосарком относительно других эффектов, оказывающих влияние на продолжительность жизни.

## R E F E R E N C E S

1. Finkel M.P., Bergstrand P.I., Riskis B.O. (1960), "The consequences of the continuous ingestion of Sr-90 by mice". Radiology, 74, 3, 458-467.
2. Finkel M.P., Bergstrand P.I., Riskis B.O. (1961), "The latent period, incidence and growth of Sr-90 induced osteosarcomas in CFWo. I. and Cba mice. Radiology, 77, 2, 269-281.
3. McClellan R.O., W.J. Clarke, J.R. McKinney and L.K. Bustad (1962). "Preliminary observations on the biological effect of Sr-90 in miniature swine". Am.JVet.Res. 23, 910-912.
4. Clarke W.J., R.F. Palmer, E.B. Howard and P.L. Hackett (1970). "Strontium-90: Effects of chronic ingestion on farrowing performance of miniature swine". Sci. 169, 598-600.
5. Mole R.H. (1958). "The dose-response relationship in radiation: Carcinogenesis. Brit.Med.Bull., 14, 2, 184-188.
6. Шведов В.Л. (1968) "Экспериментальное обоснование предельно допустимого содержания стронция-90 в организме при хроническом его поступлении". Дисс. М., 1969.
7. Goldman M., R.Pool, M.H. Momeni, F. Wilson, R.J. Romer Williams, C.Chrisp, L.S Rosenblatt and L.K. Bustad (1972). "Quantitation of Sr-90 toxicity in dogs" Там же, 17-30.
8. Finkel M.P. (1955). "Internal emitters and tumor induction" Мирное использование атомной энергии. Материалы Международной конференции в Женеве, т. II, август, 197-203.
9. Стрельцова В.Н., Ю.И.Москалев (1964) "Бластомогенное действие ионизирующей радиации". Изд. "Медицина" Москва.
10. Литвинов Н.Н. (1964). "Радиационные поражения костной системы". Изд. "Медицина", Москва.
11. Краевский Н.А., Н.Н.Литвинов (1957) "Бластомогенное действие радиоактивного стронция". Мед. радиол., II, 5, 33-39.
12. Howard, E.B., W.J. Clarke, M.T. Karagianes and R.F. Palmer (1969), "Strontium-90 induced bone tumors in miniature swine". Rad.Res. 39, 594-607.
13. Ragan H.A., R.L. Buschbom, W.J. Clarke, P.L. Hackett and B.J. Mc.Clanahan, (1972). "Late effects of chronic Sr-90 ingestion in miniature swine", Second International Conference on Strontium metabolism, Glasgow and Strontian, 145-154.
14. Rosenblatt, L.S., N.H. Hetherington, M.Goldman and L.K. Bustad (1971). " Evaluation of tumor incidence following exposure to internal emitters by application of the logistic dose-response surface". Health Phys. 21, 869-875.
15. Evans (1967), цит. по Ю.И.Москалеву, Симпозиум "Отдаленные последствия при поражении остеотропными радионуклидами" Мед. радиол., 7, 1968, 59-74

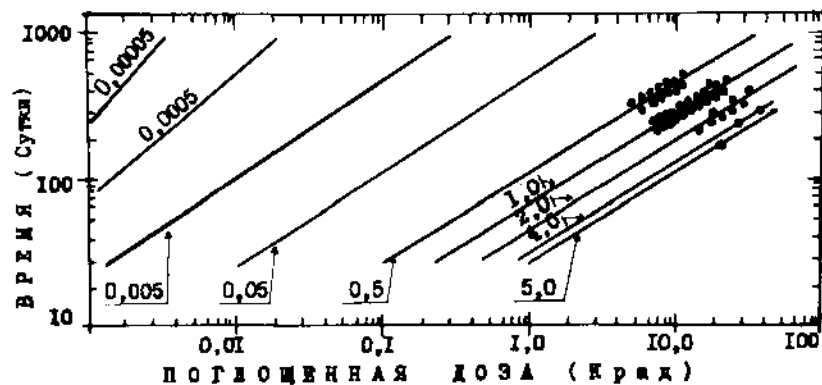


Рис. 1 Формирование поглощенной дозы в бедренной кости крыс в зависимости от суточной концентрации стронция-90 и времени его поступления.

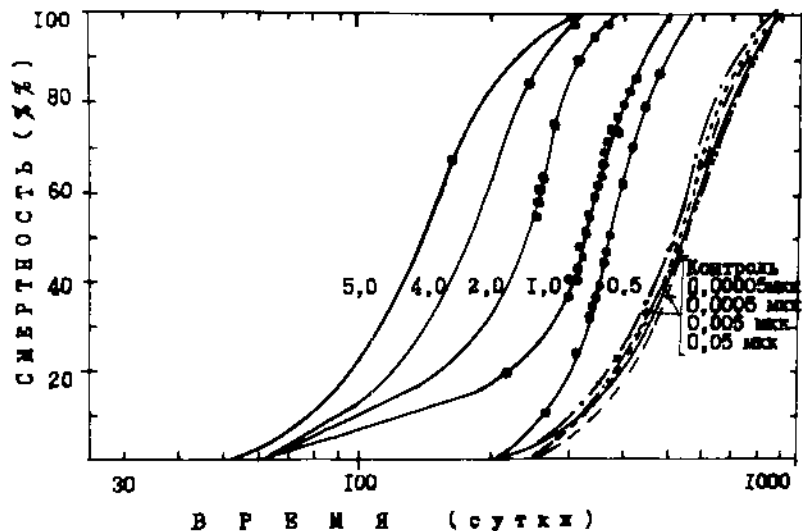


Рис. 2 Смертность крыс в зависимости от времени поступления в организм стронция-90.

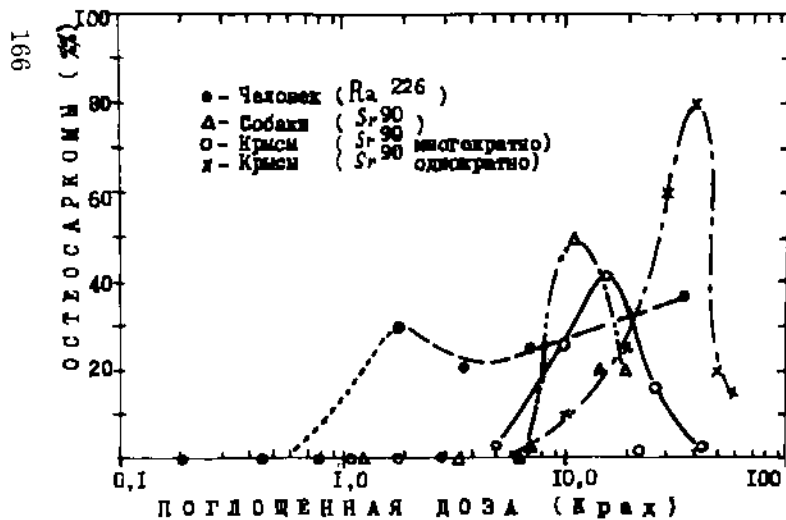


Рис. 3 Частота появления остеосарком в зависимости от поглощенной дозы.

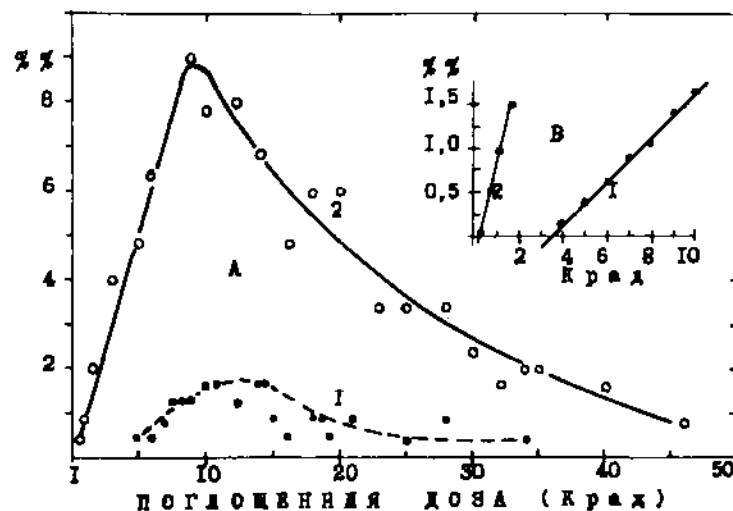


Рис. 4 Распределение случаев смерти от остеосарком (1) и других причин (2).

$$1. D_0 = 0,914 + 0,251D; D_0 = 3,6 \text{ крад}$$

$$2. D_0 = 0,742 + 1,793D - 0,1104 D^2; D_0 = 0,4 \text{ крад}$$

ОТДАЛЕННЫЕ ПОСЛЕДСТВИЯ ПРИ ПОРАЖЕНИИ  
ТРАНСУРАНОВЫМИ ЭЛЕМЕНТАМИ

Ю.И. Москалев, Э.И. Рудницкая, Л.А. Булдаков, А.П. Нифатов,  
Л.Г. Филиппова

Институт биофизики Министерства здравоохранения  
Москва, Союз Советских Социалистических Республик

Abstract

In the experiments on rats and dogs we have studied late effects of malignant and neomalignant development after the intake of various compounds of Pu-239, Am-241 and Np-237.

It has been shown that dogs and rats after the intake of transuranium elements develop osteosarcomas, leucosis, tumours of lungs, liver, kidneys and other organs, sclerotic processes (contracted liver, nephrosclerosis, pneumosclerosis) at the sites of deposition or transition of the isotope and septical processes of various localization.

After the intake of Pu-239 the minimum osteosarcomogenic dose is within 5 rad and after Am-241 - 17 rad.

The maximum frequency of osteosarcoma development after the intake of Am-241 is 33% in rats and 100% in dogs. With the decrease of dose accumulated in the bone tissue the relative osteosarcomogenic efficiency is increased in dogs as compared to rats from 3 to 30 times. When the doses are close to minimum effective ones the osteosarcomogenic activity of Pu-239 is five-fold that of Am-241. The development of leucosis after the intake of transuranium elements is more often and occurs at earlier periods relative to control animals. The maximum yield of kidney tumours (2-4,5%) is observed with Am-241 when the doses used are within 35-200 rad. After the inhalation of soluble compounds of Am-241 the frequency of pneumosclerosis and lung tumour development is lower as compared to Pu-239. The routes of Am-241 and Pu-239 intake into the body do not affect the values of carcinogenic doses for the particular tissue. The minimum osteosarcomogenic and leucomogenic dose for nitrate of Np-237 after intravenous injection is below 0.1 Ci/kg.

Введение

Хорошо известно, что при поражении ионизирующей радиацией могут возникать опухолевые и неопухолевые формы отдаленных последствий<sup>1-2</sup>. В настоящем сообщении представляются результаты экспериментальных исследований по изучению отдаленных послед-

ствий у крыс и собак при попадании в организм различных доз *Pu-239*, *Am-241*, № -237

### Результаты исследований

После инкорпорации трансураниевых элементов у животных чаще всего развиваются остеосаркомы<sup>1-3</sup>. Частота возникновения остеосарком и сроки гибели животных определяются количеством вводимой активности. В диапазоне доз от 30 до 60 рад при поражении *Pu-239* злокачественные опухоли костной ткани возникают после однократного интраперитонеального, подкожного, ингаляционного и длительного перорального введения.

После ингаляции плутоний пентакарбоната аммония остеосаркомы развились у 2 из 192 крыс, в скелете которых аккумуляровались дозы излучения, равные 3,6 и 6,4 рад, а после ингаляции цитрата *Pu-239* у 2 из 157 крыс, в скелете которых аккумуляровалась доза излучения, равная 16,6 рад. Остеосаркомы не наблюдались у контрольных крыс. Минимальная остеосаркомогенная доза при инкорпорации *Pu-239* находится в пределах 5 рад. Следует подчеркнуть, что продолжительность жизни крыс с остеосаркомами, в скелете которых аккумуляруются дозы излучения меньше 100 рад, обычно такая же как и контрольных животных, а иногда превышает ее. Так, например, после ингаляции цитрата *Pu-239* в количестве 0,008 мккюри и плутоний пентакарбоната аммония в количестве 0,045 мккюри средняя продолжительность жизни крыс с остеосаркомами, в скелете которых аккумуляровались дозы излучения, равные соответственно 16,6 и 25,2 рад были равны  $787 \pm 69$  и  $736 \pm 35$  дней. Продолжительность жизни экспериментальных крыс без остеосарком оказались равной  $635 \pm 3$  и  $519 \pm 11$  дней, а контрольных крыс не получивших *Pu-239*,  $571 \pm 8$  дней. Опыты по изучению отдаленных последствий при инкорпорации разных солей *Am-241* поставлены на 1716 крысах и 55 собаках.

После инкорпорации америция-241 у крыс и собак также возникают остеосаркомы, частота и скорость развития которых зависит от радиационной дозы в костях. Частота возникновения остеосарком у крыс при введении америция-241 в виде лимоннокислой или азотнокислой соли также не зависит от пути поступления изотопа. При введении азотнокислой соли *Am-241* наибольшая частота остеосарком у крыс обнаруживается при аккумуляции в скелете доз порядка 700-750 рад, в то время как в случае введения лимоннокислой соли при дозах 1600-3800 рад. При дальнейшем увеличении дозы процент остеосарком падает, что обусловлено вероятно сокращением продолжительности жизни крыс или тормозящим действием массивного альфа-облучения на регенерацию костной ткани. Снижение радиационной дозы также ведет к понижению частоты выхода остеосарком, которое сопровождается удлинением латентного периода необходимого для развития опухолей. Достоверное увеличение частоты остеосарком у крыс при поражении *Am-241* обнаружено при аккумуляции в скелете за время жизни дозы, равной 17 рад.

Частота возникновения остеосарком у крыс значительно ниже, чем у собак. Если у крыс частота при инкорпорации *Am-241* не превышает 33%, то у собак при дозах в скелете от 344-3320 рад остеосаркомы обнаруживаются у 100% животных.

---

I/ В работе приводятся средние дозы, аккумулярованные тканью к моменту гибели животного.

Относительно сравнительной чувствительности собак к остеосаркомогенному действию  $Pu-239$  и  $Am-241$  пока говорить преждевременно. Можно отметить, что остеосаркомы возникают у 100% собак при аккумуляции в скелете доз равных 330–3400 рад. При одинаковых тканевых дозах у собак частота остеосарком в 3–30 раз выше, чем у крыс (рис. 1). Наряду с остеосаркомами у собак, павших в отдаленные сроки после внутривенного введения  $Am-241$ , обнаружили развитие лейкозов, мезотелиомы брюшной полости, эндотелиомы печени, светлоклеточного рака почки. Мезотелиома брюшной полости (рис. 2) и эндотелиома печени развились у собаки, павшей на 882 сутки, после инъекции  $Am-241$  в дозе 1 мкюри/кг. В печени и скелете этой собаки, аккумуляровались дозы излучения, соответственно, 186 и 1000 рад. После инъекции такой же дозы  $Am-241$  у собаки, павшей на 1429 сутки от начала опыта, обнаружили остеосаркомы бедра с множественными метастазами и светлоклеточный рак почки; доза в печени и скелете к моменту гибели составляла соответственно 236 и 1600 рад.

Сравнительный анализ экспериментальных данных свидетельствует о существовании некоторых различий в остеосаркомогенной эффективности  $Pu-239$  и  $Am-241$  (рис. 1). При инкорпорации  $Pu-239$  остеосаркомы у животных возникают по сравнению с  $Am-241$  при меньших средних дозах в скелете. При дозах близких к минимально эффективным частота остеосарком при поражении  $Pu-239$  (2,5%), заметно выше, чем при воздействии  $Am-241$  (0,5%). Максимальная частота остеосарком ( $\approx 30-40\%$ ) у крыс при поражении  $Am-241$  и  $Pu-239$  оказалась практически одинаковой, и наблюдалась при аккумуляции в костной ткани примерно одинаковых тканевых доз (рис. 1). При низких дозах остеосаркомогенная активность  $Pu-239$  приблизительно в 5 раз выше таковой у  $Am-241$ . Одним из моментов, определяющих более низкую частоту развития остеосарком у крыс при введении  $Am-241$  по сравнению с  $Pu-239$ , является различие в микрогеометрии распределения их в костной ткани<sup>5-8</sup>. Следует подчеркнуть, что при инкорпорации подостро и хронически эффективных доз  $Am-241$  у подопытных животных (крысы, кролики, собаки) развились в печени изменения типа цирроза с выраженной портальной гипертензией на фоне своеобразных изменений сосудистой системы<sup>9-10</sup>.

У крыс после инкорпорации  $Am-241$  часто возникают лейкозы (рис. 3), радиационные дозы в скелете в пределах от 325 до 4650 рад. Лейкозы у подопытных животных по сравнению с контрольной группой возникают в более ранние сроки. Средний латентный период для лейкоза у контрольных крыс составляет 600 дней, а у подопытных он не превышает 500 суток. После инкорпорации  $Am-241$  лейкозы чаще всего обнаруживаются у крыс, павших на 240–400 суток, т.е. при сокращении продолжительности жизни на 30%. Интересно отметить, что лейкозы у собак также развивались значительно раньше, чем остеосаркомы при выраженном сокращении продолжительности жизни. Два случая лейкоза были обнаружены у собак павших на 204 и 371 сутки после внутривенного введения хлорида  $Am-241$  в дозах 7,5 и 2,5 мкюри/кг при аккумуляции в костной ткани доз излучения, равных, соответственно 1870 и 1130 рад. Материалы рис. 3 свидетельствуют об отсутствии линейной зависимости доза-эффект для лейкемогенного действия  $Am-241$  и существовании практического порога при дозах равных и ниже 70 рад на скелет. Истинную дозу на костный мозг предстоит оценить. Как видно из табл. 1 среднетканевые дозы в почках, при которых найдено учащение опухолей почек, сравнительно малы; 1,7–2750 рад. Максимальная частота опухолей почек обнаружена

при дозах 34–68 рад. Низкие тканевые дозы, при которых возникают опухоли, не являются истинными поскольку рассчитаны без учета неравномерности распределения америция в почке. Об этом свидетельствуют результаты гистоауторадиографических исследований (рис. 4). Повидимому, радиационные дозы в отдельных участках почки могут быть в 10–100 раз выше, чем среднетканевые, рассчитанные по данным средней концентрации изотопа в почечной ткани. Опухоли почек возникают не только у крыс, но и у собак, о чем упоминалось выше. Особенно высок процент опухолей почек был обнаружен у крыс при пероральном введении  $Am-241$ , когда в почках аккумуляровались дозы 84 и 268 рад (табл. I).

После ингаляции (собаки) или интратрахеальным введением (крысам)  $Am-241$  у животных развивается различная патология со стороны легких. Это слабо выраженный пневмосклероз и опухоли легких. Тяжесть и частота этих заболеваний при поражении  $Am-241$  несравненно ниже, чем после ингаляции  $Pu-239$ . Относительно меньшая частота возникновения патологии в легочной ткани при поражении цитратными и азотнокислыми соединениями  $Am-241$  обусловлена сравнительно быстрой элиминацией изотопа из легких <sup>11</sup>. Анализ материалов по бластомогенному действию  $Am-241$  показывает, что наряду с возникновением "типичных" для америция опухолей скелета, почек и лейкозов у животных возрастает так же суммарная частота опухолей мягких тканей. Эксперименты по биологическому действию  $№-237$  были проведены с внутривенным введением крысам (156 крыс) азотнокислой соли. Внутривенное введение  $№-237$  в дозах  $\leq 0,2$  мкк/кг не влияет на естественную продолжительность жизни животных. У животных павших в отдаленные сроки, наблюдается вся гамма отдаленных последствий, характерных для инкорпорации трансураниевых элементов <sup>12</sup>.

Остеосаркомы развивались у крыс после инкорпорации  $№-237$  в дозах 0,1; 0,5; 1 и 5 мккюри/кг. У части животных опухоли были множественными. При наименьшей из использованных доз  $№-237$  остеосаркомы были обнаружены у 2-х из 7 крыс, павших на 780 и 782 сутки, в скелете которых аккумуляровались дозы излучения, равные соответственно, 98 и 104 рад. Указанные дозы, повидимому не являются минимально остеосаркомогенными. Описано возникновение остеосарком при аккумуляции в скелете крыс и более низких доз <sup>13</sup>. Учащение случаев возникновения лейкозов типа гемоцитобластозов отмечено у крыс после внутривенного введения изотопа в дозах 0,5; 2,0 и 5,0 мккюри/кг. При инкорпорации нуклида в дозе 0,5 мккюри/кг лейкозы были обнаружены у 3 из 13 крыс, павших на 164, 367 и 420 сутки от начала опыта. В костной ткани этих крыс аккумуляровались дозы излучения равные 100–300 рад. При введении  $№-237$  в дозах 0,1 и 0,2 мккюри/кг у крыс, павших в отдаленные сроки развития лейкоза не наблюдали.

#### Заключение

Результаты проведенных исследований свидетельствуют о высокой биологической эффективности  $Pu-239$ ,  $Am-241$ ,  $№-237$ . Среди неопухолевых форм преобладают склеротические процессы в местах депонирования изотопа или транзита (цирроз печени, нефросклероз, пневмосклероз), различные воспалительные процессы. Опухолевые формы преимущественно наблюдаются в местах наибольшего отложения изотопа и среди них чаще всего остеосаркомы, лейкозы, новообразование печени, почек и раки легких. Различные уровни отложения  $Pu-239$  и  $Am-241$  в печени и неодинаковый характер микрораспределения в костной ткани, вероятно, лежат в основе относительно большей остеосаркомогенной эффективности у крыс  $Pu-239$  по сравнению с  $Am-241$ . Большая частота выхода опухолей почек в экспериментах с  $Am-241$  также, повидимому, обусловлена различными уровнями

депонирования изотопа в почках и характером его микрораспределения. В основе большей остеосаркомогенной эффективности для собак по сравнению с крысами также вероятно лежат различия в микрогеометрии поглощения альфа-излучения в костной ткани сравниваемых животных.

### Библиография

1. В.Н. Стрельцова (1964). Бластоогенное действие ионизирующей радиации. Москва, Медицина.
2. Ю.И. Москалев (1971). Отдаленные последствия лучевых поражений. Сб. работ под ред. Ю.И. Москалева. Москва, Атомиздат.
3. Л.А. Булдаков, Э.Р. Любчанский, Ю.И. Москалев, Л.П. Нифатов (1969). Проблемы токсикологии плутония. Москва, Атомиздат.
4. Yu.I. Moscalev (1972). Pu<sup>239</sup> Problems of its biological effect. Health Physics, v. 22, 723-729.
5. Ю.И. Москалев, Э.И. Рудницкая (1971). Некоторые аспекты биологического действия Am-241. В кн. "Отдаленные последствия лучевых поражений под ред. Ю.И. Москалева, с. 509-517.
6. D.M. Taylor (1962). Some aspects of the comparative metabolism of Pu and Am in rats. Health Physics. 8, 6, 673.
7. R.D. Lloyd, C.W. Mays, and as. Americium-241 studies in beagles. Health Phys. 18, 2, 149-156 (1970).
8. Э.И. Рудницкая (1971). Морфология некоторых процессов в отдаленные сроки при поражении Am-241. В кн. "Отдаленные последствия лучевых поражений", ст. 408-414.
9. Э.И. Рудницкая, Ю.И. Москалев (1970). Микрораспределение и морфологические изменения у крыс при внутривенном введении америция-241. Ж. Радиобиология, т. X, в. 4, 570-57.
10. Э.И. Рудницкая, Ю.И. Москалев (1972). Действие подостроэф-фективных доз Am-241 на кроликов. Ж. Радиобиология, т. X, 788-790.
11. A.P. Nifatov, L.A. Buldakov, and as. (1972). Some late effects after a single inhalation of Pu<sup>239</sup> and Am<sup>241</sup> in dogs. Health Phys. v. 22, p. 875.
12. Ю.И. Москалев, Э.И. Рудницкая и др. (1971). Биологическое действие Np-237, ж. Гигиена и санитария, 2, 42-47.
13. T.G. Levdiik, V.K. Lemberg, L.A. Buldakov and as. (1972). Biological effectiveness of Np<sup>237</sup>. Health Phys. v. 22, 643-645.

### Подписи к рисункам

1. Частота возникновения остеосарком у крыс и собак при поражении Pu-239 и Am-241, в зависимости от дозы, аккумулярованной в костной ткани к моменту гибели. 1/Am-241 собаки; 2/Pu-239 собаки; 3/Am-241 (цитрат)-крысы; 4/Am-241 (нитрат)-крысы; 5/Pu-239 (цитрат)-крысы.
2. Мезотелиома брюшной полости. Окр. гем.-эоз., х400.
3. Частота возникновения лейкозов у крыс в зависимости от дозы, создаваемой Am-241 в костной ткани. х-контроль; о-опыт.
4. Гистоавтограмма мозгового слоя почки AmCl<sub>3</sub>, доза 12,5 мккюри/кг, смерть на 100 сутки. Окраска гем.-эоз., х140.



БИОЛОГИЧЕСКИЕ ЭФФЕКТЫ ПРИ СОЧЕТАННЫХ  
РАДИАЦИОННЫХ ВОЗДЕЙСТВИЯХ

В.С. Калистратова, И.С. Кацапов, В.И. Трифонов  
Институт биофизики Министерства здравоохранения  
Москва, Союз Советских Социалистических Республик

Abstract

In experiments on white rats, we have studied the combined effects of  $^{131}\text{I}$ ,  $^{241}\text{Am}$ , HTO and of external radiation sources. The results obtained indicate that combining of radioactive and whole-body external gamma-irradiation in various combinations can be expressed differently, i.e. either by summation of the effects or by their increase or decrease.

Проблема изучения комбинированных форм лучевого поражения представляет большой научный и практический интерес и является мало изученной. Одним из актуальных вопросов этой проблемы является вопрос о комбинированном действии внешнего гамма-облучения и радиоактивных изотопов. В этой связи нами было изучено биологическое действие общего внешнего гамма-излучения и радиоактивных изотопов, обладающих различной тропностью к органам и тканям: йод- $^{131}\text{I}$ , окись трития и америций- $^{241}\text{Am}$ . Схематически каждый опыт с разными уровнями доз можно представить так: общее внешнее гамма-облучение - радиоактивный изотоп - общее внешнее гамма-облучение + радиоактивный изотоп - интактные животные. При изучении комбинированного действия йода- $^{131}\text{I}$  и внешнего гамма-излучения использовали соотношение доз, которое имело место у детей жителей Маршалловых островов<sup>1</sup> (щитовидная железа кумулировала дозу 1400 rad, весь организм 175 rad). В нашем эксперименте были использованы следующие комбинации: гамма-излучение Co-60 100 R + 1000 rad I- $^{131}\text{I}$  на щитовидную железу; гамма-излучение Co-60 300 R + 3000 rad I- $^{131}\text{I}$  на щитовидную железу; гамма-излучение Co-60 800 R + 8000 rad на щитовидную железу. Йод- $^{131}\text{I}$  крысам вводили через рот через 30 min после облучения. Дозы внешнего гамма-излучения и америция- $^{241}\text{Am}$ , внешнего гамма-излучения и окиси трития подбирались таким образом, чтобы они соответствовали остро-, подостро- и хронически-эффективным дозам каждого фактора. Америций- $^{241}\text{Am}$  вводили внутривенно, а окись трития внутрибрюшинно через 30 min после гамма-облучения. Были использованы следующие сочетания:  
гамма-излучение Cs-137 600 R + 0,1  $\mu\text{Ci/g}$  Am-241  
гамма-излучение Cs-137 300 R + 0,05  $\mu\text{Ci/g}$  Am-241

гамма-излучение Cs-137	150 R	+	0,025 $\mu$ Ci/g	Am-241
гамма-излучение Co-60	600 R	+	1,0 $m$ Ci/g	НТО
гамма-излучение Co-60	300 R	+	0,5 $m$ Ci/g	НТО
гамма-излучение Co-60	150 R	+	0,25 $m$ Ci/g	НТО

В контрольной группе были животные, подвергнутые изолированному действию того или иного радиационного фактора и интактные животные. Экспериментальные исследования проведены на 800 белых беспородных крысах. Биохимические исследования проводили у животных в условиях 20-часового голодания. Животных обследовали в динамике по срокам: 1, 3, 7, 14, 21 сутки, 1, 3, 6, 12 месяцев. В сыворотке крови определяли холестерин<sup>2</sup>, общие липиды<sup>3</sup>,  $\beta$ -липопротеиды<sup>4</sup>, пировиноградную кислоту<sup>5</sup>, активность холинэстеразы крови<sup>6</sup> и щелочной фосфатазы<sup>7</sup>, а также гликоген печени<sup>8</sup> и сахар крови<sup>9</sup>. Была прослежена также выживаемость животных.

Результаты исследований показали, что наиболее отчетливые эффекты при сочетании действия внешнего гамма-излучения и радиоактивного йода были получены в опытах, когда дозы радиоактивного йода на щитовидную железу превышали остро- и подостро-эффективную дозу внешнего облучения на весь организм в 10 раз. Ранее проведенными исследованиями<sup>10</sup> показано, что биологическое действие йода-131 в дозах, разрушающих щитовидную железу (200000 рад) при сочетании с внешним гамма-облучением (300 R) определяются в основном состоянием щитовидной железы, развитием явлений атиреоза. При сочетанном действии йода-131 и внешнего гамма-излучения, когда тканевая доза от йода-131 на щитовидную железу превышала дозу внешнего гамма-облучения в 5 и 10 раз отмечали более сложные закономерности, т.е. эффект сочетанного действия по ряду показателей проявлялся различно. Изученные нами показатели можно разделить на две группы. Первая группа - это показатели, в изменение которых основной вклад вносило внешнее гамма-излучение: сахар крови, гликоген печени и мышц, пировиноградная кислота сыворотки крови, холинэстеразная активность крови. Рис. 1.

Ко второй группе мы относим показатели, изменение которых характеризует взаимноослабляющее действие двух радиационных факторов - это количество общего холестерина,  $\beta$ -липопротеидов. Рис. 2. Взаимноослабляющее действие радиационных факторов может быть следствием разной направленности реакций организма после внешнего гамма-облучения или введения радиоактивного йода. Схематически изложенное можно представить следующим образом: при гипотиреозе увеличение гликогена в печени с одновременным снижением липолиза в жировых депо способствует усилению процессов синтеза белковожировых комплексов в печени, в результате чего увеличивается количество  $\beta$ -липопротеидов крови. При общем внешнем гамма-облучении в остро-эффективных дозах снижение количества  $\beta$ -липопротеидов связано с истощением запасов гликогена в печени, нарушением синтеза жировых фракций в печени. О том, что причиной так называемого "защитного эффекта" является гипотиреоз, свидетельствуют также опыты с удалением щитовидной железы. У животных с удаленной щитовидной железой оперативным путем после введения йода-131 и общего  $\gamma$ -облучения отмечается тот же "защитный эффект" при комбинированном действии, что и после поступления радиоактивного йода в дозах разрушающих щитовидную железу при одновременном внешнем облучении. Необходимо отметить, что все реакции организма при комбинированном действии наиболее устойчивы и определены после 14 суток, что связано с тем, что в ранние сроки наслаиваются неспецифические реакции. Комбинированное действие радиационных факторов на уровнях малых доз (100 R внешнего облучения и 1000 рад от радиоактивного йода

на щитовидную железу) не отличалась от изолированных воздействий по всем изученным показателям на протяжении 12 месяцев наблюдения. При совместном действии на организм двух факторов — инкорпорированного радиоактивного америция и внешнего  $\gamma$ -облучения в остро- и подостро-эффективных дозах — происходит суммация или усиление эффекта. Мы попытались провести анализ эффекта при комбинированном действии не только путем сравнения изменений показателей при изолированных воздействиях с изменением таковых при совместном действии и с контролем, а также путем сравнения комбинированного поражения с поражением от удвоенной дозы каждого радиационного фактора. Проявление эффекта суммации приходится на 7,14,30 сутки. Особенно отчетливо это выявляется для показателей, связанных с функциональным состоянием печени: гликоген печени, холинэстеразная активность и количество  $\Lambda$ -липопротеидов сыворотки крови. Табл. I. По мере накопления тканевой дозы от америция-241 в изменение ряда показателей при сочетанном действии основной вклад вносит внутренний фактор радиационного воздействия. Так, уровень молочной кислоты при действии америция в дозе  $1 \mu\text{Ci/g}$  составит  $2 \text{ mg\%}$ ; в группе с комбинированным поражением ( $300 \text{ R} + 0,05 \mu\text{Ci/g Am-241}$ ) —  $0 \text{ mg\%}$ , в то время как при внешнем облучении в дозе  $600 \text{ R}$  количество молочной кислоты составило  $22,5 \text{ mg\%}$ , в контроле —  $38 \text{ mg\%}$ .

Анализ выживаемости показал, что при комбинированных поражениях в остро- и подостро-эффективных дозах наблюдается усиление эффекта. Так при одновременном действии  $300 \text{ R} + 0,05 \mu\text{Ci/g Am}$  продолжительность жизни животных составляла  $54 \pm 1$  дня, а удвоенная доза внешнего  $\gamma$ -излучения  $600 \text{ R}$  —  $10 \pm 2$  дня. Табл. 2.

Хронически-эффективные дозы этих радиационных воздействий по показателям обмена веществ не вызвали суммации эффекта. Однако, по критерию продолжительности жизни на уровне малых доз выявлен эффект суммации. Так средняя продолжительность жизни животных при воздействии в дозе  $150 \text{ R}$  и введении америция-241 в дозе  $0,025 \mu\text{Ci/g}$  составляет  $554 \pm 33$  дня при воздействии внешнего  $\gamma$ -облучения в дозе  $300 \text{ R}$  —  $524 \pm 33$  дня. Различия в этих группах статистически не достоверны.

После поступления в организм окиси трития и одновременно внешнего  $\gamma$ -облучения, когда имеется равномерное распределение дозы по органам и тканям, комбинированное радиационное воздействие приводит к полной суммации биологических эффектов. В табл. 3 представлены изменения ряда показателей обмена веществ на 7 сутки. Результаты исследований показали, что изменения этих показателей у животных с комбинированным поражением, по сравнению с животными, подвергнутыми изолированному воздействию удвоенной дозой внешнего  $\gamma$ -облучения или окисью трития статистически не достоверны. Например, активность щелочной фосфатазы при изолированных воздействиях на 7 сутки составляла  $1,4 \text{ mg\%}$  ( $300 \text{ R}$ ) и  $1,2 \text{ mg\%}$  ( $0,5 \mu\text{Ci/g}$ ), что не отличалось от контроля ( $1,4 \text{ mg\%}$ ), при комбинированном воздействии она была достоверно ниже, чем в указанных группах и составляла  $0,89 \text{ mg\%}$ , в то же время она не отличалась от удвоенной дозы изолированного воздействия и составляла  $0,83 \text{ mg\%}$   $600 \text{ R}$   $\gamma\text{Co-60}$  и  $0,7 \text{ mg\%}$  —  $1 \mu\text{Ci/g HTO}$ . При анализе зависимости средней продолжительности жизни от дозы внешнего гамма-излучения и окиси трития при сочетанном и изолированном воздействии (табл. 4) обнаружили, что по критерию продолжительности жизни при сочетанном воздействии в остро- и подостро-эффективных дозах наблюдается эффект полной суммации. На уровне хронически-эффективных доз при сочетанном действии однозначного эффекта не выявлено.

### Заключение

Таким образом, при сочетанных воздействиях внешнего  $\gamma$ -облучения и изотопов на уровне остро- и подостро-эффективных доз наиболее отчетливый эффект выявляется на 7-14 сутки, когда реакции организма становятся наиболее стабильными. При этом наиболее показательными оказались результаты опытов с подострыми дозами внешнего  $\gamma$ -облучения и радиоактивных изотопов, что связано с возможностью более длительного наблюдения за животными. Для хронически-эффективных доз по большинству показателей четких закономерностей выявить не удалось. Для окончательного суждения представляли интерес на уровне этих доз изучение отдаленных эффектов, главным образом бластомогенных.

Из представленных данных так же вытекает, что в определении эффекта сочетанного действия большая роль принадлежит тропности изотопов к той или иной ткани. Именно поэтому гипофункция щитовидной железы является определяющим фактором в эффекте сочетанного действия и "защитный эффект" на уровне определенных доз наблюдается при оценке показателей, связанных с изменением ее функции. По этой же причине суммация и усиление эффекта биологического действия наиболее выражены для показателей, которые связаны с функциональным состоянием печени - критического органа при сочетанном действии внешнего  $\gamma$ -облучения и америция-241. Одинаковая направленность изменений в обмене веществ при действии на организм внешнего облучения и окиси трития способствует полной суммации эффекта на уровне организма.

### Библиография

1. "Pathological effects of thyroid irradiation" . Federal Radiation Council Washington.
2. М.А. Левченко (1955). Метод определения холестерина в крови. Лаб. дело 2, 28.
3. H.G. Kunkel, S.H. Ahrens (1948). I. "Gastroenterology", 4, 499.
4. М. Ледвина (1960). Определение  $\beta$ -липопротеидов турбидиметрическим методом. Лабораторное дело 3, 13.
5. А.М. Петрунькина (1961). Практическая биохимия. III изд. Медгиз, Ленинград.
6. S. Hestrin (1949). I. "Biochem". 180, 249.
7. Н.В. Новикова, Е.П. Прокофьева (1964). Лабораторное дело. 12, 713.
8. S. Seifter (1950). "Arch. Bioch.", 25, 1, 191.
9. А.Н. Волков (1965). Методика определения сахара крови на фотоэлектроколориметре. Врачебное дело. 5, 147.
10. В.С. Калистратова, Г.С. Оксентук, В.Ф. Крюк (1970). Особенности сочетанного действия йода-131 и лучей Co-60. В сб. "Распределение, кинетика обмена и биологическое действие радиоактивных изотопов йода". Медицина, 186.

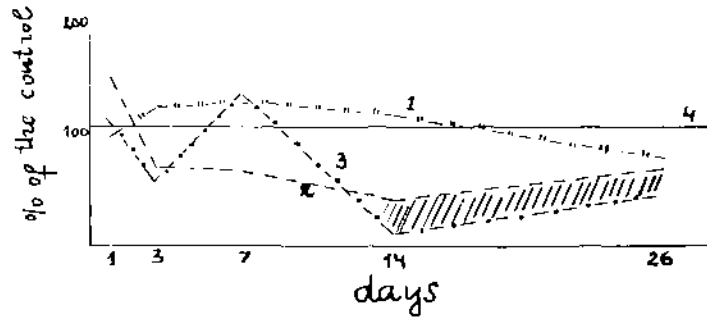


Рис. 1. Изменение количества гликогена в печени у крыс.  
 1 - I-131; 2 -  $\gamma$ Co-60; 3 -  $\gamma$ Co-60 + I-131

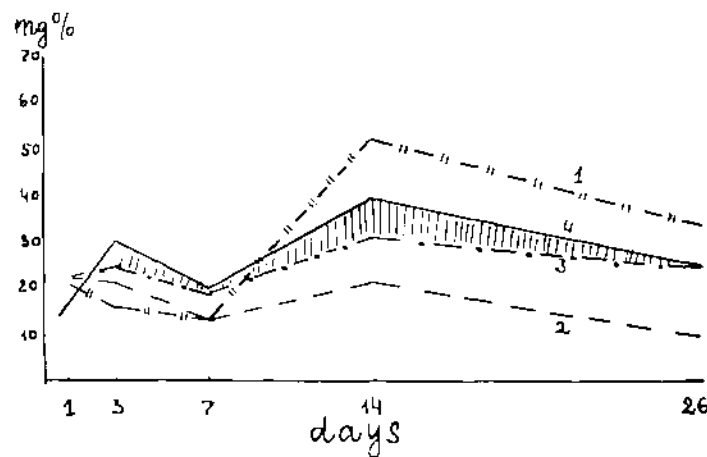


Рис. 2. Изменение содержания  $\beta$ -липопротеидов крови у крыс. Обозначения как на рис. 1.

ТАБЛИЦА 1

ИЗМЕНЕНИЕ ПОКАЗАТЕЛЕЙ ОБМЕНА ВЕЩЕСТВ НА 14 СУТОК ПОСЛЕ СОЧЕТАЮЩЕГО И ИЗОЛИРОВАННОГО ВОЗДЕЙСТВИЯ  
 АМЕРИЦИЯ-241 И ВНЕШНЕГО  $\gamma$  - ОБЛУЧЕНИЯ И ПОДАСТРОФИЧНЫХ ДОЗ

ПОКАЗАТЕЛЬ	300 p $\gamma$ Co <sup>60</sup>	0,05 $\mu$ Ci/g Am. 241	300 p $\gamma$ Co <sup>60</sup> 0,05 $\mu$ Ci/g Am. 241	600 p $\gamma$ Co <sup>60</sup>	0,1 $\mu$ Ci/g Am. 241	Контроль
Гликоген печени g%	1,76(1,0-2,5)	1,66(1,4+1,92)	1,34 <sup>M</sup> (1,04+1,68)	2,38(2,1+2,66)	1,46 <sup>M</sup> (1,03+1,89)	3,4(3,18-3,63)
Гликоген мышц g%	0,22(0,16-0,28)	0,33(0,26+0,39)	0,2(0,19+0,21)	0,3(0,26+0,34)	0,44(0,34+0,54)	0,2(0,14-0,26)
Сахар крови mg%	104,6(90,6+116,6)	78,2(58,0+98,4)	127,1 <sup>M</sup> (102,1+152,1)	129,5 <sup>M</sup> (120,1+138,5)	108,7(90,7+126,7)	96,3(72,6-120)
Молочная кислота mg%	32(24+40)	3(2,2+3,6)	0 <sup>M</sup>	22,5(18,5+26,5)	2 <sup>M</sup> (0,16+2,4)	38(30,8-45,2)
Пировиноградная к-та mg%	1,4(1,0+1,8)	1,28(1,06+1,4)	1,15 <sup>M</sup> (0,75+1,55)	1,72(1,51+1,93)	1,08 <sup>M</sup> (0,97+1,63)	1,98(1,8-2,16)
$\beta$ -липопротеиды mg%	26(20,2+31,8)	28(13+43)	14 <sup>M</sup> (11,9+16,1)	16 <sup>M</sup> (10+22)	20(16+24)	38(25+51)
Общий холестерин mg%	44(46+76)	65(47+83)	56 <sup>M</sup> (39+73)	55 <sup>M</sup> (41+65)	82(73+91)	85(74+96)
Общие жиры mg%	568(539+597)	605(532+678)	375 <sup>M</sup> (309+457)	476 <sup>M</sup> (418+534)	451 <sup>M</sup> (378+524)	605(532+678)
Холинэстеразная активность КРД (мг/мл)	24(19+29)	32(19+45)	11 <sup>M</sup> (6+16)	8 <sup>M</sup> (2+14)	7 <sup>M</sup> (2+13)	34(29+39)
Активность молочной дегидратазы mg%	1,01(0,96+1,06)	0,98(0,84+1,16)	0,85(0,75+0,95)	1,48(1,3+1,66)	1,46(1,3+2,66)	1,0(0,82+1,18)

<sup>M</sup> - НЕДОСТОВЕРНЫЕ РАЗЛИЧИЯ В СРАВНИВАЕМЫХ ГРУППАХ С СОЧЕТАЮЩИМ ВОЗДЕЙСТВИЕМ И ИЗОЛИРОВАННЫМ ОТ УДВОЕННОЙ ДОЗЫ ВНЕШНЕГО  $\gamma$  - ОБЛУЧЕНИЯ И АМЕРИЦИЯ-241.

Характер воздействия		Средняя продолжительность жизни крыс, дни (P=0,05)
Внешнее гамма-облучение,	Америций-241,	
1200	-	5 (4+8)
-	0,2	12 (9+15)
600	-	11 (7+18)
600	0,1	11 (8+12)
-	0,1	40 (37+43)
300	-	485 (385+610)
300	0,05	57 (44+56)
-	0,05	176 (138+225)
150	-	554 (373+745)
150	0,025	256 (219+300)
-	0,025	288 (253+328)
Контроль		594 (453+675)

Таблица 2.

Таблица 3.

Изменение показателей обмена веществ на 7 суток после сочетанного и изолированного воздействия окиси трития и внешнего  $\gamma$ -облучения в подостроэффективных дозах.

Показатель	300 p $\gamma$ Co <sup>60</sup>	0,5 mCi/g НТО	300 p $\gamma$ Co <sup>60</sup> + 0,5 mCi/g НТО	1 mCi/g НТО	Контроль
Гликоген печени g%	3,99(3,19+3,87)	2,62(2,12+3,12)	1,53 <sup>m</sup> (1,37+2,60)	2,38 <sup>m</sup> (1,47+3,29)	4,96(4,71+5,21)
Пирровиноградная кислота mg%	0,66(0,53+0,79)	0,48(0,22+0,74)	0,31 <sup>m</sup> (0,18+0,44)	0,47 <sup>m</sup> (0,27+0,67)	1,4(0,7+2,1)
Щелочная фосфатаза mg%	1,4(1,27+1,53)	1,2(1,1+1,25)	0,89 <sup>m</sup> (0,7+1,08)	0,7 <sup>m</sup> (0,53+0,87)	1,4(1,3+1,5)

<sup>m</sup> — недостоверные различия в сравниваемых группах с сочетанным воздействием и изолированным от удвоенной дозы окиси трития.

Характер воздействия		Средняя продолжительность жизни крыс, дни (при P=0,05)
Внешнее гамма-излучение, R	Окись трития, mCi/g	
1200	-	5 (4 ± 5)
-	2,0	5 (5 ± 6)
600	1,0	5 (5 ± 6)
600	-	13 (11 ± 15)
-	1,0	21 (13 ± 32)
300	0,5	14 (12 ± 16)
300	-	375 (335+420)
-	0,5	80 (43 +121)
150	0,25	254 (216+296)
150	-	392 (308+498)
-	0,25	346 (262+457)
Контроль		575 (513+678)

Таблица 4.

ОСОБЕННОСТИ КОМБИНИРОВАННОГО ДЕЙСТВИЯ НА  
ОРГАНИЗМ РАДИОАКТИВНЫХ ВЕЩЕСТВ (ПОД-131,  
ПРОМЕТИЙ-147) И ВНЕШНЕГО ИЗЛУЧЕНИЯ В  
ЭКСПЕРИМЕНТЕ.

Л.И.Бурыкина, Р.Л.Орлянская, В.Л.Пономарева,  
Л.А.Васильева, Н.А.Веселовская, Ш.Жанадилов,  
Н.Н.Мельникова.

Институт гигиены труда и профзаболеваний АНН СССР  
Москва СССР.

Summary

The paper presents some data based upon experiments on rats. The authors studied the combined forms of radiation diseases caused by external ( $\gamma$ , X-rays) and internal ( $^{147}\text{Pm}$ ,  $^{131}\text{I}$ ) sources of radiation. A distinctive effect of the summed up diseases has been discovered in adult rats during the  $^{147}\text{Pm}$  and external radiation. The age of the animals has been found effective for the course of the combined radiation diseases (due to  $^{131}\text{I}$  and X-rays). The younger rats (36-42 days old) have proved to be more radio-sensitive as compared to the adult ones (2-2.5 months old). They have shown somewhat summed up effects of the radiotoxic action of  $^{131}\text{I}$  and external radiation as drawn by the obtained hematological and honodotropic criteria.

The authors have proved  $^{131}\text{I}$  to have a higher blastomogenic efficiency in chronic administration as compared to its short term one.

Введение.

Проблему комбинированных форм радиационных поражений, которые наиболее полно моделируют воздействие радиационной компоненты производственной среды на организм человека, следует отнести к весьма актуальным и недостаточно изученным проблемам современной радиационной медицины труда и радиотоксикологии. В настоящее время нет научно обоснованных предпосылок, позволяющих оценить ожидаемые результаты комбинированных радиационных воздействий, а именно, развитие независимого действия, суммирование эффекта, либо потенцирование его.

Литературные данные по изучаемому вопросу противоречивы. Ряд авторов<sup>1,2</sup> приходит к выводу о возможном проявлении антагонизма в совместном действии  $^{131}\text{I}$  и внешнего излучения, другие<sup>3,4</sup> указывают на аддитивность их действия. Сведения о комбинированном действии  $^{147}\text{Pm}$  и внешнего излучения в литературе отсутствуют. Целью настоящего исследования явилось изучение ближайших и отдаленных последствий комбинированных форм радиационных поражений, вызванных радиоактивными веществами с различным типом распределения ( $^{131}\text{I}$ ,  $^{147}\text{Pm}$ ) и внешними источниками радиации ( $\gamma$ , X) в

зависимости от дозы и ритма воздействия и от возраста подопытных животных.

## 1. Материал и методы исследования.

1.1. В эксперименте на 293 крысах изучали биологическую эффективность  $^{147}\text{Pm}$  (7,0 мкюри/г) в сочетании с общим внешним гамма-облучением в среднесмертельной дозе (740р), а также действие каждого фактора в отдельности (табл. 1).

Облучение крыс проводили на установке "Стебель" с мощностью дозы 617р/мин.  $^{147}\text{Pm}$  был введен внутрибрюшинно через 2 часа после облучения. Содержание  $^{147}\text{Pm}$  в биологических пробах определяли по методу С.Э.Иноя. Биологическое действие  $^{147}\text{Pm}$  и гамма-излучения оценивали по общеклиническим, гематологическим и гистологическим показателям. Использовали общепринятые методики.

Поглощенные дозы через месяц после введения  $^{147}\text{Pm}$  (7,0 мкюри/г) оказались равными в скелете  $2400 \pm 500$  рад, в печени  $4100 \pm 800$  рад, в селезенке  $700 \pm 150$  рад.

1.2. Оценка биологического действия  $^{131}\text{I}$  и внешнего рентгеновского излучения в зависимости от возраста животных проведена на крысах в возрасте 2-2,5 месяцев и 36-42 дней. Схема эксперимента и уровни радиационного воздействия приведены в таблице 2.  $^{131}\text{I}$  вводили крысам через рот в течение 10 дней. Животных облучали на аппарате РУМ-11 с мощностью дозы 38,8 р/мин. (сила тока 15мА, напряжение на трубке 180 кв., фильтры 1 мм Сс и 1мм А1, кожно-фокусное расстояние 35 см). Использованы те же методы исследования, что и в эксперименте 1.1.

Среднетканевая полная поглощенная доза в щитовидной железе при воздействии  $^{131}\text{I}$  у молодых крыс оказалась равной 6900 рад, у взрослых - около 11000 рад; при комбинированном воздействии внешнего и внутреннего облучения эти величины соответственно равнялись 6900 и 15000 рад (дозы определены с точностью  $\pm 20\%$ ).

1.3. Изучение действия малых доз  $^{131}\text{I}$  и гамма-излучения в хроническом эксперименте проведено на 548 крысах-самцах, разделенных на 8 групп. Животные трех групп ежедневно получали с питьевой водой различные дозы  $^{131}\text{I}$ , животные двух других групп подвергались комбинированному воздействию  $^{131}\text{I}$  и внешнего гамма-излучения, крысы двух других групп - только внешнему облучению. Схема эксперимента и уровни радиационного воздействия указаны в таблице 3.

## 2. Результаты исследований.

### 2.1. Комбинированное действие $^{147}\text{Pm}$ и внешнего гамма-излучения.

Условия проведения эксперимента и показатели смертности крыс различных групп представлены в таблице 1. Введение переносимой дозы  $^{147}\text{Pm}$  (7,0 мкюри/г) на фоне предшествующего внешнего общего облучения в среднесмертельной дозе (740р) привело к повышению смертности крыс примерно в 2 раза (92,3% против 50%). Гематологические данные также свидетельствуют о суммации повреждений при воздействии двух указанных факторов (рис. 1).

Гистологические исследования, проведенные в разгар лучевой болезни (10-е сутки) и в период восстановления (30-е сутки), подтвердили значительное усиление эффекта при воздействии внешнего и внутреннего облучения по сравнению с изолированным воздействием.

Структурные изменения в селезенке развивались по типу острого лучевого поражения. К 30 дню отмечено некоторое увеличение числа лимфоцитов, появление очагов эктопического кроветворения и



избыточного количества мегакариоцитов. У выживших после внешнего облучения крыс отмечено активное восстановление лимфопоэза. В селезенке крыс, которым был введен  $^{147}\text{Pm}$  (1 группа), в этот период и ранее обнаружено некоторое уменьшение размеров фолликулов и снижение числа малых лимфоцитов, слабая фиброадения.

Следовательно, проведенное нами исследование позволило установить суммацию действия двух факторов радиационной природы при их совместном применении.

## 2.2. Ближайшие и отдаленные последствия комбинированного действия $^{131}\text{J}$ и общего рентгеновского облучения в зависимости от возраста животных.

Показатели смертности взрослых крыс различных групп в течение 21 месяца от начала опыта представлены в таблице 2. Выявлено более быстрое вымирание крыс, в течение года, подвергавшихся комбинированному воздействию, по сравнению только с внешним облучением (19,2% и 7,6% соответственно). Смертность в указанных группах за 21 месяц после воздействия оказалась близкой (91 и 96%) и значительно превышала таковую значения в контроле (45,4%). Гибель крыс, подвергшихся воздействию  $^{131}\text{J}$ , за 21 месяц наблюдения была существенно выше (68%), чем в контроле.

Гематологическое исследование показало, что при всех видах воздействия (особенно при их комбинации) поражение гемопоэза у крыс было более глубоким, чем у половозрелых крыс (рис. 2). Комбинация двух факторов не вызвала у взрослых животных никакого изменения по сравнению с тем, что наблюдалось при одном внешнем облучении. У крыс при комбинированном воздействии  $^{131}\text{J}$  и внешнего излучения имело место утяжеление эффекта - поражение крови было более глубоким, восстановление начиналось позднее и шло медленнее (особенно красной крови).

Через 6 месяцев после радиационного воздействия обнаружено отчетливое нарушение генеративной функции самцов, подвергшихся комбинированному воздействию. Более глубокое угнетение функции воспроизводства наблюдалось у молодых животных, облученных в возрасте 36-42 дней. При скрещивании процент беременных самок составил  $37,0 \pm 3,1$  (при  $74,0 \pm 8,4$  в контрольной группе). В группе взрослых животных эти величины были соответственно равны  $53,0 \pm 8,1$  и  $75,0 \pm 5,6$ %.

При изолированном воздействии  $^{131}\text{J}$  или одном внешнем облучении крыс-самцов понижения скрещиваемости их с интактными самками не установлено.

У подопытных самок способность к зачатию нарушалась после всех видов воздействия. Если количество бесплодных самок (в возрасте 6-8 месяцев) в контрольных группах составляло 43-47%, то в опытных группах оно возрастало до 68-80%.

У самцов, подвергнутых внешнему облучению и комбинированному воздействию, через 30 дней гистологически выявлено угнетение сперматогенеза, запустевание канальцев, появление многоядерных клеток, увеличение количества интерстициальной ткани. У взрослых крыс снижение веса семенников было более значительным, чем у молодых.

В тот же срок не найдено структурных изменений в гонадах крыс, получавших  $^{131}\text{J}$ .

На 19-й день после внешнего облучения отмечалось увеличение высоты эпителия щитовидной железы, усиление его пролиферации, появление зоны пристеночной вакуолизации коллоида. Эти признаки свидетельствовали об усилении синтеза и выброса гормонов. В последующие сроки отмечалась нормализация строения железы.

При введении  $^{131}\text{J}$  наряду с атрофией фолликулов отмечалось усиление роста паренхимы, увеличение высоты эпителия. Наблюдался мезфолликулярный и периваскулярный склероз.

При комбинированном воздействии строение щитовидной железы было близким к таковому при воздействии  $^{131}\text{I}$ , но атрофия фолликулов и рост паренхимы были выражены менее значительно, чем под влиянием  $^{131}\text{I}$  и внешнего излучения отдельно. При этом виде воздействия также отмечен значительный склероз.

В отдаленный период при всех видах радиационного воздействия частота развития опухолей щитовидной железы была близкой и хотя превышала контрольные значения, но не отличалась достоверно от них (табл. 4). При этом установлено, что при внешнем облучении развиваются только доброкачественные опухоли, а при введении  $^{131}\text{I}$  и при сочетанном воздействии наряду с аденомами отмечаются и раки.

### 3.2. Бластомогенное действие $^{131}\text{I}$ и гамма- и рентгеновского излучения на щитовидную железу.

При хроническом воздействии малых доз  $^{131}\text{I}$  и общем гамма-облучении утяжеления опухолевого эффекта по сравнению с изолированным применением  $^{131}\text{I}$  в тех же дозах не выявлено (табл. 4).

Для опухолей, возникающих под влиянием  $^{131}\text{I}$  и при комбинированном воздействии, была характерна множественность и разнообразие гистологического строения. Наиболее часто встречались аденомы папиллярного строения. Число их у одного животного могло достигать 4-5. Они часто сочетались с опухолями из клеток Ашнмаса. Выявлено также сочетание различных форм рака.

Наши данные расширяют сведения, полученные другими авторами, о бластомогенном действии на щитовидную железу  $^{131}\text{I}$  (5-3).

Важен в практическом отношении вывод о том, что хроническое облучение щитовидной железы  $^{131}\text{I}$  при сравнимых уровнях поглощенных доз оказывало большее бластомогенное действие, чем 10-дневное его поступление: суммарная частота развития опухолей щитовидной железы 44,4 и 15,3%, злокачественных - 20,3 и 4,6% соответственно.

### Заключение.

Проведенное изучение комбинированных форм радиационных поражений позволяет высказать суждение о том, что стягивающее действие радиоактивного изотопа по непосредственным эффектам определяется тем вкладом, который он вносит в повреждение, кроветворения - критической системы организма, ответственной за острую лучевую гибель. В отдаленные сроки риск бластомогенного действия малых доз  $^{131}\text{I}$  при длительном его введении возрастает. Эффект суммации развития опухолей щитовидной железы при комбинированном облучении в наших условиях опыта не обнаружен.

### Литература.

1. Василенко И.Я., Классовский Л.А., Малахов А.Э. В сб. "Радиационная эндокринология", Обнинск, 1967, стр. 16.
2. Москалев Ю.И., Егорова Г.И., Мишкин В.Ф. В сб. "Распределение, кинетика обмена и биологическое действие радиоактивных изотопов йода". Под ред. Л.А.Ильина, Ю.И.Москалева. М., 1970, стр.173
3. Голиков В.В., Наркличская О.Н., Классовский Л.А., Лобкова Н.П. "Мед. радиол.", 1969, 14, 10, 42.
4. Назаргина А.Г., Балановская Л.М. В сб. "Распределение, кинетика обмена и биологическое действие радиоактивных изотопов йода". Под ред. Л.А.Ильина, Ю.И.Москалева, М., 1970, стр. 209.
5. Пинель С.Э. "Бюлл. exper. биол. и мед.", 1955, 4, 76.
6. Lindsay S., Potter G., Chaikoff J.L. "Cancer Res", 1957, 173, 189-189.
7. Doniach S, "Health Phys", 1963, 9, 12, 1357-1362.
8. Potter G., Lindsay S. Chaikoff J.L. "Arch. Pathol.", 1960, 69, 3, 257-269.
9. Marks S, Bustad L.K. "J. Nat. Cancer. Inst.", 1963, 30, 661.

Таблица 1.

Схема эксперимента и характеристика гибели крыс в течение 30-ти дней после воздействия  $^{146}\text{Pm}$  и  $\gamma$ -лучами.

Группа	Радиационное воздействие		Количество во крыс в группе	Смертность	
	фактор	уровни		абсол.	%
		р	мкюри/г		
I	$^{147}\text{Pm}$	-	70	51	0
II	$^{147}\text{Pm}$	-	24,0	15	88,7
III	$\gamma$ -лучи	740	-	80	50,0
IV	$\gamma$ -лучи	825	-	52	96,1
V	$\gamma$ -лучи+ $^{147}\text{Pm}$	740	7,0	65	92,3
VI	Биол. контроль	-	-	30	0

Таблица 2.

Схема эксперимента и характеристика гибели крыс после воздействия  $^{131}\text{I}$  и  $x$ -лучами (подострое воздействие)

Группа	Вид и уровень воздействия	Число крыс в группе	Смертность % за периоды (месяцы)					t
			1	6	12	18	21	
Ia	$x$ -лучи (3000)	125	0,8	7,8	7,3	61,7	91,1	5,7
IIa	$^{131}\text{I}$ (0,1 мкюри/г за 10 дней)	120	0	0	5,4	32,0	68,0	2,1
IIIa	$x$ -лучи+ $^{131}\text{I}$ (в тех же дозах)	123	6	6	19,2	58,3	95,8	5,4
IVa	Биол. контроль	120	0	0	10,8	30,3	45,4	-

Таблица 3.

Схема эксперимента с хроническим воздействием  $^{131}\text{I}$  и  $\gamma$ -лучами.

Группа	Число крыс	Вид воздействия	Суммарная эквивалентная доза за 6 месяцев, р	Общее кол-во $^{131}\text{I}$ , введенное за 6 месяцев, мкюри	Средняя поглощенная доза в щитовидной железе, рад.
IIб	65	$^{131}\text{I}$	-	12,8	3000
IIIб	63	$\gamma$ -лучи+ $^{131}\text{I}$	100	12,8	2700
IVб	70	$\gamma$ -лучи	50	-	50
Vб	70	$^{131}\text{I}$	-	5,4	1800
VIб	63	$\gamma$ -лучи+ $^{131}\text{I}$	50	5,4	1700
VIIб	70	$^{131}\text{I}$	-	1,6	700
VIIIб	64	Биол. контроль	-	-	-

Таблица 4.

Частота развития опухолей щитовидной железы в подостром и хроническом эксперименте.

Группа	Средняя поглощенная доза в щитовидной железе, рад ( $\bar{x} \pm 20\%$ )	Число крыс, вскрытых со времени развития первой опухоли	Число крыс	
			с опухолями	с раком
1а	300	40	7 (17,6%)	-
Па	11000	43	7 (16,3%)	2 (4,6%)
IIa	15000	36	6 (16,6%)	2 (5,3%)
1б, 1У5	50-100	44	3 (6,8%)	1 (2,2%)
IIб, У6	1800-3000	54	24 (44,4%)	11 (20,3%)
IIIб, У16	1700-2700	44	18 (40,9%)	7 (15,9%)
УП6	700	27	4 (14,8%)	2 (7,4%)
1Уа, 1.16	-	32	6 (7,3%)	2 (2,4%)

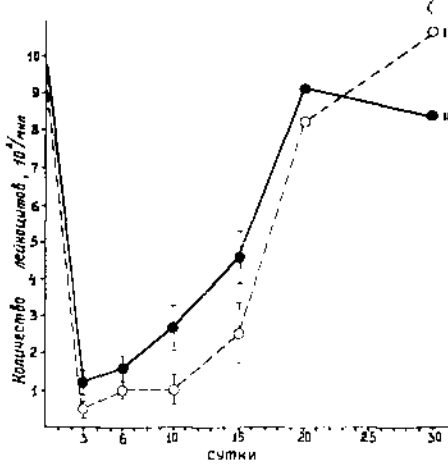
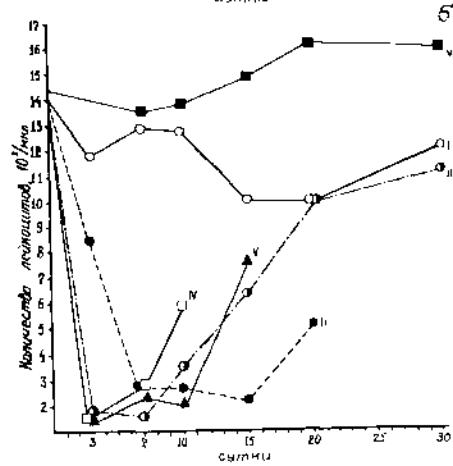
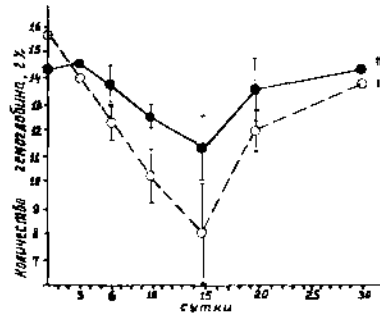
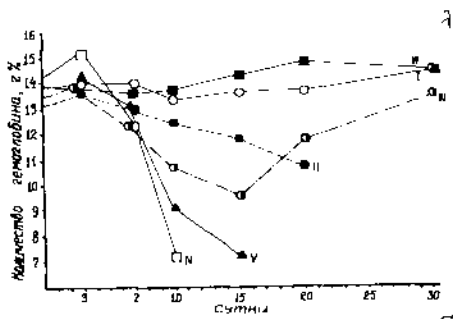


Рис. 1. Изменение количества гемоглобина (а) и лейкоцитов (б) у контрольных и подопытных крыс: I- $^{147}\text{Pm}$  (7мкюри/г); II- $^{147}\text{Pm}$  (24мкюри/г); III- $\gamma$ -лучи (740р); IV- $\gamma$ -лучи (826р); V- $^{147}\text{Pm}$  (7мкюри/г +  $\gamma$ -лучи (740р); VI-биологический контроль.

Рис. 2. Изменения количества гемоглобина (а) и лейкоцитов (б) у молодых (I) и взрослых (II) крыс при комбинированном воздействии ( $^{131}\text{I}$  + X-лучи).

QUANTIFICATION OF POST-IRRADIATION LEUKOCYTE DATA  
RELATIVE TO ESTIMATING EFFECTIVE EXPOSURE\*

George M. Angleton, Ph.D.  
Colorado State University, Fort Collins, Colorado 80521

Charles D. Robinette, Ph.D.  
National Academy of Sciences, Washington, D.C. 20418

Peter B. Carter, M.D.\*\*

Louis F. Wailly, Ph.D.\*\*  
\*\*United States Air Force Academy, Colorado

Abstract: The exponential model  $R = \alpha \cdot \text{Exp}(\beta \cdot E)$  was used in a study of the post-irradiation response pattern of leukocytes in young male beagles given whole-body subacute bilateral exposures to Co-60 radiations. The post-irradiation sensitivity of the minimum relative leukocyte count as characterized by the rate parameter  $\beta$  was estimated to be -0.7% per R. Conversely, a maximum post-irradiation depression of 25% could be used to infer that an individual had been subjected to an effective equivalent exposure of 40 R.

Introduction

The general characteristics of the time-dependent post-irradiation response patterns of the various components of the mammalian hemopoietic system have been known for many years. When the irradiation treatment consists of a whole-body, single, sub-lethal exposure given over a period of time not exceeding twenty minutes, many of the hemopoietic components exhibit a similarity in their response patterns. With respect to time post-exposure, the concentrations of these components initially show a decrease, they reach a minimum and eventually increase back to a level commensurate with the homeostasis requirements of the individual subject. The magnitudes of the rates with which these processes occur and the magnitude of the maximum depression are observed to be dose dependent.

One of these components which has been studied rather extensively is the group of cells collectively referred

to as the leukocytes.<sup>2,3,4</sup> The time-dependent post-irradiation pattern of the leukocyte response for sub-lethal exposures is such that a minimum count is manifested about ten to fifteen days post-exposure, the level of which is proportional to the dose received.

The mere existence of such a general pattern does not justify the use of an inverse procedure for determining an effective dose when the level of exposure is not known. The reasons for this are twofold. The first reason is related to very large variations in response patterns both among individuals and within individuals on a day to day basis. The second reason is that only minimal effort has been put forth relative to quantification of this dose-response relationship. The principal requirement is that methods of analysis must be developed which will permit the making of inferences with greater certainty by simultaneously taking into account sources of variations as related to several variables.

With such problems in mind, the purpose or goal of this paper is to

\*This work supported in part by Contract AF29 (601)-6782, Kirtland Air Base, New Mexico and Contract FDA 72-302 Food and Drug Administration.

explore the use of some rather simple and direct biometrical methodologies to be used in the making of inferences about the leukocyte response patterns of irradiated individuals.

#### Biometrical Considerations

The general method of analyses utilized here is to first separately characterize the time dependent response patterns of the individuals in terms of single parameters or parameter sets, then to study the dependence of these parameter sets on level of treatment.

In the case of leukocyte depression following irradiation one way by which this goal may be partially achieved is to obtain an estimate of the magnitude of the minimum level of response, normalize it with regard to a pre-irradiation level and then to adjust this level relative to a control population in order to allow for seasonal or time dependent variability.

The minimum level of response sought is referred to as an effective minimum. Obtaining this estimate requires the use of a smoothing technique. This may be accomplished by fitting a polynomial to the data in the regions where the minima occur. In general, if data are taken on a daily basis the fourth-degree polynomial is a desirable equation or model to use. Namely, it allows for the data to exhibit considerable non-symmetry of the response pattern in the vicinity of the minimum response and the adjacent points of inflection.

In a similar manner estimates of average pre-irradiation responses may also be obtained. The ratio of the minimum response level to the pre-treatment level becomes a relative or normalized response. The importance of the use of this procedure is that individuals with low pre-treatment counts tend to have proportionately low post-treatment counts. Once such estimates are obtained, their dependence on level of treatment and other variables may be studied. Previous studies show that the dependency of these estimates can be expected to decrease with dose but at a decreasing rate. The maximum level of depression need not be zero.

Other relevant factors which might

influence the level of response of the end-point under considerations include sex, age and weight. It appears reasonable to assume that the dependence of the response (R) on exposure (E) might be approximated by a decreasing exponential fraction with either a zero or non-zero asymptote. The dependence of R on age (A) and weight (W) might be expected to be somewhat linear over the restricted ranges of these variables that one usually incurs. The dependence of R on sex (S) can be treated in a similar manner giving 0, 1 values to S for males and females respectively. The first order approximation for the dependence of R on E, A, W, and S is expressed as follows:

$$R = \alpha_0 + \alpha_1 \cdot \text{Exp}(\beta \cdot E) \alpha_3 W + \alpha_4 A + \alpha_5 S.$$

Analysis of data using this type of model can be performed using conventional techniques of analysis<sup>5</sup>. The estimate of the parameter  $\beta$  becomes the estimate of the expected relative rate of deviation per unit of exposure.

#### Procedures

The plausibility of using the previous model to describe the dose-response relationship of leukocyte depression following irradiation was studied using data obtained from an investigation involving forty young adult male beagles<sup>6</sup>. Whole body bilateral irradiations were performed using a Co-60 source. The midline dose rate was 5 R per minute. Four levels of exposure were used in this study: 0-R, 100-R, 200-R and 325-R. Eight beagles were originally assigned to the 0-R group and sixteen each to both the 100-R and 200-R groups. Four of the original 0-R group beagles were subsequently used for the 325-R exposures. Blood samples were drawn at a regular time early in the morning. The cephalic vein was used for obtaining the samples. Standard cellular and differential data were then obtained. The current analysis is based only on the early leukocyte data.

#### Results

Representative data for each of the four exposure groups are plotted on the graphs of Figure 1. They represent five animals each for the 0-R, 100-R, and 200-R groups and four animals for the 325-R group. One of the 325-R

animals died 20 days post-exposure.

In regard to the O-R data a rather wide variation in the data exists, a condition which was also characteristic of the pre-irradiation data for all four exposure groups. The post-irradiation data for the other exposure groups exhibit much smaller variance, these variances appear to decrease somewhat with increased exposure. This general pattern of decreased variance following exposure is also seen in the data for animals when they are examined individually. The solid lines on the graphs are plots of fourth-degree polynomials fitted to the data. General trends in the data can be discussed relative to these curves.

Two types of trends are present. The first is a time dependent decrease in the average leukocyte count for the O-R group, a decrease which must be taken into account in the overall assay of the irradiation effects. The second type of trend is the dose dependent reduction in leukocyte count. This depression is seen to be significant for all three exposure groups and to occur ten to fifteen days post-exposure.

As indicated, the plotting of the combined data is useful for trend analysis. However, as also indicated previously, separate analyses of the data for the individuals are required if dose-response curves are to be generated which are to show variances attributed to individual differences. Accordingly, the data for each animal were analysed using a fourth-degree or less polynomial. Only the data in the vicinity of the exposure for the minimum of the response were used to obtain the estimates of the minimum levels of response.

The fine structures of these response patterns for the individuals actually reveal three types of minima. Frequently, one sees a large transient drop in the leukocyte count one to two days post-exposure. However, it is not a consistent observation for all animals. Subsequently a second minimum occurs, a consistent event for all subjects. This event may take place anywhere from three to fifteen days post-exposure. The trend is for it to occur later for the higher exposures. However, it is not a very

reliable statistic when used alone. Coupled with other information it may contain some useful information. Finally, in the time range of 14 to 30 days post-exposure a third minimum is frequently detected. Again, though, this is not observed with a high degree of consistency. Accordingly, only the magnitudes of the response in the vicinity of the time range around the second minimum were studied. The same method of analysis was applied to the individuals of the O-R group in the ten to fifteen day post-irradiation time range.

The estimates of the absolute counts for the minimum response levels are plotted in Figure 2. The corresponding normalized data are plotted in Figure 3.

The correlations between the magnitude of these minimum response levels and the ages of exposure or the total body weights at the time of exposure were not found to be statistically significant. Accordingly, these variables were not used in the analyses discussed in this paper. However, the estimated correlations were not zero and the use of these variables may be justified in subsequent analyses of a more refined nature. The linear term for sex was deleted since only males were used in this study.

The resulting model after the deletion of the linear terms for weight, age, and sex contains only the exponential term and the constant term.

Subsequent evaluation of the model showed that the constant terms did not make a significant contribution to the description of the dose-response relationship and hence it was also deleted from the model. The model thus became a one term nonlinear exponential equation;  $R = \alpha \cdot \text{Exp}(\beta \cdot E)$ .

A logarithmic transformation of the model was made to facilitate analyses. Least squares estimates of the parameters under the transformation were obtained and the resulting equations plotted, the solid lines of Figures 2 and 3. The estimates of the parameter so obtained are:

Count	$\hat{\alpha}$	$\hat{\beta}$
Absolute	12.65	-0.00774
Normalized	0.97	-0.00722

The parameter of interest is  $\beta$ . The estimate of  $\beta$  for the two cases do not differ appreciably. However, since the normalized procedure leads to a decreased variance and also to less overlapping of the response patterns at the different treatment levels, the corresponding estimates of  $\beta$  would appear to be the more reliable estimate. In terms of this estimate, the predicted post-irradiation minimum response level of a subject would be expected to decrease exponentially with a rate constant of  $-0.007 R^{-1}$ .

#### Summary

Quantification of the post-irradiation leukocyte data for beagles was performed in terms of the dependence of the estimated minimum leukocyte count on levels of exposure. Polynomial analyses were used to obtain the estimates of the minimum level of response for each individual. These estimates were normalized with respect to averaged pre-exposure levels. The exponential equation  $R = \alpha * \text{Exp}(\beta * E)$  was fitted to these data. The resulting estimate of  $\beta$  was such that the minimum leukocyte count level could be expected to decrease exponentially with a rate constant of 0.7% per R of exposure.

The dose-response curve so obtained may be used for a calibration curve for use in the inverse process of estimating an equivalent effective exposure when the actual exposure is not known. Thus for a subject showing a maximum post-irradiation depression in the leukocyte count of 50%, the estimated effective exposure would be 96 R. The reliability of this estimate might not appear to be very high in lieu of the observed scatter of the data points from which the calibration curves were generated. However, if one makes a non-statistical value judgement, one perceives that the recovery characteristics of those subjects whose data points fall below the calibration curve are poorer than for those whose data points fall above the curve. That is, the former appear to have received a greater effective exposure than the latter, a condition which leads to having greater reliability associated with the estimated dose.

Finally, the material presented here represents the results of applying some rather basic biometrical techniques to a type of biological problem which is of general importance. Additional work needs to be done and is being done in an attempt to derive alternate methods of analysis using increasingly meaningful mathematical models.

#### References

1. L.O. Jacobson, The Hematologic Effects of Ionizing Radiation. In Radiation Biology, Vol I: High Energy Radiation (A. Hollender, ed.) McGraw-Hill Book Co., Inc. (1954).
2. E.J. Ainsworth and F.A. Mitchell, Postirradiation Leukocyte Patterns in Mice and Dogs Treated With Endotoxin. Rad. Res. 33, 325-336 (1968).
3. S.J. Baum and D.E. Wyant, Hematopoietic Recovery in Irradiated Dogs. AFRRI SR70-2 (1970).
4. D. Sturrouch, E.R. Balliner, and J.E. Traynor, Daily Total-Body Exposures of Primates to Proton-, X-, or Gamma Radiation, SAM-TR-70-1 (1970).
5. N.R. Draper and H. Smith, Applied Regression Analysis. John Wiley and Sons, Inc., New York (1966).
6. G.M. Angleton, Radiation Recovery Studies, AFWL-TR-66-161 (1967).



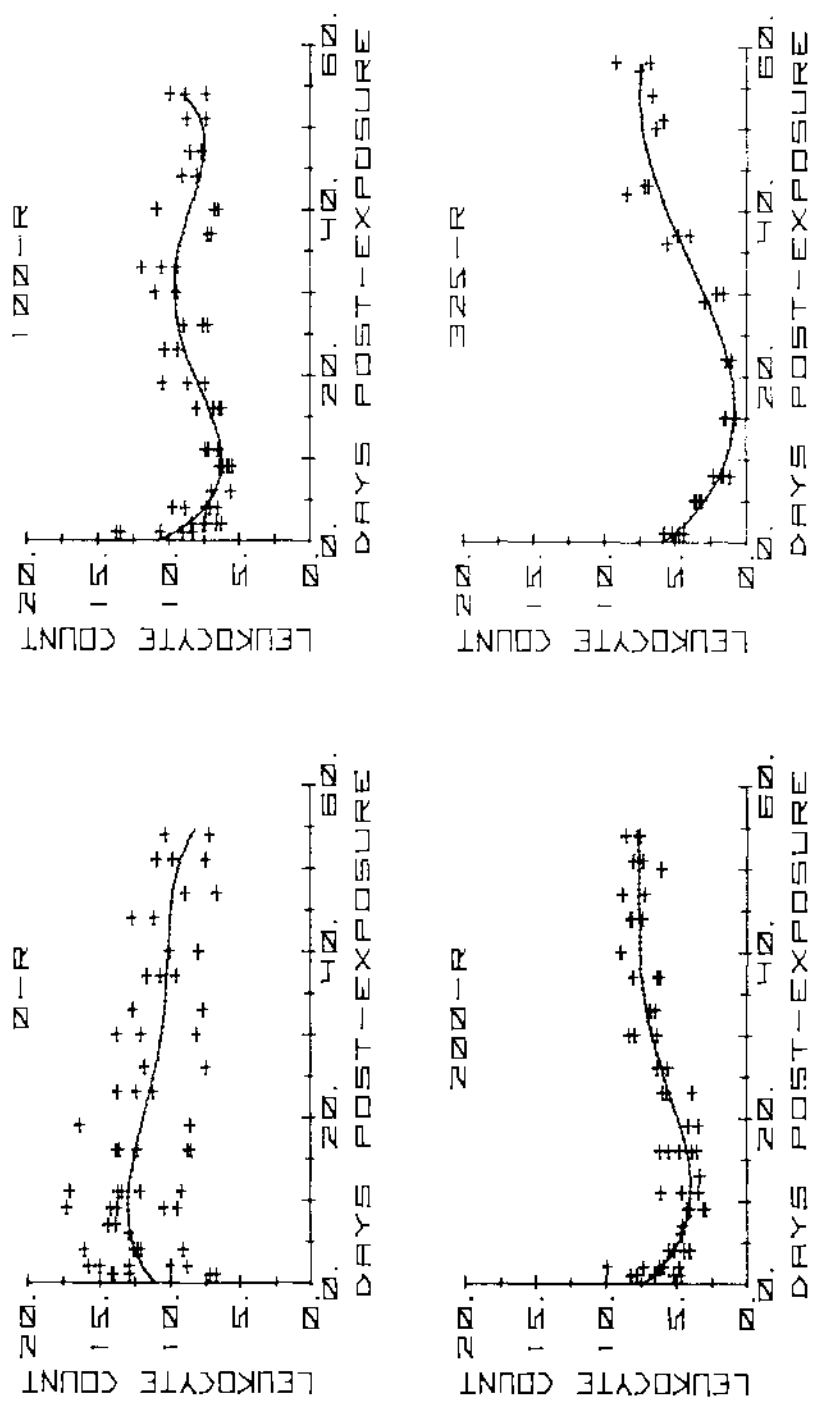
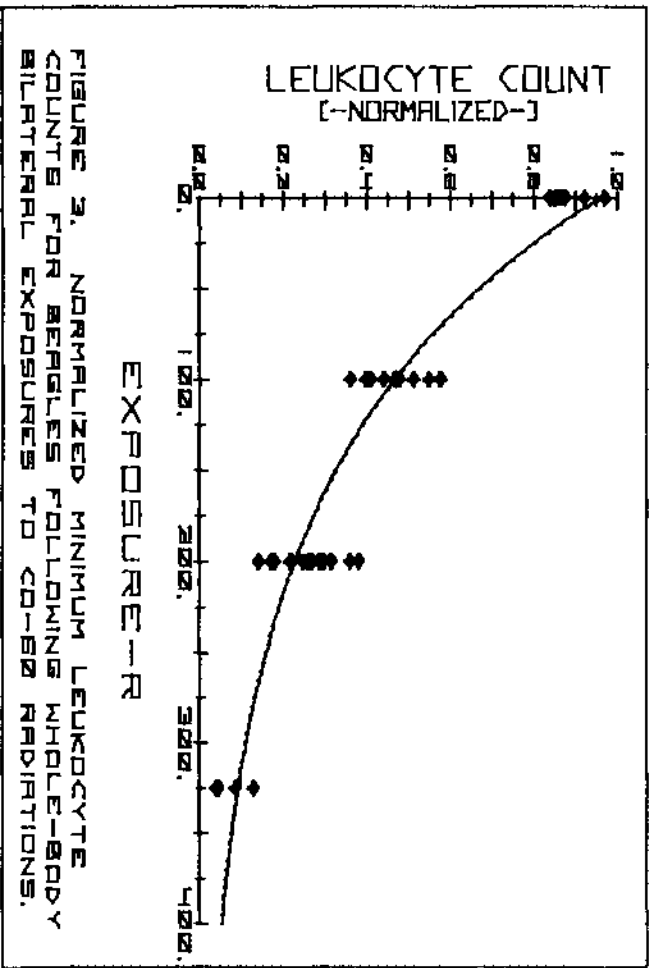
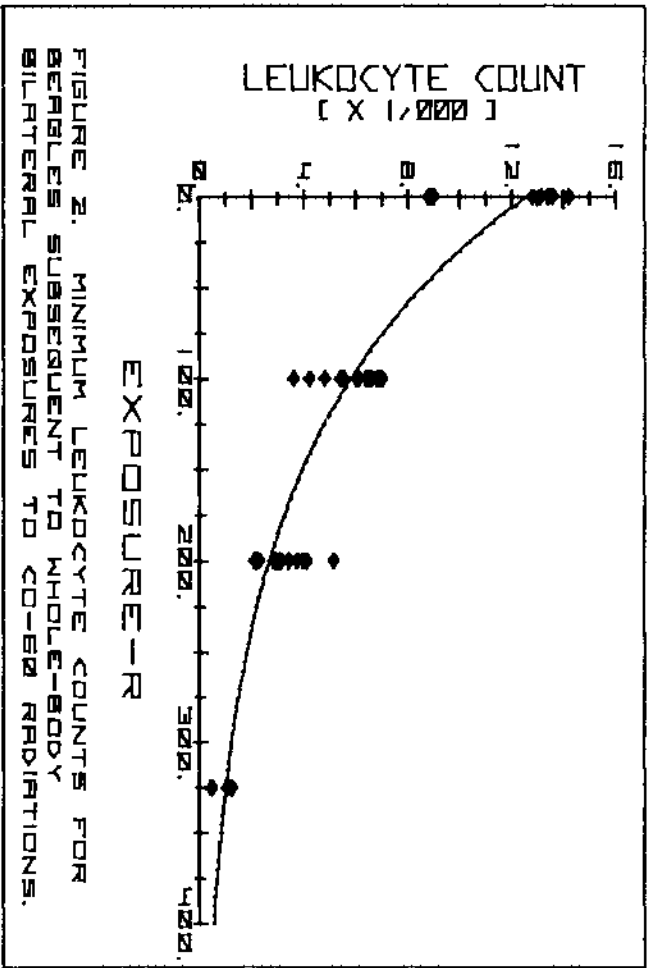


FIGURE 1. LEUKOCYTE COUNTS [X1,000] FOR BEAGLES FOLLOWING WHOLE-BODY BILATERAL EXPOSURES TO CO-60 RADIATIONS.



THIRD INTERNATIONAL CONGRESS OF THE  
INTERNATIONAL RADIATION PROTECTION ASSOCIATION  
Washington D.C., 9 - 14 Septembre 1973

-----

STUDY OF PURIC AND PYRIMIDIC COMPOUNDS IN URINE  
OF RABBITS AND RATS AFTER IRRADIATION

by

Ch. VALLE<sup>+</sup>, G. PEPIN, F. DUPREY, L. BREUIL<sup>++</sup>

+ Commissariat à l'Energie Atomique- France  
Département de Protection - S.P.T.E.  
B.P. n° 6 - 92260 - Fontenay aux Roses

++ Service de Santé des Armées - Centre de Re-  
ches - Rue Raoul Batany - 92140 Clamart

Abstract:

A chromatographic technic is described to study the urinary excretion of nucleic acid catabolit after irradiation. The results obtained for those metabolites that the chromatographic method had shown to change after irradiation are presented for animals uric acid and hippuric acid.

-----

The urinary excretion of nucleic acid catabolit after irradiation has already been studied by several authors, most of them being interested in basis and nucleosides only.

We think, we are the first to have undertaken a comprehensive study of the whole of puric and pyrimidic metabolites and their variations following irradiation.

A chromatographic technique has been developed to that purpose and the results obtained for those metabolites that method has shown to change after irradiation are presented for many animals

The substances retained by the resin are eluted by a gradient of different molarities (HCOOH - HCOONa)

Twenty two hours' chromatograms of standard nucleotides have allowed to determine the elution time of some twenty compounds.

Elution time	peak
0 to 45 minutes	puric and pyrimidic bases nucleosides and desoxynucleosides creatinine
50 minutes	d. CMP
55 minutes	C.M.P.
1 h. 20	NAD
3 hours	5' AMP - 5' d. AMP
3 h. 15	2' AMP
3 h. 40	3' AMP
4 h.	3' 5' AMP
5 h.	hippuric acid
8 h.	G M P
9 h.	N A D P
10 h.	U.M.P.
12 h. 30	ADP - U. DPG
14 h.	FMN
16 h.	GDP - UDP
17 h.	ATP
20 h.	UTP
21 h.	GTP

The contours of the chromatograms of biological liquids are less sharply outlined than standard chromatogram contours.

After undoubtable identification of the peaks by specific methods, it is possible to say that there are noticeable quantitative differences between normal and post exposure condition

in rats and rabbits delivered supra-lethal dosis of  $^{60}\text{Co}$  gamma rays.

Compounds identified	Variations after irradiation	
	24 h.	48 h.
Puric and Pyrimidic Bases, nucleosids and desoxynucleosids. Amino-Acids.	↑↑↑	↑↑
Uric acid	↑↑↑	↑↑
?	↓	
3'5' AMP ?	↓	↑↑
Hippuric acid	→ ○	→ ○

On the basis of these results a few metabolites varying after exposure have been studied.

Measurements have been made on a large number of animals with the respective methods.

Uric acid is changed by the specific uricase enzym into allantoin which will reduce  $\text{H}_2\text{O}_2$  in presence of an indicator which can be detected at  $440\text{ n.m.}$

Acid hippuric is determined by fluorescence in sulfuric acid at excitation wavelength of  $260\text{ n m}$ , and measurement wavelength  $370\text{ n m}$ .

As early as 1904 an increase of urinary uric acid has been noticed after exposure in irradiated patients, dogs, rabbits and rats.

Our results are in a good agreement and our measurements on some 30 rats (L D 100%/10 D and L D 20%/30 D) are summarized in tables 3 and 4

## L.D. 100% / 10 D

	Excretion uric acid mg/24 h.	$\sigma$	n
Normal	1.77	0.78	83
D + 1 post	2.48	0.05	5
D + 2 exposure	0.74	0.28	5
D + 3	1.24	1.07	5
D + 4	1.31	0.44	4
D + 5	0.95	0.47	4
D + 6	0.95	0.47	4
D + 7	3.22	2.41	3
D + 8	1.22	0.69	2

- Table 3 -

## L.D. 20% / 30 D

	Excretion uric acid mg/24 h.	$\sigma$	n
Normal	1.7	0,76	234
D + 1 )	2,29	1.1	9
D + 2 ) fast	1.48	1.032	9
D + 3 ) time	0.87	0.39	9
D + 1 post	3.37	1.57	9
D + 2 exposure	2.27	0.59	9
D + 3	1.9	0.8	9
D + 4	0.98	0.39	9
D + 5	1.67	0.65	8
D+6 - D+7	1.45	0.51	9
D + 8	2.41	1.6	9
D + 9	2.09	1.34	9
D + 10	1.78	0.6	7
D + 11	2.09	0.7	7

- Table 4 -

A increase of uric acid is noticed on the 1<sup>st</sup> day after exposure (about 50%) followed by a decrease on the second day down to a level much lower than normal, then there occurs an increase to the initial value for lot II, and a very high value for animals dying from irradiation, usually on the preceding death.

A significant decrease of hippuric acid is noticed after exposure (by a factor 5). (Tables 5 and 6). The decrease of Hippuric acid can be compared with the decrease induced by a complete fast (table 6) (the animals do not want food after exposure).

L.D. 100% / 20 D

	excretion Hippuric acid mg/24 h.	$\sigma$	n
Normal	19.81	4.59	59
D+1 after expo- sure	4.10	3.59	5
D + 2	6.43	8.6	5
D + 3	7.88	10.55	5
D + 4	12.23	9.24	5
D+5 - D+6	25.05	17	4
D + 7	34.13	28	3
D + 8	28	6	2

- Table 5 -

L.D. 20% / 30 D

	excretion Hippuric acid mg/24 h.	$\sigma$	n
Normal	10	3,3	130
Fast time			
D + 1	6,1	3	5
D + 2	2,1	1,7	5
D + 3	0,6	0	2
After exposure			
D + 1	6,3	2,7	5
D + 2	6,4	0,8	5
D + 3	5,5	2,2	5
D + 4	5,2	1,1	5
D + 5	9,1	2,6	5
D+6 - D+7	9,6	4,4	4
D + 8	12,5	5,2	5
D + 9	9,9	3,4	4
D + 10	11,75	1,6	4
D + 11	12,8	5,2	3

- Table 6 -

However, excretion of Hippuric acid seems to be decreased by factor 2 in surviving rats, no secondary increase was observed in the fasting animals, as noticed in the animals dying after exposure.

The results on urinary excretion of Hippuric acid and Uric acid allow to conclude that, they are highly influenced by  $\gamma$  exposure.

Fasting plays in the same way as exposure for Hippuric acid (lower excretion) and in the opposite way for Uric acid (lower excretion in fasting rats increased excretion after exposure).

However these effects do not allow to predict the fate of the animals death or survivals.



STRUCTURAL ABERRATIONS IN BONE MARROW CELLS  
AFTER TRITIATED WATER ADMINISTRATION IN RATS

Irina Andreuță and N.T.Racoveanu  
Institute of Hygiene and Public Health, Bucharest, Romania

Abstract

Structural aberrations in bone marrow cells were used as a biological test of HTO irradiation.

Three groups of nine rats were used : two groups received a single intraperitoneal dose of 75 and 100  $\mu$ Ci/g HTO ; one group received daily 30  $\mu$ Ci/g HTO for 18 days and 5 animals receiving saline solution served as controls.

Structural aberrations incidence in loo cells for each animal was established using the MOORHEAD and LAM-PO-TANG techniques.

The structural aberrations were of chromatidian (gaps and breaks) and chromosomal type (acentric fragments and translocations).

A relationship between calculated bone marrow dose and structural aberrations incidence was established.

Rats receiving once 100  $\mu$ Ci/g HTO (20 rads) have 5.0 $\pm$ 2.0 p.c. structural aberrations and rats receiving 18 days 30  $\mu$ Ci/g HTO daily ( $\simeq$  600 rads cumulative dose) displayed 7.3 $\pm$ 3.2 p.c. structural aberrations. Control rats displayed only 0.6 $\pm$ 0.5 p.c. aberrations.

1. Introduction

The aim of this paper is to present the alterations in number and structure of bone marrow cells chromosomes, in rats, after unique and repeated administration of tritiated water (HTO).

Chromosomal aberrations were used as a biological dosimeter, due to the great sensitivity mentioned in the literature (BENDER)<sup>1</sup>.

2. Material and methods

32 female WISTAR rats were divided in 4 groups. Groups A and B received intraperitoneally 75 and 100  $\mu$ Ci/g body weight HTO. Group C received for 18 days intragastric administration of 30  $\mu$ Ci/g HTO. Group D has served as control and received saline solution. Groups A, B and C were of 9 animals and D of 5 animals.

Amersham tritiated water was used, the dilution to 20

µCi/ml being made with saline solution.

Three animals from groups A and B were killed at 24 hours, 3 and 7 days interval, after HTO administration. From group C three animals were killed at 24 hours, 7 and 14 days after the last intragastric administration of HTO.

A technique combining the methods of MOORHEAD<sup>2</sup> and LAN-PO-TANG<sup>3</sup> was used for emphasizing chromosomes. 100 metaphases were analysed for each animal, scoring the aneuploid mitosis and the various types of chromatidian and chromosomal aberrations.

The pattern of normal rat caryotype described by VRBA<sup>4</sup> and FITZGERALD<sup>5</sup> was used.

### 3. Results and discussions

DAWEY's work shown that RBE of tritium particles for structural aberrations is 1.2 by comparison with <sup>60</sup>Co gamma rays.

In our work we have attempted to assess the magnitude of the process of alteration in structure and number of chromosomes and the elimination with the time of altered cells, the selection of damaged cells in renewing cellular systems being well known (FABRIKANT)<sup>7</sup>.

We did not notice differences in aneuploid cells between irradiated animals and controls at any interval after irradiation. The maximum occurrence of aneuploid cells was 13.6 p.c. and the minimum 9.0 p.c., in controls the incidence was 11.2 p.c.

Table 1 - Numerical abnormalities

GROUPS		Time of study days-	No. animals	Total cells analyzed	Euploid cells %	Aneuploid cells %
Single Administration	A 100 µCi/g	1	3	300	87 +9.3	13 +9.3
		3	3	300	86.4+1.1	13.6+1.1
		7	3	300	87.4+2.3	12.6+2.3
Single Administration	B 75 µCi/g	1	3	300	91 +1.7	9 +1.7
		3	3	300	88 +3.4	12 +3.4
		7	3	300	90 +1.1	10 +1.1
Repeated Administration	C 30 µCi/9/day	1	3	300	90.7+2	9.3+2
		3	3	300	87.4+2.3	12.6+2.3
		14	3	300	90 +1.7	10 +1.7
CONTROL			5	500	88.8+3.6	11.2+3.6

Structural aberrations observed were chromatidian (gaps and chromatid breaks) and chromosomal type (acentric fragments and translocations). The gaps were not scored because these are considered by EVANS<sup>8</sup> repairable lesions.

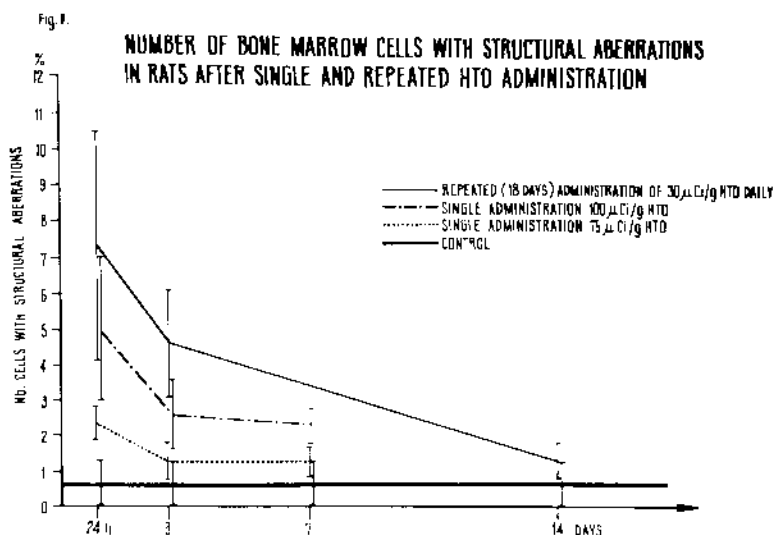
After single administration of HTO the incidence of chromatidian aberrations (table 2) is not significantly increased.

Table 2 - Structural chromosome aberrations

GRUPS		Time of study -days-	Nr. animals	Total cells analyzed	Nr. Chromatid Type Aberrations	Nr. Chromosome Type Aberrations		Cells with structural abnormalities	
					Breaks	Fragments	Translocations	Total	%
Single Administration	A 100 $\mu$ Ci/g	1	3	300	4	9	2	15	$5 \pm 2$ (p 0.025)
		3	3	300	3	3	2	8	$2.6 \pm 1$ (p 0.5)
		7	3	300	1	5	1	7	$2.3 \pm 0.5$ (p=0.02)
	B 75 $\mu$ Ci/g	1	3	300	2	5	-	7	$2.3 \pm 0.5$ (p=0.02)
		3	3	300	3	1	-	4	$1.3 \pm 0.5$ (p 0.5)
		7	3	300	4	-	-	4	$1.3 \pm 0.5$ (p 0.5)
Repeated Administration	C 30 $\mu$ Ci/g/day	1	3	300	7	9	6	22	$7.3 \pm 3.2$ (p=0.025)
		3	3	300	3	10	1	4	$4.6 \pm 1.5$ (p=0.02)
		14	3	300	-	3	1	4	$1.3 \pm 0.5$ (p 0.1)
D	CONTROL	-	5	500	3	-	-	3	$0.6 \pm 0.5$

The chromosomal aberrations increased significantly after 100  $\mu$ Ci/g HTO, and were gradually eliminated 3 and 7 days later. 75  $\mu$ Ci/g also produce a statistically significant increase after 24 hours and the decrease could be detected 3 days after HTO administration.

The repeated HTO administration produced a higher increase, 7.3 + 3.2 p.c. of cells with structural aberrations both the chromatidian and chromosomal type being involved. Three days after, cessation of HTO administration, the incidence of chromosomal aberrations is still significantly higher than in controls but 14 days later the elimination of damaged cells is almost complete.



From the calculation of bone marrow absorbed dose, we obtained for single administration of 100  $\mu$ Ci/g HTO approximately 27 rad, for 75  $\mu$ Ci/g - 20 rad and for repeated administration of 30  $\mu$ Ci/g the cumulative bone marrow absorbed dose was about 600 rad.

The pattern of structural aberrations after single administration of HTO, shown before, may be related with bone marrow cells irradiation, if we consider that the bone marrow receives an irradiation with a dose rate which attains the maximum a few hours after the HTO administration and decreases gradually during the first days. Such a distribution of irradiation could explain the reason for which 75  $\mu$ Ci/g HTO deliver enough bone marrow dose to produce statistical significant increase of structural aberrations only 24 hours after administration. With 100  $\mu$ Ci/g HTO, the dose obtained after administration produce enough altered cells to be detected until the 7 days.

During daily administration of HTO, the bone marrow dose increases gradually, the tritiated water eliminated in 24 hours being only a part of that administered the following day.

The equilibrium of tritium content in bone marrow with the tritium in blood reached, the dose attains a steady state until

the cessation of administration, afterwards the decrease begins. The production of damaged cells is at the same time partly compensated by elimination of these cells and this explains the figures observed in group C.

From the above discussions we have to conclude that tritiated water irradiation induces in bone marrow system with a cellular population inhomogeneous as structure and division phase, structural aberrations roughly dose dependent.

This fact needs a more accurate establishing on a homogeneous cellular system as peripheral lymphocyte.

#### References

1. H.A.Bender - Ann.of the New York Academy of Sciences, 1964, 114, 294.
2. P.S.Moorhead - Exp.cell.res., 1960, 20, 613.
3. P.R.L.G.Lam-Po-Fang. - Scand.J.Haemat, 1968, 5, 158.
4. M.Vrba - Folia biol.(Praha), 1964, 10, 75.
5. P.H.Fitzgerald - Exp.cell res., 1961, 25, 191.
6. W.C.Dewey - Radiat.res., 1970, 43, 561.
7. J.U.Fabrikant - Radiology, 1967, 88, 767.
8. H.J.Evans - Radiat.res., G.Silini, Ed. Amsterdam, 1967, North Holland Publ.Co.

MODIFICATION OF SOME BIOLOGICAL INDICATORS INDUCED BY SINGLE  
AND REPEATED ADMINISTRATION OF TRITIATED WATER

Valentin Stanculescu and Ariadna Ciubotaru-Bordeianu  
Institute of Hygiene and Public Health, Bucharest, Romania.

Abstract

The experiments were performed on white rats exposed to the action of tritium from tritiated water (HTO) by single (i.p.) and repeated (oral) administration. The results showed that in the lots which have received HTO, as compared to the controls, there was a lower percentage of iron incorporation in the red blood cells, and an early accumulation of  $^{59}\text{Fe}$  was noticed in bone marrow; however longer biological half time (Tb) indicate a prolonged stay of iron which situation has been also found in the spleen. The relative spleen weight was noticeably reduced in the animals which received larger quantities of HTO. The unsaturated iron binding capacity (UIBC) evinced a decreasing tendency, which shows that, after exposure to HTO, the degree of iron saturation of transferrin is higher.

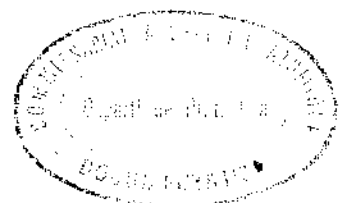
The comparison with the extent to which the same indicators are modified after whole-body X-ray exposure suggests that exposure to HTO causes a lesser effect than expected for the calculated dose.

Tritiated water has been the object of numerous studies, as well as its effects upon the organism, due to its particularities, as in its composition it has a low energy beta emitter which might be dangerous through the possibility of its creating local large energy absorption in the cell<sup>1,2</sup>. This possibility is enhanced by the fact that it has all properties of water, the biological medium in which metabolic reactions take place. Research workers have expressed various opinions with regard to the risk of HTO irradiation, and some authors have claimed to have evinced biochemical modifications even after very low exposures to HTO<sup>3</sup>. This position is backed by observation of Bond according to which after prolonged exposure to HTO, there appears tritium bound to organic compounds, which implied an additional irradiation hazard<sup>4</sup>.

The present authors have followed a series of biological indicators in single and repeated exposure to HTO and have attempted a comparison of the magnitude of the effects with those of X-irradiation.

Material and method

The experiments were performed on male white rats averaging 120 g. Before the experiments the rats were adapted to laboratory conditions for least one week.



Repeated exposure to HTO. Daily for 18 days running, the animals were given a quantity of 30  $\mu$ Ci HTO/body wt. through a gastric tube, the HTO having been diluted to the working activity with tap water. The necessary quantity for whole experiments was distributed in vials and sterilized through boiling. HTO Amersham was used. In the last day of administration the animals were i.p. injected with 3  $\mu$ Ci  $^{59}\text{Fe}$ -citrate Amersham with specific activity of the order of mCi/ $\mu$ g. The controls were administered tap water during the same interval.

Single administration of HTO was given to 3 lots of animals; the first lot received i.p. 75  $\mu$ Ci HTO/g body wt. in saline solution together with 3  $\mu$ Ci  $^{59}\text{Fe}$ ; the second lot was injected 150  $\mu$ Ci HTO/g body wt. and 3  $\mu$ Ci  $^{59}\text{Fe}$ ; and the controls received the same quantity of saline solution with  $^{59}\text{Fe}$ .

In the both types of experiments the animals were killed at 1, 3, 7 and 14 days after radioiron administration, in lots of 4-5 animals.

X-ray irradiation with 50 R was achieved as follow: 180 kVp, 5 mA, 0,5 mm Cu filter, distance 80 cm, dose-rate 6 R/min. Part of animals were injected with  $^{59}\text{Fe}$  immediately and killed at an interval of 1, 3, 7, 14 and 21 days, and the rest received  $^{59}\text{Fe}$  24 hours before the killing, which occurred at the same time interval.

Blood samples were taken on heparine, as well as samples from the following organs: spleen, liver and bone marrow (femur).

The following determinations were performed on the samples: percentage of red blood cell incorporation of iron (% RBC Fe-incorporation) was calculated by measuring radioactivity of the red blood cells from 1 ml blood after repeated washing with saline solution according to the usual formula.

$^{59}\text{Fe}$  distribution in organs was calculated by measuring the radioactivity of the fragment of the organ, and results were expressed in:

$$N = \frac{\text{counts/g wet organ}}{\text{injected counts/g body}}$$

relative weight of organs was expressed in the ratio organ wt./body wt. (in the case of the spleen this has been multiplied by 100 for the graphic representation).

the unsaturated iron binding capacity (UIBC) was performed after the technique described by Brozovich<sup>2</sup>.

## Results

After repeated administration of HTO the investigated indicators evinced the following variations:

Bone marrow incorporation of iron shows that in the first day a significantly increased quantity is retained in the lot exposed to HTO; however, in the groups killed at other time intervals no difference is found any longer. Calculating the biological half time ( $T_b$ ) of iron in the two lots, it is seen that disappearance of iron from the bone marrow occurs after an exponential law with two terms corresponding to two  $T_b$ 's as follow: for the control 0.8 and respectively 7.6 days, and for the lot exposed to HTO 0.6 and respectively 12.7 days. The second  $T_b$  of the exposed lot is by approximately 67% longer of the controls. (Fig. 1)

$^{59}\text{Fe}$  uptake in the spleen constantly show higher values in the HTO exposed animals but the differences are not statistically significant. The  $T_b$  calculated for the spleen show the existence of two  $T_b$ 's for iron elimination with the following valu-

es:control lot 2.1 and 14.5 days and exposed lot 1.7 and 23.0 days. As in the case of bone marrow, the second Tb of the exposed lot is by 65% longer than the corresponding time of the control.

In the liver the differences between the values of the two lots are not statistically significant, though there is a slight tendency of accumulation in the exposed lot.

On the first day after the period of administration was ended, the relative weight of the spleen in these animals was greatly diminished, amounting to only 55% of the value of the controls, which difference is statistically significant.

The percentage of red blood incorporation of iron is visibly affected, in the lot exposed to HTO: the utilization of iron in erythropoiesis is delayed, as may be seen from the first part of the curve, and the yield of iron utilization is diminished (the values of the exposed lots are constantly lower during the whole interval).

UIBC variations in the animals which received HTO ranges between the extreme limits of the controls. It may be seen in the first days the value of UIBC is below the control average, in the 7th day it rises to the superior limit of the control, after which it again approaches the average of the control lot.

The evolution of the same indicators, with the exception of bone marrow incorporation of iron, followed in animals which were given single administration of HTO i.o. may be seen in Fig. 2.

No significant difference is noticed in the spleen and liver between the control and exposed lots as regards the iron uptake.

The relative weight of the spleen is lower in the animals given 150  $\mu$ Ci HTO/g body wt. by 11-21%, whereas in the lots which received 75  $\mu$ Ci HTO/g body wt. it is higher in the animals killed after 3 and 7 days, as compared to the control.

% RBC Fe-incorporation is delayed and noticeably reduced in the lot which received 150  $\mu$ Ci HTO/g body wt., whereas the curve obtained in the animals given 75  $\mu$ Ci HTO/g body wt. is similar to that of the controls.

The UIBC values of animals injected with HTO vary within the limits of the values of the controls; still they seem to be influenced by quantity of HTO administered. The evolution of these values is divergent in the two exposed lots, the difference being statistically significant in the 7th and 14th days; there is however no significant difference between the exposed lots and the control.

The results obtained on the rats X-irradiated with 50 R show (Fig. 3) that the %RBC Fe-incorporation undergoes the same modifications as described above, a delayed and reduced incorporation in the irradiated lot, with a recovery in the 14th day. There are no modifications in the  $^{59}\text{Fe}$  uptake in the spleen and liver.

The distribution of iron in the organs was followed at the same intervals after the irradiation in animals injected radio-iron 24 hours before the killing, and it was seen that there is no difference between the exposed and the control lots as regards the %RBC Fe-incorporation; however there was noticed an obviously increased iron uptake in the bone marrow of the irradiated animals; the same tendency was less markedly present in the spleen, whereas in the liver the control values seemed to be higher.

### Discussion

The majority of indicators followed in this experiment belong to the metabolism of iron. Our interest in them is justified by



the various observations reported in literature which demonstrate the high radiosensitivity of the haemopoietic system.

Numerous reports show that, of the four main cellular sub-systems of the bone marrow, the erythropoietic line has the highest radiosensitivity<sup>6-9</sup>. It could be estimated that following the exposure of mice to LD<sub>50</sub> out of 1,000 stem cells only two or three survive which retain their proliferative capacity uninjured. Both in mice and in rats during the recovery period the contribution of the spleen to the process of erythropoiesis is considerably increased<sup>6,8</sup>.

Investigations using radioiron show that after irradiation the iron uptake in the bone marrow is increased in the first 24 hours as a result of disturbances in maturation and a delayed outflow of cells in circulation<sup>10</sup>. These effects are reperculated upon the percentage of red blood cell incorporation of iron. This indicator has been considered up to the present to be the most sensitive indicator in irradiation<sup>11,12</sup>, some authors claiming to identify exposures to 5 R<sup>13</sup>. After body irradiation a quantitative redistribution of iron at the level of various organs follows in general<sup>14,15</sup>. It has been found that following the action of any agent which alters the normal balance between the metabolic compartments of iron<sup>16</sup>, which action affects mainly the bone marrow, the iron tends to be bent to the storage organs<sup>17,18</sup>. The involvement of the transferrin in the general picture of the irradiation-induced modifications is obvious, as it appears with a higher degree of iron saturation<sup>18,19</sup>.

It emerges from our experiments that internal irradiation with tritium from HTO in quantities applied by us, determines noticeable modifications of the indicators we followed, similarly to the pattern described in literature. Thus, the curves showing the percentage of incorporation of iron in the blood cells are flattened in the first part, which modifications are the more important the greater the dose administered. In the lots which received HTO during 18 days and in the lot which received 150  $\mu$ Ci HTO/g body wt. the magnitude of the effects is comparable, whereas in the lot injected 75  $\mu$ Ci HTO/g body wt. they are similar to those of the controls.

The early accumulation of iron in the bone marrow and in the spleen and its longer T<sub>b</sub> in these organs is in agreement with above mentioned observations.

The behaviour of UIBC is different in the experiment with animals injected with a single dose of HTO depending on the administered dose, though it does not exceed the limits between which range the values of the control. Thus at the lowest dose, UIBC shows a tendency to increase with time, whereas a double quantity of HTO determines a decrease, suggesting that the transferrin is more heavily loaded with iron at higher doses.

Taking the T<sub>b</sub> of HTO to be 3.7 days<sup>20</sup> the following cumulated doses were calculated: in the repeated administration for the intervals of 1, 3, 7 and 14 days, 620, 720, 820 and 820 rad respectively and for the single administration of HTO in the amount of 75  $\mu$ Ci HTO/g body wt. the values were of 20, 50, 85 and 108 rad respectively; for the quantity of 150  $\mu$ Ci HTO/g body wt. the values were of 40, 100, 170 and 216 rad respectively.

In the lot irradiated with 50 R an alteration of the curve of  $\%RBC$  Fe-incorporation is seen, which however is small compared to those obtained from animals which were administered HTO. It may also be asserted with the difference due to the small number of animals in these lots that is a tendency to redistribute the iron

among the various organs which is less marked than in the case of the animals treated with HTO. Previous data<sup>19</sup> have demonstrated that there is a very strong decrease of UIBC in rats irradiated with doses greater than 200 R.

Comparing the modification of the followed indicators after irradiation with HTO and after  $\gamma$ -irradiation, it results that the dose of exposure to tritium in our experiment is located between 50 and 200 R, which is a by far inferior value to calculate dose. Evidently in order to make a firm statement in this respect the influence should be determined of some factors such as the character of the irradiation (internal or external), the dose-rate, the degree of reliability on the indicator's adequate response to the dose a.s.o.

We can however mention that up to the end of the experimental period we could not find any exterior sign of discomfort as regards the animals' aspect and behaviour, as they appeared to be in perfect state of health (shiny fun, liveliness, a normal weight growth curve, etc), what makes us believe that the actual effects of HTO are less important than foreseen on the basis of the calculated dose.

This disagreement could be accounted for if the fact were accepted that tritiated water irradiation is largely wasted in nonvital zones. This is the tantamount to asserting that the tritium of HTO only to a small extent reaches the intracellular volume, as the equilibrium between the volume of the extracellular fluid and this volume is slowly established<sup>21</sup>. If this hypothesis could be proved to be real, the critical organs for HTO could be considered to be those, which are highly irrigated (bone marrow, spleen, etc).

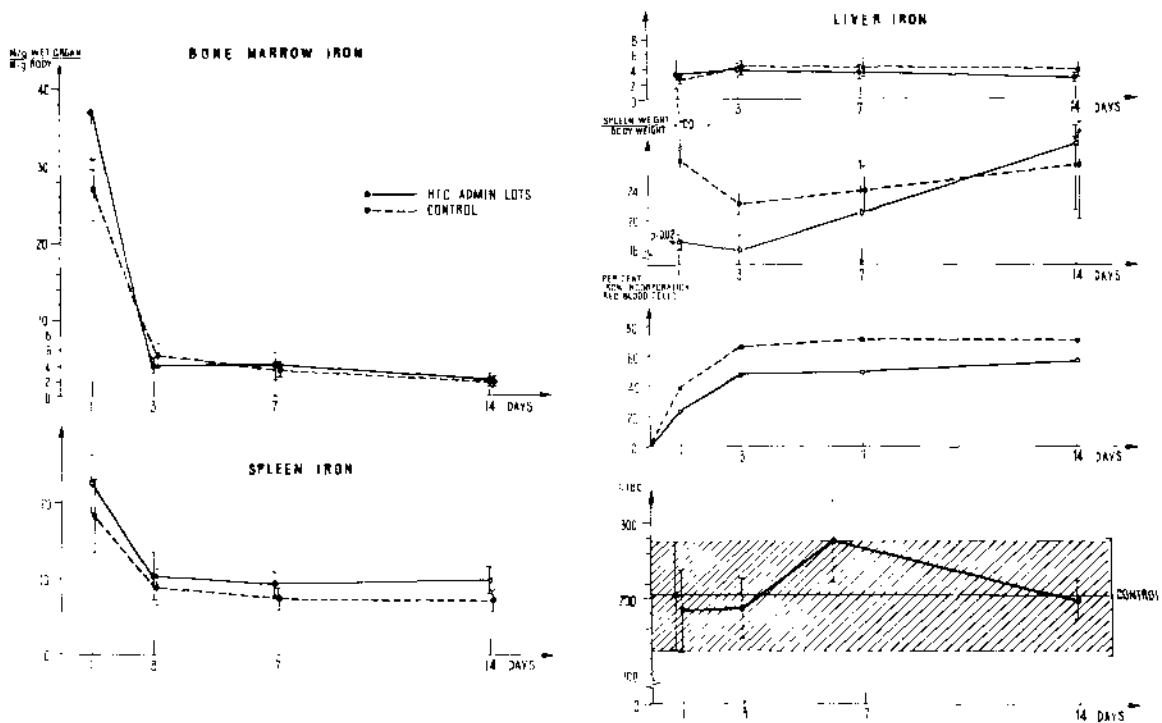
As regards the hazard of exposure to HTO, in spite of results of the present report, it is our opinion that any statement should be made with the greatest prudence, especially in cases of long term exposures to HTO when circumstances prevail in which an equilibrium is reached between the extracellular fluid and intracellular fluid, which could result in the binding of tritium to macromolecular compounds of metabolic importance.

#### Bibliography

1. Shiragai A., J. Rad. Res., 12, 73, 1971.
2. Shiragai A., J. Rad. Res., 13, 208, 1972.
3. Lyaginskaya A.M., Zaukova I.V., Veselovskaya K.A., Med. Radiol., 14/8, 61, 1969.
4. Bond V.P., Environ. Aspects of Nuclear Power Station, Proc. of Symp. N.Y. 10-14 Aug. 1970, 287.
5. Brozovich B., Copestake J., J. Clin. Pathol., 22, 509, 1969.
6. Bond V.P., in Manual on Radiation Haematology, Tech. Report Series No. 123, IAEA Vienna, 1973, 71.
7. Najean Y., Failla A., Dresch C., Rev. Europ. Etudes Clin. et Biol., XVI, 642, 1971.
8. Hellman S., Helen E. Grate, Blood, 38, 174, 1971.
9. Scaro J.L., Carrera M.A., De Tombolesi R.A.P., Texas Rep. Biol. Med., 29, 125, 1971.
10. Vacek A. et colab., Folia Biol., 14/6, 471, 1968.
11. Streffer C., Biochemical Indicators of Radiation Injury in Man, IAEA Vienna, 1971, 11.
12. Koch R., Irma Seiter, Strahlentherapie, 124, 79, 1964.
13. Harris E.B., in Manual on Radiation Haematology, Tech. Report Series No. 123, IAEA Vienna, 1971, 99.

14. Uteshev A.B., Satkojina E.A., Altibaeva Z.K., *Izv. Akad. Nauk. Kaz. S.S.R., Ser. Biol.*, 3, 66, 1970.
15. Dame J.V., in *Manual on Radiation Haematology Tech. Report Series No. 123*, IAEA Vienna, 1971, 215.
16. Maillie H.D., Mermagen H., *Health Phys.*, 20, 179, 1971.
17. MacDonald R.A., MacSween R.N.M., *Ann. N.Y. Acad. Sci.*, 165, 156, 1969
18. Fawwaz R.A., Winchell H.S., Pollycove M., Sargent T., *Blood*, 30, 417, 1967.
19. Stanculescu V., Ariadna Ciubotaru-Bordeianu, Herscovici H., Racoveanu N., Maria Iliescu, *Rev. Roum. Biochem.*, 8, 329, 1971.
20. Moscalev Iu., *Radiobiologhiceschii experiment i celovec*, Atomizdat, Moscova, 1970.
21. Albert S.N., Hirsch E.F., Economopoulos B., Albert C.A., *J. Nucl. Med.*, 9, 1, 19, 1968.

Fig. 1. The evolution of the indicators followed in animals which were given repeated administration of HTO.



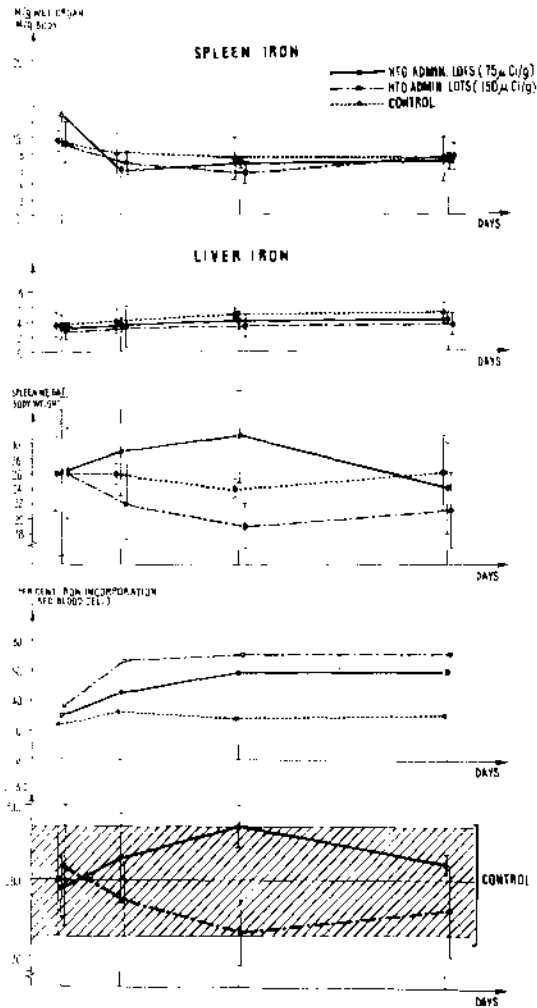


Fig. 2. The evolution of the indicators followed in animals which were given single administration of HTO.

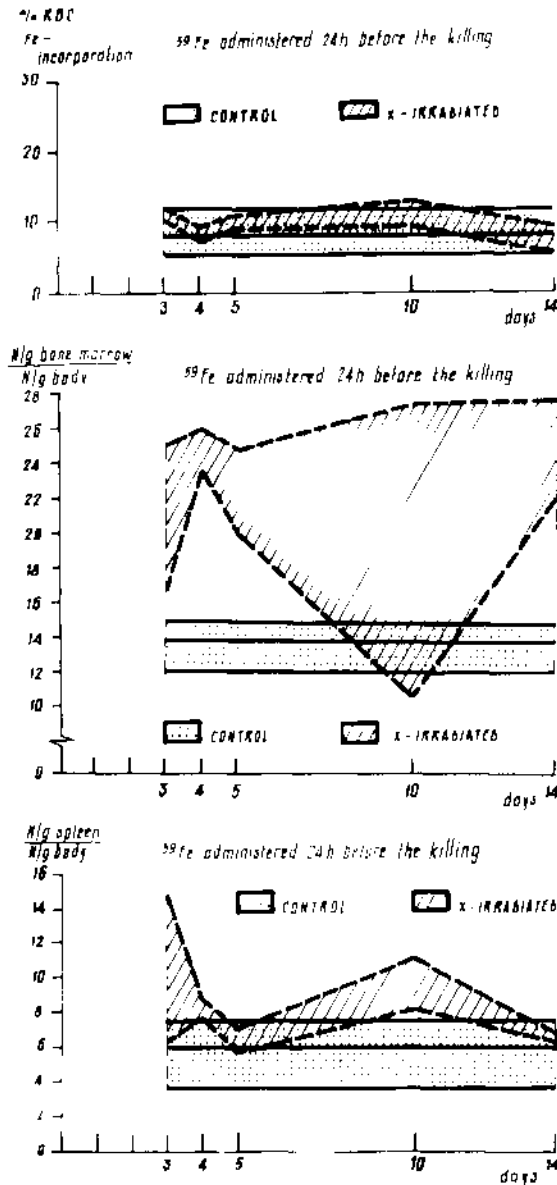
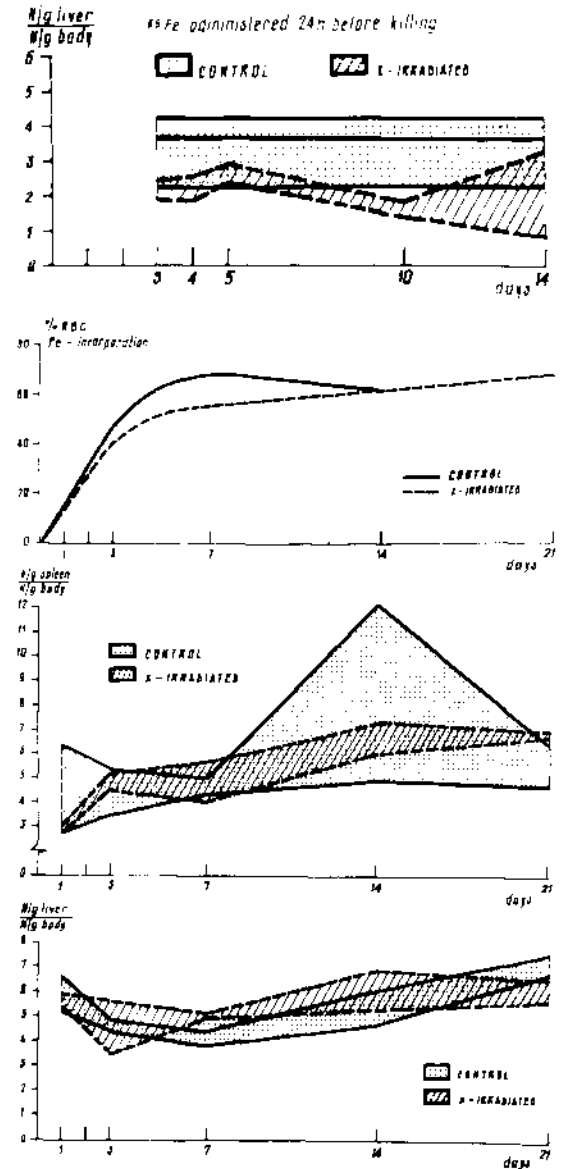


Fig. 3. The evolution of the indicators followed in animals X-irradiated with 50 R.



COMPARATIVE TOXICITY OF INHALED BETA-EMITTING  
RADIONUCLIDES IN BEAGLE DOGS\*

R. O. McClellan, B. B. Boecker, F. F. Hahn, C. H. Hobbs,  
R. K. Jones and M. B. Snipes

Inhalation Toxicology Research Institute  
Lovelace Foundation for Medical Education and Research  
Albuquerque, New Mexico 87108

Abstract

Four lifespan studies are being conducted in which Beagle dogs have been exposed via inhalation to aerosols of fused clay particles containing  $^{90}\text{Y}$ ,  $^{91}\text{Y}$ ,  $^{144}\text{Ce}$  or  $^{90}\text{Sr}$  resulting in graded initial lung burdens of radioactivity. With all four aerosols, the labeled particles are tenaciously retained in the lung with effective retention half-times of 2, 6, 53, 178 and 400 days, respectively. With the  $^{91}\text{Y}$ ,  $^{144}\text{Ce}$  and  $^{90}\text{Sr}$  in fused clay, the physical half-life is sufficiently long to allow translocation of small quantities of radioactivity, presumably in particulate form, to tracheobronchial lymph nodes and movement of solubilized  $^{91}\text{Y}$  and  $^{144}\text{Ce}$  to liver and skeleton and  $^{90}\text{Sr}$  to skeleton. With the highest initial lung burdens, deaths related to radiation pneumonitis and pulmonary fibrosis were observed in all four studies within 400 days post-inhalation exposure. The radiation dose required to produce early deaths was lowest for  $^{90}\text{Y}$  and highest for  $^{144}\text{Ce}$  and  $^{90}\text{Sr}$  being related to the lung retention half-times (and rate of decrease of radiation dose rate). Later deaths (>640 days) related to primary pulmonary hemangiosarcomas have been observed in dogs that inhaled  $^{144}\text{Ce}$  or  $^{90}\text{Sr}$  in fused clay with cumulative lung doses >20,000 rads. The current status of the studies (to 1800 days post-inhalation exposure) is discussed and compared with similar studies conducted with  $^{239}\text{PuO}_2$  at Battelle-Northwest. Of special interest is the development of primary pulmonary hemangiosarcomas (endothelial tumors) with chronic alpha irradiation from plutonium.

Introduction

The toxicity of inhaled radioactive materials is qualitatively well established. If high levels of radioactivity are inhaled, experimental animals die within a few hundred days from radiation pneumonitis and pulmonary fibrosis. Lower, but still substantial levels of exposure, produce primary pulmonary neoplasia in both experimental animals and man. Within this qualitative framework, however, there are relatively little quantitative data relating various levels of radiation exposure to biological endpoints such as pulmonary neoplasia and lifeshortening. Especially lacking is information that extends over a broad range of exposure levels and provides comparative data on different types of exposure such as alpha emitters versus beta emitters or radionuclides with short versus long effective half-times in lung. Recognizing these deficiencies in our knowledge and their significance in the establishment of radiation protection standards, studies were initiated to evaluate the toxicity of four beta-emitting radionuclides whose predicted effective retention half-times in lung varied from several days to more than several hundred

---

\*Research performed under AEC Contract AT(29-2)-1013 and in animal facilities fully accredited by the American Association for Accreditation of Laboratory Animal Care.

days. A range of exposure levels was used from high levels predicted to produce radiation pneumonitis, pulmonary fibrosis and early death to lower levels predicted to have a high probability of producing pulmonary neoplasia.

#### Experimental Approach

Beagle dogs, 12-14 months of age at time of exposure, were used; 365 exposed to radioactive aerosols and 51 sham exposed to serve as controls. The basic approach used in the studies has been described.<sup>1,2,3</sup> Radiation dose and experimental parameters are shown in Table 1. All dogs received a clinical examination before being placed on experiment and regularly thereafter.

Table 1  
Radiation Dose and Experimental Parameters  
for Radionuclides Inhaled in Fused Clay

Radiation Dose Parameters	Radionuclides			
	<sup>90</sup> Y	<sup>91</sup> Y	<sup>144</sup> Ce	<sup>90</sup> Sr
Physical Half-Life (Days)	2.6	59	285	10,500
Effective Half-Life in Lung (Days)	2.6	53	178	400
Average Beta Energy (Mev)	0.9	0.62	1.27	1.13
Radiation Dose to Lung Initial Dose Rate (rads/day/ $\mu$ Ci ILB*)	0.29	0.23	0.47	0.41
Potential Infinite Dose (rads/ $\mu$ Ci ILB)	1.2	16	110	250
<u>Radionuclide Exposed Dogs</u>				
Number	89	96	108	72
Range of ILB ( $\mu$ Ci/kg body wt)	80-5200	11-360	0.0024-210	3.7-94
Number of Deaths (< 500 days PE**)	37	36	17	32
Number of Deaths (> 500 days PE)	0	2	6 †	10 †
Current Survivors	52	59	85	30
<u>Control Dogs ‡</u>				
Number	12	12	15	12

\*ILB = Initial Lung Burden

\*\*PE = Post-Inhalation Exposure

† All due to primary lung cancer

‡ None have died to date

The aerosols labeled with either <sup>90</sup>Y, <sup>91</sup>Y, <sup>144</sup>Ce\* or <sup>90</sup>Sr\* were prepared by exchanging the radionuclide into montmorillonite clay and filtering and washing the clay. Aerosols generated with a nebulizer were fused to spherical insoluble clay particles by passing through a heating column at 1100°C and delivered to an exposure cone, through which the dog's nose projected, after being cooled with diluting air. The Activity Median Aerodynamic Diameter (AMAD) of the <sup>91</sup>Y, <sup>144</sup>Ce and <sup>90</sup>Sr aerosols as determined by cascade impactor analysis was about 1.8  $\mu$ m with a  $\sigma_g$  of 1.8. To reduce upper respiratory deposition, and thus the gastrointestinal dose, the AMAD was reduced to 0.9  $\mu$ m for the <sup>90</sup>Y exposures.

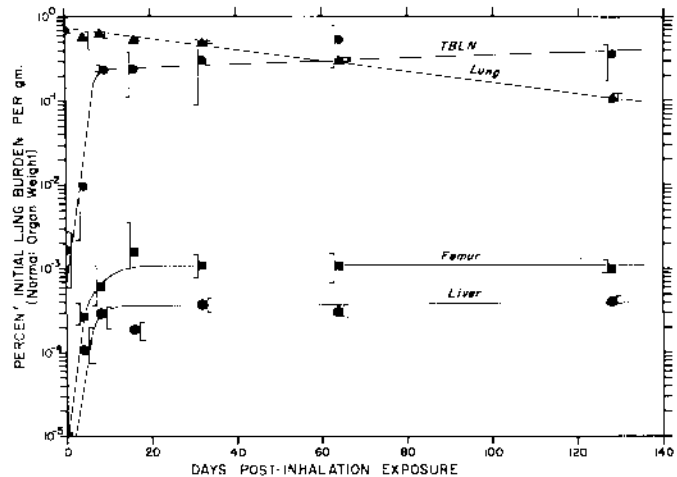
Gross necropsies and histopathological examinations were performed on all animals that died or were euthanized. If any radioactivity remained at death, the tissues were radioanalyzed. Data from periodic *in vivo* measurements of body and lung burden, excreta analysis and tissue radioactivity at death were used with data from studies of the radiation dose pattern for the specific radioactive aerosol to estimate the radiation dose for each animal.

\*<sup>144</sup>Ce in equilibrium with <sup>144</sup>Pr and <sup>90</sup>Sr in equilibrium with <sup>90</sup>Y.

### Experimental Findings to Date

In all four studies, the radioactive fused clay particles have been retained tenaciously in lung with effective half times as shown in Table 1. With  $^{90}\text{Y}$ -fused clay, the physical half life of  $^{90}\text{Y}$  was sufficiently short that minimal translocation to tracheobronchial lymph nodes and accumulation in liver and skeleton occurred.<sup>2</sup> With  $^{91}\text{Y}$ ,  $^{144}\text{Ce}$  and  $^{90}\text{Sr}$  fused clay, the effective half lives were sufficiently long to allow translocation of small quantities of radioactivity, presumably in particulate form, to tracheobronchial lymph nodes. In addition, with  $^{91}\text{Y}$  and  $^{144}\text{Ce}$ , radioactivity was accumulated by liver and skeleton and with  $^{90}\text{Sr}$  by skeleton; this probably represents solubilized material since it was distributed as it would be if a soluble form of the element were introduced into the bloodstream. The distribution pattern for  $^{91}\text{Y}$  in fused clay, shown in Figure 1, is typical of the translocation patterns observed. The highest radiation doses were to lung and tracheobronchial lymph nodes with much lower doses to bone, and for  $^{90}\text{Y}$ ,  $^{91}\text{Y}$  and  $^{144}\text{Ce}$ , to liver.

Figure 1. Tissue concentration of  $^{91}\text{Y}$  in Beagle dogs after inhalation of  $^{91}\text{Y}$  in fused clay; tissue weights: lung, 140 gm; TBLN, 0.5 gm; femur, 32 gm; and liver, 270 gm; mean and range of 3 dogs at each sacrifice time.



The current status of each of the four studies is summarized in Table 1 and schematically represented in Figures 2-5. In all four studies, a number of deaths occurred within 500 days after exposure with a suggestion that the deaths occurred earliest with  $^{90}\text{Y}$  followed by  $^{91}\text{Y}$  with the  $^{144}\text{Ce}$  and  $^{90}\text{Sr}$  deaths somewhat later. This apparent difference may be related to the initial radiation dose rate to lung which was highest for  $^{90}\text{Y}$  followed by  $^{91}\text{Y}$  and  $^{144}\text{Ce}$ , and then  $^{90}\text{Sr}$ , because higher levels of radioactivity were used with the radionuclides with short physical half life. The high dose rate exposures, although they decreased rapidly, produced death with lower cumulative doses than when the dose rate was low but sustained. The total radiation dose associated with early deaths was lowest for  $^{90}\text{Y}$  (>7000 rads), followed by  $^{91}\text{Y}$  (>14,000 rads), and was highest for  $^{144}\text{Ce}$  (>38,000 rads) except for one case in which death occurred with 19,000 rads, and  $^{90}\text{Sr}$  (>34,000 rads).

Early deaths were typically due to radiation pneumonitis and pulmonary fibrosis. The alveolar septa were thickened with hypertrophic and hyperplastic alveolar lining cells. The alveoli were frequently filled with proteinaceous material, macrophages and neutrophils. The fibrosis ranged from fine fibrillar thickening of alveolar septa to large dense collagenous scars. In many cases there was fibrous thickening of the pleura.

Late deaths, >500 days after inhalation exposure, are very radionuclide dependent at this time in the studies (Table 1). No late deaths have been observed with  $^{90}\text{Y}$ , only 2 with  $^{91}\text{Y}$ , 6 with  $^{144}\text{Ce}$  and 10 with  $^{90}\text{Sr}$ . The 2 late deaths with  $^{91}\text{Y}$  were due to pulmonary fibrosis with no evidence of neoplasia. Fifteen of the late deaths with  $^{144}\text{Ce}$  and  $^{90}\text{Sr}$  were caused by primary pulmonary hemangiosarcomas with one late death due to a hemangiosarcoma which arose in either the lung or rib. In addition, other tumors were found with the

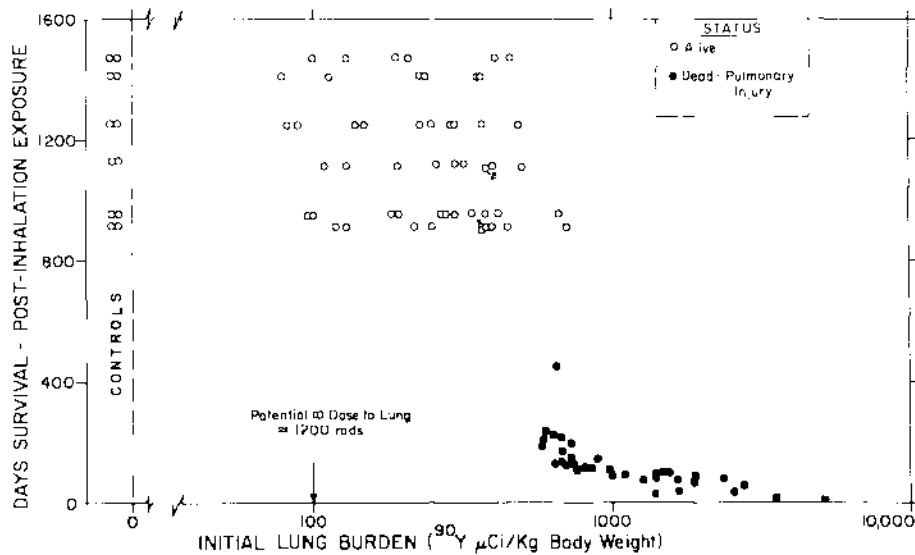


Figure 2. Survival of Beagle dogs after inhalation of  $^{90}\text{Y}$  in fused clay.

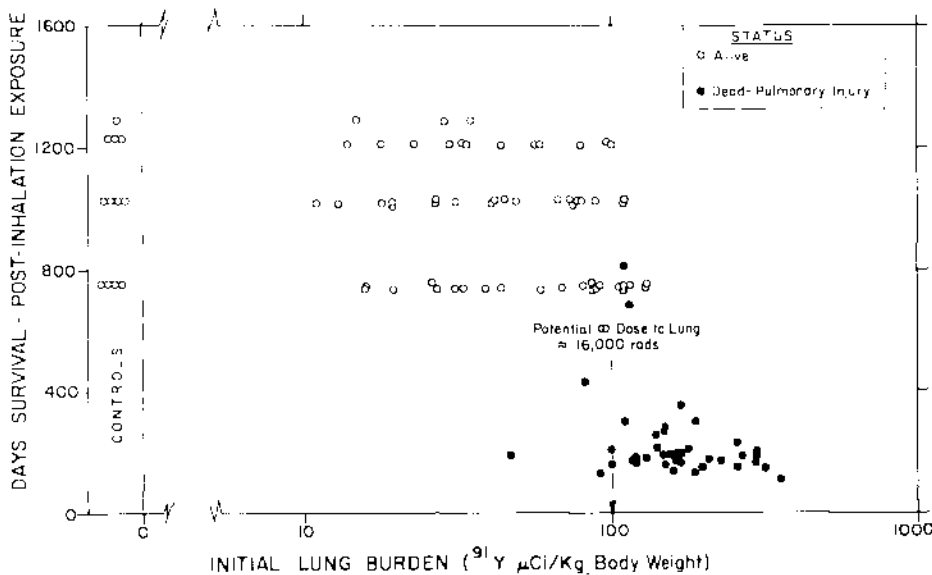


Figure 3. Survival of Beagle dogs after inhalation of  $^{91}\text{Y}$  in fused clay.

hemangiosarcomas; two bronchiolo-alveolar carcinomas, one fibrosarcoma primary to the lung and a squamous cell carcinoma of the nasal cavity.

The hemangiosarcomas were rapid growing tumors with death occurring within a few months after initial diagnosis. In most cases, there were multiple masses of tumor tissue in lung and there were numerous metastases to other tissues.<sup>4</sup> The high incidence of pulmonary hemangiosarcomas observed after inhalation of beta-emitting radionuclides is new and unexpected. No hemangiosarcomas of the lung have been found in control dogs in this colony, however, hemangiosarcomas of the skeleton have been observed in this colony after inhalation of  $^{90}\text{SrCl}_2$ .<sup>5</sup>

The contrast between the late effects of long-lived beta emitters in the lung reported here and the late effects of alpha-emitting  $^{239}\text{PuO}_2$  in the lung is striking. Howard<sup>6</sup> reported on 30 dogs followed for long periods of time after inhaling  $^{239}\text{Pu}$ . Twenty-two of the dogs died with primary lung neoplasms 1151 to 3313 days post-inhalation exposure while 8 dogs died with pulmonary



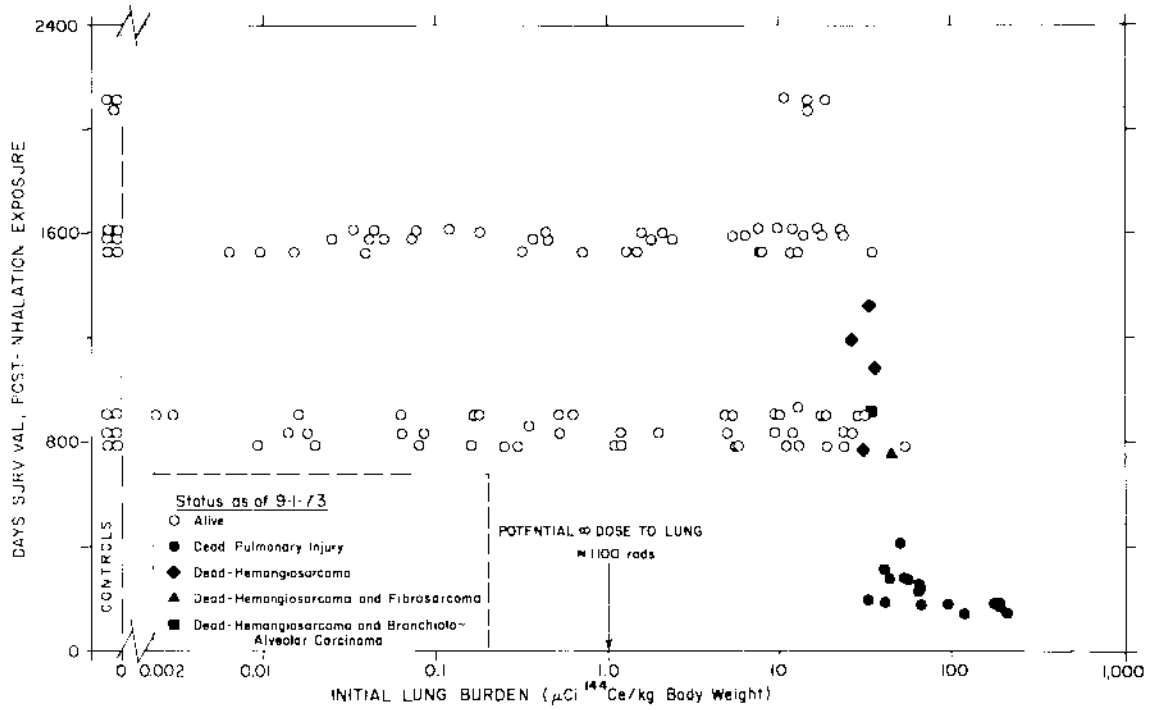


Figure 4. Survival of Beagle dogs after inhalation of  $^{144}\text{Ce}$  in fused clay.

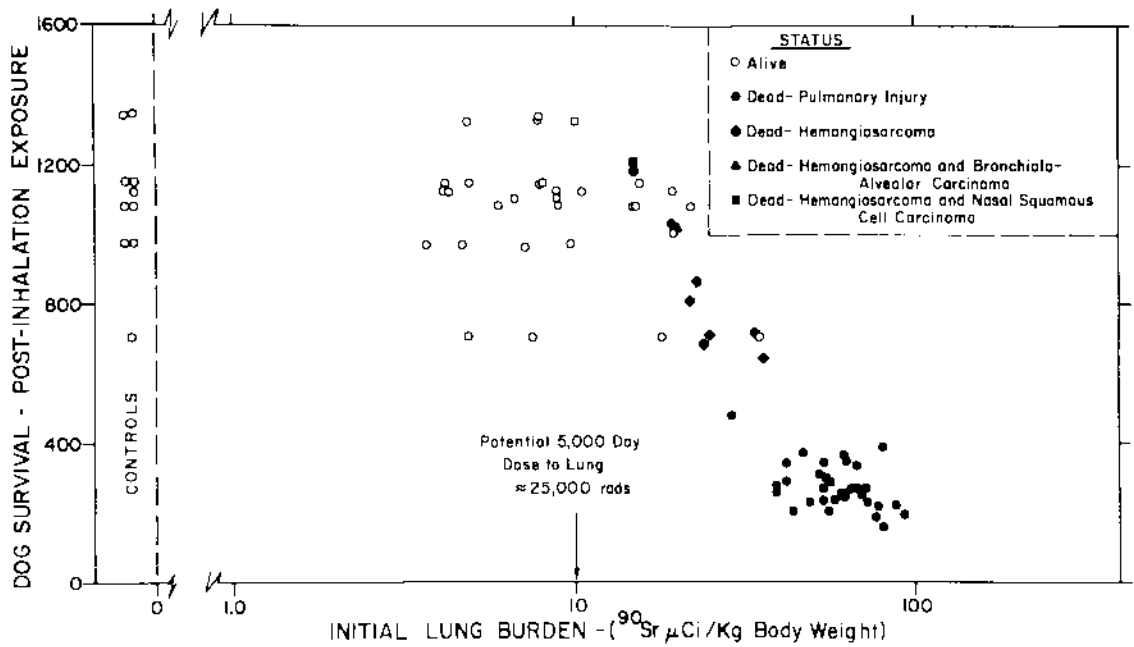


Figure 5. Survival of Beagle dogs after inhalation of  $^{90}\text{Sr}$  in fused clay.

fibrosis. Most of the neoplasms were bronchiolo-alveolar carcinomas of peripheral origin with two peripheral squamous cell carcinomas and three epidermoid carcinomas. Three dogs had tumors that appeared to originate from the pleura or mediastinum and two dogs had malignant lymphoma. Two dogs were observed to have benign appearing hemangiomas.

The basis for the predominance of different tumor types; hemangiosarcoma of endothelial origin with the beta emitters and the bronchiolo-alveolar carcinomas of epithelial origin with alpha emitters is not clear at this time. It is of note that the bronchiolo-alveolar carcinomas originated from areas of severe fibrosis and hyperplasia with associated alveolar epithelial metaplasia. This was also true for the bronchiolo-alveolar carcinoma observed in one of the  $^{144}\text{Ce}$  dogs. In general, however, in the dogs with hemangiosarcomas the pulmonary fibrosis and alveolar hyperplasia and metaplasia were not as marked as that described for the  $^{239}\text{PuO}_2$  dogs. The pathogenesis of bronchiolo-alveolar carcinomas may be intimately related to the process of fibrosis and associated alveolar hyperplasia and metaplasia. In contrast, perhaps the development of hemangiosarcomas from the substantial number of endothelial cells within the alveolar septae is related to the relatively uniform exposure and sublethal radiation damage produced by beta-irradiation of the lung. Perhaps  $^{239}\text{PuO}_2$  exposed animals are spared from this type neoplasm because of the more non-uniform radiation dose with numerous hot-spots and the higher LET of the alpha-emitting  $^{239}\text{PuO}_2$  particles.

Additional work is required to clarify the pathogenesis of lung cancer related to the several types of radiation exposure and especially to provide a firmer basis for extrapolating these findings to man. Future observations on dogs that still survive following exposure to lower initial lung burdens will be of special significance in considering radiation protection standards for man for long-lived beta emitters such as  $^{144}\text{Ce}$  or  $^{90}\text{Sr}$  deposited in lung. Comparative observations on the shorter-lived  $^{90}\text{Y}$  and  $^{91}\text{Y}$  in which exposures have not yet resulted in lung cancer will aid in establishing the dose-response significance of variations in the radiation dose pattern to lung.

Acknowledgements - The generous assistance of the staff of the Inhalation Toxicology Research Institute is gratefully acknowledged.

#### References

1. McClellan, R. O., Barnes, J. E., Boecker, B. B., Chiffelle, T. L., Hobbs, C. H., Jones, R. K., Mauderly, J. L., Pickrell, J. A. and Redman, H. C., Toxicity of Beta-Emitting Radionuclides Inhaled in Fused Clay Particles - An Experimental Approach. In Morphology of Experimental Respiratory Carcinogenesis, pp. 395-415, AEC Symposium Series 21, 1970.
2. Barnes, J. E., McClellan, R. O., Hobbs, C. H. and Kanapilly, G. M., Toxicity in the Dog of Inhaled  $^{90}\text{Y}$  in Fused Clay Particles: Distribution, Retention Kinetics and Dosimetry. Radiat. Res. 49: 416-429 (1972).
3. Hobbs, C. H., Barnes, J. E., McClellan, R. O., Chiffelle, T. L., Jones, R. K., Lundgren, D. L., Mauderly, J. L., Pickrell, J. A. and Rypka, F. W., Toxicity in the Dog of Inhaled  $^{90}\text{Y}$  in Fused Clay Particles: Early Biological Effects. Radiat. Res. 49: 430-460 (1972).
4. Hahn, F. F., Benjamin, S. A., Boecker, B. B., Chiffelle, T. L., Hobbs, C. H., Jones, R. K., McClellan, R. O., Pickrell, J. A. and Redman, H. C., Primary Pulmonary Neoplasms in Beagle Dogs Exposed to Aerosols of  $^{144}\text{Ce}$  in Fused Clay Particles. J. Natl. Cancer Inst. 50: 657-698 (1973).
5. McClellan, R. O., Benjamin, S. A., Boecker, B. B., Chiffelle, T. L., Hobbs, C. H., Jones, R. K., Pickrell, J. A. and Redman, H. C., Neoplasms in Dogs that Inhaled  $^{90}\text{SrCl}_2$ . In Radionuclide Carcinogenesis, pp. 181-200, AEC Symposium Series 39, 1972.
6. Howard, E. B., The Morphology of Experimental Lung Tumors in Beagle Dogs. In Morphology of Experimental Respiratory Carcinogenesis, pp. 147-160, AEC Symposium Series 21, 1970.

BENEFITS AND RISKS TO NUCLEAR POWER IN THE UNITED STATES  
OF THE "AS LOW AS PRACTICABLE" PHILOSOPHY

Morton I. Goldman, Sc.D.  
Senior Vice President  
NUS Corporation  
Rockville, Maryland

At the outset I should apologize to this group for the somewhat laborious title of this presentation. It resulted from a somewhat hasty consideration on my part of a title for the subject matter which I would like to present today; an attempt to communicate to you some of the difficulties that have been created in the United States by the enthusiastic application of the philosophy with which we in the radiation protection field have been living for several decades at least.

However, in attempting to present at least one individual's view of these problems, I am confronted with several difficulties, particularly for a distinguished international audience. One of these difficulties is created by the innate complexity of the agencies, organizations, and entities involved in U.S. radiation protection programs, as well as their assorted guides, standards, and regulations which apply in varying degrees to nuclear facilities. A second difficulty is established by attempting to relate the significance of other non-radiological laws, court decisions, and agency actions on radiological as well as non-radiological issues.

In the first area, we are involved with recommendations, evaluations, guides, standards, and regulations which at one time or another have been promulgated by the National Council on Radiation Protection and Measurements, the Federal Radiation Council, the National Academy of Sciences, the Environmental Protection Agency, and the Atomic Energy Commission, to say nothing of the influences exerted by the United Nations' Scientific Committee on Effects of Atomic Radiation and, of course, the International Commission on Radiological Protection, both of which have had a substantial influence on U.S. policies and practices in the radiation protection field. In the second instance of difficulty, we need only to look at the effects of the National Environmental Policy Act, or NEPA, and several judicial decisions about its interpretation to identify the significant impact these have had on radiological protection aspects of nuclear power, as well as on other non-radiological aspects of these facilities.

In this presentation, I would like to describe the potential threats as I see them to the viability of the generation of energy from nuclear sources in the U.S. which owe at least as much to the application of the "as low as practicable" philosophy as to any other source. It is these somewhat perhaps overzealous applications of the concept in the U.S. that I would like to explore this morning in the hope perhaps that other nations may avoid some of the pitfalls we appear to be creating in our own country.

Much of the zeal exhibited within the past few years has derived from efforts to reclaim the environment in general from a number of misapplications of technology which have resulted in the degradation of significant portions of our environment. At the same time and as part of the same movement, an intensive examination has been initiated of those environmental factors, whether natural or man-made, which may have more subtle effects on human health and welfare now that modern medicine and public health engineering have generally extended our life spans by eliminating, at least in large measure, the more obvious communicable diseases.

For the nuclear community, one of the more significant controversies over the past few years has been about the issue of radiation exposures deriving from the operation of commercial nuclear power plants. Certainly the names of Drs. Gofman, Tamplin, and Sternglass can evoke very predictable responses from those members of the nuclear community whose operations or plants have been affected by that controversy.

Additionally, the National Environmental Policy Act, as interpreted by the courts, has stimulated a broad inquiry into all other aspects of potential environmental impact with the objective of achieving a balance between benefits and costs, both economic and environmental. In many cases, however, pressures have been directed toward eliminating environmental effects regardless of cost or of the significance of the effect. In some of these instances, however, the application of a narrowly-focused effort to minimize one effect has created other, more significant effects.

As a quite recent, if peripheral example, I would cite a news article this weekend that quoted a scientist with the Environmental Protection Agency's Air Program research activity to the effect that catalytic devices proposed for removal of pollutants from automobile exhausts might in fact themselves discharge more harmful particulates of sulfuric acid and platinum.

More significant to the nuclear power industry (and in fact to the power industry in general) is the legislative goal of eliminating all discharges to waterways within 10 years. Although the guidelines for power plants are not yet promulgated, preliminary manifestations of the EPA policy are already evident. Actions in individual power plant cases, as well as a draft report on feasible technology, strongly suggest a mandate of closed cycle cooling using evaporative cooling towers for power plants regardless of site. Thus, "as low as practicable" may well become "as low as possible" in the water--with such potential adverse side effects as salt drift and deposition at coastal sites, evaporative depletion of fresh water resources in water-short areas, fogging and, in colder climates, icing of structures and roads. However, since these are not water pollution problems, they appear not to have been considered.

We have our own example of this type of "tunnel-vision" in the radiation protection field, however, and it is on this issue I would like to concentrate. It is related to the perspectives presented by Dr. Lapp yesterday--but presents still another aspect of considerable significance.

It is probably entirely superfluous to remind this audience that the ICRP, in 1950, stated: "It is strongly recommended that every effort be made to reduce exposures to all types of ionizing radiations to the lowest possible level." Although this injunction was directed primarily at occupational exposures, it did reaffirm for the radiation protection community the principle of minimizing exposure. In 1954 and again in 1957, the NCRP recommended that "exposure to radiation be kept at the lowest practicable level in all cases." In the 1957

statement, NCRP also made recommendations for the limitation of exposure to individuals outside of "controlled areas," as well as for population exposure.

Although the recommendations of the ICRP and NCRP were widely adopted by operating organizations, these recommendations as such carried no statutory force. To provide a federal policy in the U.S. for human radiation exposure, the Federal Radiation Council was formed in 1959 to "advise the President with respect to radiation matters directly or indirectly affecting health." In 1960, the first report and recommendation by the Federal Radiation Council staff essentially adopted the recommendations of the ICRP and NCRP, but selected the term "radiation protection guide" for the "maximum permissible dose" concepts of the other organizations. Again the staff report stated: "Every effort should be made to encourage the maintenance of radiation doses as far below this guide as practicable."

During the 1950's and 1960's the Atomic Energy Commission, which provided the regulatory control over all major nuclear activities, had adopted its own radiation protection standards. These were in two forms: the first, a codified section of the Code of Federal Regulations, the so-called Part 20, applicable to licensees of the Atomic Energy Commission; and the other, a portion of the AEC manual appropriate for internal operations of the AEC and its contractors at such facilities as Oak Ridge, Hanford, Savannah River, etc. It is, of course, Part 20 of the AEC regulations that apply to the nuclear power facilities that are being built and operated by electric utility companies in the U.S.

In 1968 and 1969, substantial controversies were generated in the U.S. by a number of individuals with respect to the projected effects of exposure of populations to doses from nuclear plant operations at the limits prescribed by FRC guides for the population at large. Needless to say, the allegations by Gofman and Tamplin created a widespread controversy not only in the United States but elsewhere as well. Although these allegations were refuted at the time and more convincingly since by BEIR, one of the difficulties in dealing with them derived from the fact that Part 20 of the AEC regulations dealing with licensed facilities nowhere explicitly included the ICRP/NCRP/FRC recommendation that exposures be maintained at the minimum practicable value.

I pointed this out in testimony before the Joint Committee on Atomic Energy in January 1970, although I indicated then that in practice the Regulatory Staff of the Atomic Energy Commission had in fact applied that philosophy in their review and approval of facilities seeking permits and licenses. In April of 1970, the AEC proposed amendments to their regulations which would include the "as low as practicable" mandate in the Part 20 standards. Additionally, AEC proposed amendments to that section of their regulations covering licensing of nuclear facilities\* which would require applicants to identify the design objectives for discharges and the means to be employed to keep discharges as low as practicable, as well as a requirement for operating plants that equipment installed in effluent management systems be maintained and used to assure that discharges were as low as practicable.

At that time, the Commission indicated its plans to consult with the industry and with other interested groups and persons to determine the feasibility of developing more definitive criteria for the design objectives and the means for keeping radioactive discharges from light-water-cooled nuclear plants as as low as practicable. The proposed amendments to Part 20 and Part 50 were

---

\* Part 50

adopted in December of 1970, and early in 1971 the AEC carried out its announced intention to meet with industrial and other representatives to review the feasibility of establishing more definitive numerical guides. In December of 1970 also, a reorganization of part of the Federal establishment transferred the functions of the Federal Radiation Council to the newly established Environmental Protection Agency.

In June of 1971, the Atomic Energy Commission issued for comment a proposed Appendix I to their regulations for licensing of nuclear facilities which established numerical guides for design objectives and limiting conditions for operation governing discharges of radioactive materials in effluents from light-water-cooled nuclear power plants. These were intended to implement the "as low as practicable" concept and basically provided for discharge limits which would result in exposures 1% of the ICRP/NCRP/FRC recommendations for individuals in uncontrolled areas, or 5 millirem per year. The position of the Regulatory Staff was that the discharges reported for existing plants indicated the general feasibility of meeting these new lower limits at modest expense and that the values needed to be in that range in order to protect the public health and safety as nuclear power became a more significant contributor to both electric generating capability of the United States and to the dose to the population of the United States.

A very large number of comments were received from utility organizations, from reactor manufacturers, from architect-engineers, from consultants, from environmentalists, and from other government agencies, and predictably enough ranged from the viewpoint that the guides were unnecessarily restrictive to the opposite.

A rulemaking hearing was held during the first five months of 1972 with participation by a combined group of utilities, by a reactor manufacturer, by a consolidated group of environmental intervenors, and by the State of Minnesota. During the course of these rulemaking hearings, several factors became apparent. The first, and perhaps most important, was the intention of the Regulatory Staff to apply the same degree of conservatism in arriving at projections of radioactive releases and in assessing the resulting doses as had been applied when the standards were greater by two orders of magnitude. In particular, it was proposed to include not only normal waste releases but also those that might result from off-standard operation, so-called miscellaneous leakages. Few if any of these had been measured and reported by the operating plants, so the basis on which the practicability of meeting the new guide had presumably been demonstrated was immediately in question. In the dose assessment area, perhaps the most notable of the Staff conservatisms became the "hypothetical future cow" which was to be assumed to be chained to the nearest site fencepost, with an infant permanently attached to its hind leg, even if such farming uses did not exist in the present or foreseeable future.

It appeared obvious to several of us who participated in that hearing that the Staff had not really done its homework in evaluating the impact of their proposed rule either in terms of costs of compliance in an economic sense or with the benefit to be obtained as a result of these costs. One of the AEC's own biomedical witnesses, Dr. Leonard Sagan, indicated that in his view the expenditures required for meeting these proposed limits might better be spent in controlling and reducing exposures of the public from X-ray machines. I think Ralph Nafer might even agree with that position if he had better advisers than John Gofman. In the instance of the proposed limit for radioactive iodines, the combined utility group produced testimony which indicated that it was highly unlikely that the proposed limit was even feasible to

achieve, particularly under the assessment ground rules established by the licensing Staff, and that the costs of compliance with the proposed rule could reach very large figures very easily.

Even the environmentalist intervenor group witnesses, with the exception of Dr. Sternglass, generally supported higher radiation exposure limits than those proposed by the AEC. A final point, and one which I will expand on a little later, was made by the combined utility group--that the staff had given no consideration to the potential increase in plant worker exposure which might result from the adoption of these more stringent restrictions on releases from the plant.

At the end of the hearing in May of 1972, the AEC announced that it would prepare and issue an Environmental Statement on the proposed rule to assess the costs and benefits of its implementation and to consider alternatives to the proposed rule. This Environmental Statement was an outgrowth of the requirements of the National Environmental Policy Act that any Federal action having a significant impact on the environment must be preceded by an environmental statement which examines the costs, benefits, and alternatives to the proposed Federal action. The Draft Environmental Statement was issued for comment in January 1973.

The direction and scope of the Draft Environmental Statement tended to support the view of some individuals that the AEC decision to establish 1% of the NCRP limits as a design objective may not have been as much technically motivated, but rather politically stimulated by a desire to silence their critics. The Draft Statement was a fairly substantial exercise in numerical calculations which attempted to demonstrate by the sheer weight of numbers that the 5 millirem per year dose objective was indeed achievable although at some cost. It did not, for example, consider in any real sense alternatives to the proposed numerical guidance of 5 millirem per year. It appeared in summary to be a massive exercise in self-justification.

The methodology employed in the Draft Statement was to establish models of boiling water and pressurized water reactors; to hypothesize radioactivity releases from these plants using alternative waste treatment schemes which started with the base cases being essentially open sewers; to establish these hypothetical plants in so-called representative sites; and to assess the doses to individuals and to populations resulting from the hypothesized releases from alternative waste processing schemes. Estimates were made of the costs of these additional waste systems and the differential costs associated with the resultant reduced exposures.

In these assessments, many of the same conservatisms employed by the Regulatory Staff in individual plant licensing actions were largely reiterated. Normal operation was assumed to include a wide variety of off-standard operations, including fluid leakages, fuel failures, etc., which resulted in a tendency, as the statement indicated, "to overestimate the quantity of radioactive material released in the effluents."

To determine the impact of these releases on hypothetical individuals and population groups around these hypothesized plants, a number of interesting characteristics were ascribed to the so-called maximum individual. This individual was used to represent the "maximum" dose which could be postulated for an individual in the station environs. This in itself was at variance with the stated intent of Appendix I to deal with "expected" rather than maximum situations. "The individual is assumed to have been conceived, born, and expected to live his entire lifetime on a farm adjacent to the station site boundary. He is assumed to eat nominal amounts of fish,

crustacea, mollusks, and aquatic vegetation (if applicable) taken from near the outfall of the liquid effluent; to obtain his drinking water from near the site boundary (for sites adjacent to fresh water); to swim and boat in the waterway near the outfall; to engage in normal recreational activities on the shoreline near the station; to eat fresh green vegetables grown on the farm; and to drink fresh milk obtained from cows on local pasture. Further, the farm is assumed to be located in the sector downwind of the predominant wind direction."

From the numerical attributes assigned to this individual in the Draft Environmental Statement, he was truly heroic. He ingested two and one-half times as much fluid as does the ICRP standard man; he ate as much seafood annually as does a commercial fisherman, except near the sea coast site where he ate twice as much seafood as does a U.S. commercial fisherman; he also spent 500 hours per year on the shoreline near the plant outfall, and 200 hours more swimming or boating near the outfall, presumably to catch all the fish he eats. At the same time, he survived largely on spinach, lettuce, and cabbage grown in the most exposed off-site area in an amount equal to half the U.S. average total annual vegetable intake, and drank about three times as much milk from his own cow (presumably pastured in the vegetable patch) as does the average individual in the U.S., while simultaneously remaining naked all year 'round on the downwind fencepost next to the cow. Sometime during this period, he also found time to arrange for a new infant every year. While the concept of the critical population group is not a new one in the radiation protection field, this exercise did appear to be advancing the state of that art by a substantial amount.

Having gone through this exercise, the Draft Environmental Statement came to the conclusion that with the exception of radioactive iodines, it appeared feasible and not economically unreasonable to achieve the design objective of 5 millirem per year. However, in order to achieve this design objective for radioactive iodine discharges, it would have appeared to be necessary to exhaust all ventilation air flows from turbine, auxiliary, and reactor buildings through perhaps a thousand high efficiency filters and charcoal absorbers units and 100 meter tall stacks. Since there are a large number of plants which are being designed and built in the U.S. under the apparent misconception that "clean nuclear power" does not require chimneys, this came as somewhat of a shock.

In fact, the Draft Environmental Statement confirmed what the combined utility group had stated during the hearing; e.g., there did not seem to be any biological rationale for the dose objective of 5 millirem per year and, in any event, it appeared highly infeasible to assure meeting the restrictions on release of radioactive iodine. What was not considered in the Draft Environmental Statement was the necessity for meeting the proposed guideline for that substance let alone for the other discharges.

The costs of achieving the desired degree of protection were not insignificant even if the estimates made on behalf of the AEC were accepted at face value. These estimates indicated that the annual incremental costs for control of radioiodines might range between \$500,000 and \$1,000,000 per year for pressurized water reactors and between \$1,000,000 and \$2,000,000 per year for boiling water reactors for such control. Needless to say, one could buy significant amounts of land or cows or liters of milk for that annual cost. It was suspected, in addition, that the costs of control contained in the Draft Environmental Statement were low perhaps by as much as a factor of two.



Despite the information introduced in the hearing by the utilities, not one word appeared in the Draft Environmental Statement about the potential impact on in-plant personnel exposures resulting from the adoption of the proposed rule, nor was any consideration given to those facilities which might incorporate more than two units per site.

Over three hundred pages of comments on the Draft Environmental Statement were filed, the bulk of which were critical of the approach and the substance of the Draft Environmental Statement. Late in July of this year, the Final Environmental Statement was published by the AEC. It is substantially bulkier but not significantly better than the Draft Statement. The Final Environmental Statement consists of three volumes: the first of which is the Statement itself; the second, a description of the analytical models and calculations employed; and the third, a compendium of comments and the responses of the Staff to those comments.

Many, if not most, of the deficiencies in the Draft Environmental Statement remain. No significant consideration has been given to the costs and risks of a guideline higher than 5 millirem per year; the conservatism in estimation of source terms for releases from plants has been maintained in much the same form as in the original draft statement; the heroic proportions of the so-called maximum individual are essentially unchanged from those in the Draft Statement, although at least a step in the direction of assuming more reasonable parameters for the population at large has been taken for estimation of population dose; the cost figures used by the Staff in the Final Environmental Statement appear to be about a factor of two lower than the cost estimates for comparable pieces of equipment in the Draft Environmental Statement (the original estimates were felt perhaps to be low by as much as a factor of two).

There has been some relaxation of the attitude of the Regulatory Staff in respect to the potential capabilities to deal with radioactive iodine. The Final Environmental Statement does indicate that perhaps the 5 millirem per year objective is not a practically achievable limit at this time, and the figure of 15 millirem earlier produced in an Atomic Energy Commission Safety Guide is presented as a potential alternative for the initially proposed value.

However, in my view, the document still neglects a major consideration in the cost-benefit balancing that needs to be done prior to the adoption of these extremely stringent limits on discharges. In the Draft Statement, the AEC recognized the values of dollar cost per man-rem of exposure that have been assigned by a number of authorities over the years. (Dr. Lapp yesterday mentioned \$60 per man-rem, for example.) It, perhaps understandably, chooses not to balance directly the costs of systems to provide a man-rem reduction against the value of the man-rem's so saved, although in a number of instances the ratio between the cost of saving a man-rem and the highest value that might be assigned to it are so far out of balance that the expenditures must be regarded as nonsensical. There is one area, however, in which a direct comparison could have been attempted but was not. This area is the one to which I referred earlier and about which a number of comments were made by those who reviewed and commented on both the initial Appendix I and the Draft Environmental Statement. I refer here to the potential impact on occupational exposures. There is no treatment of occupational exposure impact whatsoever in the Final Environmental Statement, despite the evidence put forth in the hearing itself, the comments made on the Draft Environmental Statement, and in fact comments and findings made by at least one Atomic Safety and Licensing Board and an Appeal Board.

In one power plant licensing case in which the Staff evaluation deliberately excluded the dose received by on-site personnel, the Licensing Board stated, "Since the significance (if any) of general population exposure at these low levels lies in the impact on the gene pool of the entire population rather than in its relatively trivial somatic effects, from the point of view of the cost-benefit analysis, the environmental cost of the total genetic dose received by the entire population (including employees) is the important factor. The genetic dose to employees should not be considered as a voluntary occupational exposure, but rather as a dose to future generations and, therefore, fully comparable to the dose to non-employees." The Appeal Board in that case added the following: "We would add to these observations only the obvious fact that the more stringent the limitation on liquid and gaseous emissions from the facility, the greater the radiation exposure likely to be experienced by on-site plant personnel. Thus, a concomitant of undue conservatism in the matter of computation of off-site emissions may be the subjection of the plant personnel to an unnecessary genetic dosage."

The growing significance of occupational exposure to operating and maintenance personnel at nuclear power facilities has been the subject of a number of papers over the past few years. We identified this problem in 1971 presentations based on data from a number of our clients, and others. Its growing significance has been attested to by the incorporation in the 1972 report of UNSCEAR of the doses from occupational exposure at nuclear power plants around the world. For these data which cover twenty stations with a gross electrical output of almost 19,000 megawatt-years, the average person-rads per MWe-year is almost 1 with a range of 0.2 to 8.9 man-rads per megawatt-year.

We have examined film badge exposure records of utility staff and contractors employed at nuclear power stations in the U.S. This analysis has treated 13 operating stations of the light-water type which have generated some 12,870 megawatt-years of electric energy and in the process reported total man-rem external exposures of over 18,000 man-rem, an average of about 1.4 man-rem per megawatt-year of electric energy. For these light-water plants, the range of exposure has been between 0.12 man-rem per megawatt-year and 4.5 man-rem per megawatt-year. Generally, the higher values tend to be associated with the older, smaller plants although this is not universally the case. The lower end of the spectrum tends to represent the newer, larger plants which have been operating only for one to two years and have not undergone either refueling or significant major maintenance activities which tend to substantially increase the total man-rem exposure.

Although it has not been possible at the present time in more than a very few cases to identify man-rem exposures associated with particular jobs at a representative number of nuclear power plants, the data that are available indicate that the overwhelming majority of the accumulated exposure derives from activities carried on during the refueling and major maintenance periods at these reactors. In any event, it is quite difficult to relate particular job exposures to potential requirements for maintenance work on systems demanded by the Appendix I requirements. It is not difficult, however, to extrapolate some of the job exposures that have already been reported to the potential requirements of implementing the "as low as practicable" policy of the Regulatory Staff and to observe intuitively the great dis-benefit that is quite likely to accrue as a result.

For example, typical values of exposure for specific operating and maintenance items have been reported by some operating nuclear power plants. At one plant, for example, it was reported that, exclusive of supervision or technician assistance, the removal of spent demineralizer resins for shipment

involved an exposure of 4.8 man-rem. This would certainly be a more frequent occurrence under the expanded systems proposed to meet the requirements of the "as low as practicable" limits. The sipping or sampling of fifty fuel bundles for the detection of leaky fuel elements, which would almost certainly be required more frequently under the "as low as practicable" restrictions, at one plant produced an integrated exposure of 9.6 man-rem. The changing of waste filter cartridge units yielded an exposure of 0.2 man-rem. Doses accumulated during the inspection and repair of steam generators have ranged from a low of about 9 man-rem to a high in excess of 350 man-rem. In most, if not all these cases, the basic motivation for repair of the steam generator rested in the inability to comply with discharge limitations with continued primary-to-secondary leakage although in no case would population exposure to the public have been contributed which was remotely equivalent to that generated in-plant by the inspection and repair operations.

It is somewhat discouraging to me that it was only a few months ago, in April of this year, the Regulatory Staff began to request man-rem occupational exposure data from plant operators by job function. Considering the significance of these data, it seems hardly logical to have proceeded with the establishment of discharge standards in their absence.

No doubt much can be done to improve the ease and safety with which maintenance activities can be conducted, although for many of the maintenance jobs required, substantial reductions could only be accomplished by a significant change in the way nuclear plants are currently designed to one more closely resembling the fuel recovery plants in which remote maintenance activities are standard. Failing this, it does not appear as though substantial reductions in occupational exposure are likely to result from improved shielding, for example, or other similar plant modifications. Much of the equipment requires direct contact for maintenance and replacement of parts which cannot conveniently be performed in any other way.

On this basis and projecting the current experience in the U.S., it appears to me that the occupational contribution to the population genetic burden from nuclear power plants alone will substantially outweigh the general population contribution to that total by factors approaching two orders of magnitude. If you recall, ICRP suggested in 1958 in its illustrative apportionment of genetic dose that the exposure of the population at large might be twice as great a contribution to overall population exposure as would the occupational contribution.

The AEC has estimated that by the year 2000, light-water-reactors will provide a capability for generating 500,000 megawatts of electric power. Assuming a capacity factor of 80%, in that year a total of 400,000 megawatt-years of electric energy would be generated, and projecting current occupational exposure experience, this would result in a population exposure from this source of about 560,000 man-rems or almost 2 millirem per capita in that year. On the other hand, the Atomic Energy Commission in their Final Environmental Statement projects a population exposure from the operation of these plants of 24,900 man-rem, less than 0.08 millirem per capita-year, or 0.06 man-rem per megawatt-year. An examination of assessments by the AEC in their Environmental Statements for some 35 individual nuclear plants totalling 56,580 megawatts of capacity, yields a total projection of 728 man-rem per year or about 0.016 man-rem per megawatt-year (at 80% capacity) for these plants.

Given the conservatism inherent in the Staff analyses of population dose with respect both to transport and uptake, the ratio between occupational and the general public contributions to population exposure of between

20 and 90 can probably be increased by at least a factor of 2 to 3. However, accepting even that range of about 20 - 90 as it stands, it seems to me that there is a serious deficiency in any approach to reduction of environmental exposure standards of this magnitude without providing at least equal attention to the potential hazards of the trade-off that almost certainly will occur as a result.

Certainly the in-plant staff exposures are real and, on average, substantially higher than those to which even the nearest neighbors of nuclear plants have been exposed to in the past or are likely to be exposed in the future. Exposures, which have been experienced by neighbors to the plant, on the other hand, have been essentially invisible when compared to the natural background in the area. Under the new policy, these would be even more theoretical than real and the resulting population exposures would also be hypothetical rather than genuine exposures. It seems to me, then, that we are facing a trade-off of an increase in real, measurable exposures for an invisible decrease in hypothetical exposures, one which does not comport well with the basic radiation protection philosophies, at least those with which I am familiar.

While the Atomic Energy Commission has been vigorously defending its proposal to limit these releases from light-water plants, the National Academy of Sciences' Advisory Committee on the Biological Effects of Ionizing Radiation has been assembling an analysis of current knowledge relating to risks from exposure to ionizing radiation. This was published in November, 1972, and some extracts from this report, which was extensively discussed yesterday, provide an interesting counterpoint to the performances of the Atomic Energy Commission. Several of the recommendations of the BEIR Committee and their conclusions are of interest, particularly when examined in the context of the present controversy. I would like to quote some of these recommendations and conclusions.

"It is suggested that numerical radiation standards be considered for each major type of radiation exposure based on the results of cost-benefit analysis."

"In addition to individual and average population guidelines, we recommend that an additional limitation be formulated... that takes into account the product of the radiation exposure and the number of persons exposed; this might be expressed in terms of person-rem. This need arises from acceptance of the non-threshold approach in risk estimates which implies that absolute harm in the population will be related to such a product."

"Guidance for the nuclear power industry should be established on the basis of cost-benefit analysis, particularly taking into account the total biological and environmental risks of the various options available and the cost-effectiveness of reducing these risks."

"Thus we say: The total future cost of one man-rem, in terms of health costs paid for in present dollars, is between \$12 and \$120. This may provide one way for putting a dollar value on a dose commitment of one rem that could be used in cost-benefit calculations. The cost would be distributed over many generations in the future."

"The public must be protected from radiation but not to the extent that the degree of protection provided results in the substitution of a worse hazard for the radiation avoided. Additionally, there should not be attempted the reduction of

small risks even further at the cost of large sums of money that spent otherwise, would clearly produce greater benefit."

The AEC position on the "as low as practicable" guidelines results from a substantial overestimation of the likely public exposures and underestimation of the cost of minimizing these exposures, producing a totally unrealistic cost-effectiveness index. When this is coupled with the total neglect of the in-plant exposure cost that may accrue, there exists a real and substantial basis for questioning whether the AEC has not, by this proposed action, generated more risk to the public of the United States by over-reacting to what has been in the past and will undoubtedly continue to be in the future an insignificant threat to the health of the public. It is my sincere hope that other nations will not blindly follow these actions of the U.S. Atomic Energy Commission without a very careful consideration of the implications of such a move on the total exposure budget of their populations. Certainly the blind appeasement of a small but loud segment of the public has not, in my view, contributed materially to the overall radiological welfare of the United States.

Thank you.

# RADIATION QUANTITIES AND UNITS

## THE DOSE EQUIVALENT EQUATION

H.O. Wyckoff

International Commission on Radiation Units and Measurements  
Washington, D.C. 20014

In 1962, discussions between the International Commission on Radiation Units and Measurements (ICRU) and the International Commission on Radiological Protection (ICRP) resulted in the formulation of a statement on dose equivalent, and this statement with minor modifications was utilized in subsequent publications of the Commissions.

At the time the statement was first published it was noted that, "although this statement does not cover a number of theoretical aspects (in particular the physical dimensions of some of the quantities) it fulfills the immediate requirement for an unequivocal specification of a scale that may be used for a numerical expression in radiation protection". One might view the dose equivalent as having the dimension of absorbed dose, being a biophysical quantity or representing risk and therefore physically dimensionless. A brief discussion of the reasons for each interpretation is included. The interpretation recommended by the ICRU -with ICRP concurrence- is presented.

### Introduction

The dose equivalent is a quantity determined in an organ or tissue of the body of a person and related to the presumed risk of radiation induced injury to that organ or tissue. The use of this quantity is limited to radiation protection applications and should not be used for high level accidental exposures. It is defined by the equation

$$H = \underline{D} Q N$$

where  $\underline{D}$  is the absorbed dose determined at the site of the presumed radiation induced injury,

Q is a modifying factor related to radiation quality at the site and whose numerical values are given in terms of the collision stopping power in water in figure 1 and,

N represents the product of all other modifying factors.<sup>1</sup>

When  $\underline{D}$  is in rads,  $H$  is in rems. This definition is adequate for practical determinations, but does not spell out the meaning of  $H$  and in particular its dimensions. The dose equivalent could be, among other things, either a purely physical quantity, a biophysical quantity, or a representation of the risk of radiation induced injury and therefore dimensionless. Each interpretation has its following within the scientific community. These three interpretations will be discussed in the

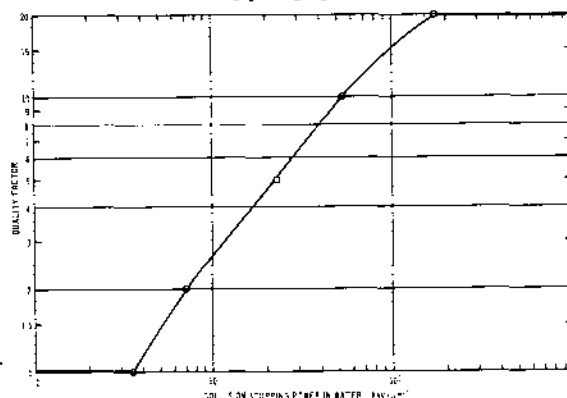


Fig. 1 The quality factor as a function of the collision stopping power in water.

present paper and the ICRU recommended interpretation outlined.

#### A Physical Quantity

A physical quantity denotes a physical entity or concept used for the precise description of a phenomena and defined so as to be measurable. Proponents of a purely physical interpretation of dose equivalent view the modifying factors  $Q$  and  $N$  as purely physical factors. They point out that  $Q$  is related to the radiobiological effectiveness (RBE) which is merely the inverse ratio of absorbed doses of different types and/or energies of radiation that produce the same biological effect. It is recognized that the magnitude of the RBE is obtainable experimentally only for large absorbed doses and absorbed dose rates and that its magnitude may change with absorbed dose and absorbed dose rate. Thus, the value of  $Q$  should be obtained ideally by extrapolation of observed RBE values to the absorbed dose and absorbed dose rates encountered in the radiation protection area. Such an extrapolation will not modify the dimensions so that  $Q$  is dimensionless.

Other factors that might be included in  $N$  are obtained in a similar fashion. For example, if one were to incorporate an absorbed dose rate factor, it would be obtained from the inverse ratio of the absorbed doses at different rates that would produce the same effect. This factor is also dimensionless. Again, these experimental data obtained at high doses ideally would be extrapolated to the absorbed doses of interest for radiation protection.

With such an interpretation,  $Q$  and  $N$  are dimensionless and the dimensions of  $\underline{D}$  and  $\underline{H}$  are identical (energy/mass). In this case, the units of  $\underline{D}$  and  $\underline{H}$  are the same. This means that both  $\underline{D}$  and  $\underline{H}$  can be expressed in rads or joules per kilogram.

#### A Biophysical Quantity

Proponents of this interpretation agree that the magnitude of  $Q$  and  $N$  are inferred from physical measurements-- for example, the value of  $Q$  being inferred from the inverse ratio of absorbed doses required to produce the same biological effect. However, they point out that the magnitude of the ratio depends upon the biological end point. In their view, this is a different type of situation than those in the purely physical area. For example, the effective current for an alternating voltage wave form is equal to the constant current required to produce the same heating in an electrical resistance. Such a definition gives an unequivocal relation between the two electric currents because a particular physical effect has been chosen for the relation. However, the value of the RBE (and presumably that of  $Q$  inferred from it at low dose and dose rate) depends upon the biological end point. While the value of  $Q$  for a given collision stopping power in water is currently assumed to apply to all tissues or organs, there are as yet insufficient data to determine whether this is actually true.

The proponents of this view point out also that a modifying factor,  $n$ , is used for radionuclides deposited in bone which is likely to be related "among other things to non-uniform spatial distribution of absorbed dose, the essentiality of the damaged tissue and the radiosensitivity of the particular type of irradiated cells".<sup>2</sup> Such a factor is not used for other tissues even though the tissues may be exposed to a similar non-uniform spatial distribution of absorbed dose. Incidentally, "absorbed dose" is used here because it is part of a quotation. A better term would be "specific energy imparted".

Thus, those who believe that the dose equivalent is a biophysical quantity, base their argument on the requirement for the specification of the biological end point for the determination of the modifying factors. To reinforce their argument, they point out that the value of the dose equivalent may be different in different organs or tissues even though the purely physical parameters are identical.

## A Risk

ICRP Publication 14- a report of two task groups to the parent commission- contains a discussion of how one might set relative permissible levels to the different tissues or organs of the body.<sup>3</sup> This scheme requires 1) a commonly agreed upon scale for "hurt or suffering" resulting from radiation damage of different tissues or organs and 2) the relative sensitivity of the various organs or tissues expressed as the ratio of doses required for equal "hurt or suffering".

If the permissible levels were based on such a scheme, the risk from a given dose equivalent in a particular organ would be proportional to this dose equivalent divided by the permissible level for that organ. While such a scheme for setting permissible levels is not now in effect, primarily because there is no generally accepted scale of "hurt or suffering", the dose equivalent to a given organ is presumed to be proportional to the radiation risk in that organ. However, for equal dose equivalents in different organs the risk will generally not be the same.

## Discussion

It is generally assumed that the dose equivalent to a particular organ or tissue is proportional to the risk of radiation induced injury for that organ. However, there is no commonly agreed upon scale of "effect" for all organs and tissues, so that the risk in any one organ may not be the same as for another organ even though their dose equivalents may be the same. For this reason, the dose equivalent equation cannot be considered generally to give an indication of the risk.

A fundamental assumption in the current use of the dose equivalent equation is that the value of the quality factor is independent of the organ under consideration and of the biological end point. Conceptually it primarily depends upon experimentally determined values of RBE. Thus, this factor is dimensionless. The magnitude of  $Q$  depends upon an agreed relation with the collision stopping power in water and is evaluated at the site of interest. The value of any modifying factor that might be included under  $N$  is also envisaged as being obtained from the ratio of the absorbed doses required to produce the same effect and is therefore also dimensionless. With these assumptions, and as the equation is currently used, it becomes a purely physical equation and the units of absorbed dose and dose equivalent may be the same.

One could think of  $H$  as either a weighted value of the absorbed dose or the absorbed dose of a reference radiation that would produce the same effect. If the latter is chosen, the proper term for  $H$  would be the "equivalent absorbed dose" of a reference radiation. It must be remembered, however, that the values of  $Q$  are merely estimates of the true value. Furthermore, biological variability has not been factored into these values. Thus, the same effect may not be presumed to be produced by an equivalent absorbed dose of a reference radiation. The ICRU, with ICRP concurrence, has agreed to retain the name "dose equivalent" for  $H$  and to treat the factors  $Q$  and  $N$  as dimensionless weighting factors.<sup>4</sup>

While both the dose equivalent and the absorbed dose may be given in either rads or joules per kilogram, the ICRU, with ICRP concurrence, has recommended that a specially named unit -the rem- be used for dose equivalent.<sup>4</sup> The redundancy resulting from the use of this special unit is deemed important by these commissions because the employment of the same unit for both absorbed dose and dose equivalent might lead to erroneous decisions in the matter of radiation safety. They point out that this redundancy conforms with the general safety policy in this technical area and is a continuation of established practice.



### References

1. International Commission on Radiation Units and Measurements, ICRU Report 19, Radiation Quantities and Units (International Commission on Radiation Units and Measurements, Washington, 1971)
2. International Commission on Radiological Protection, ICRP Publication 9, Recommendations of the International Commission on Radiological Protection, 1965. (Pergamon Press, Oxford, 1966)
3. International Commission on Radiological Protection, ICRP Publication 14, Radiosensitivity and Spatial Distribution of Dose. (Pergamon Press, Oxford, 1969)
4. International Commission on Radiation Units and Measurements, Supplement to ICRU Report 19, Dose Equivalent, (International Commission on Radiation Units and Measurements, Washington, 1973)

THE "INDEX QUANTITIES" OF THE ICRU  
Harald H. Rossi  
Radiological Research Laboratories, Department of  
Radiology, Columbia University, New York, N.Y.

Abstract

In its last report on radiation quantities and units (ICRU Report 19, 1971) the International Commission on Radiation Units and Measurements (ICRU) has defined the Absorbed Dose Index ( $D_1$ ) and the Dose Equivalent Index ( $H_1$ ) for employment in radiation protection. The explanatory statements provided by the Commission are rather brief and it appears that some misconceptions have arisen with regard to these quantities.

It will be the purpose of this paper to present the reasons for the introduction of these quantities and to examine their utility as well as their limitations.

-----

Radiation protection is based on a broad range of sciences and dedicated to the task of protecting man from a potentially dangerous agent that has a variety of forms and acts in manifold ways. In view of both the scientific nature and the practical importance of the subject it is essential that precise terminology be employed.

The interactions between the various ionizing radiations and the bodies of individuals exposed to them are usually quite complex and it is unavoidable that various approximations be made in practical health physics. This results in assessments that often have comparatively wide margins for error. This is quite understandable and as a rule also entirely acceptable, but substantial uncertainty in the numerical values of various quantities need not be accompanied by ambiguity concerning the quantities themselves.

"Dose" is perhaps the most important word in the vocabulary of radiation protection. Yet it has been, and still is, being employed in a variety of meanings. This may in part be due to the fact that the unmodified term has not been officially associated with a definite physical quantity and that the quantity under discussion is often evident from the context. Thus, a "dose" of 10 rads is obviously an absorbed dose, a "dose" of 10 rem is plainly meant to be a dose equivalent and a "dose" of 10 roentgens must be an exposure. There is perhaps some merit to the contention that in such instances the precise name of the quantity involved is redundant and that insistence that it be completely specified is pedantry. However, if SI units were to be used the name of the unit for dose and dose equivalent would be the same.

When absorbed dose and kerma are confused substantial errors can ensue. Thus, it has been customary to relate radiation epidemiological data to what are termed "dose", "air dose" or "first collision dose" when the quantity under consideration is in fact tissue kerma in free air. There have been evaluations of the "RBE" of neutrons relative to gamma rays (as in comparisons of leukemogenic effects in Hiroshima and Nagasaki) that are based on the ratios of the kermas of gamma radiation and neutrons that

produce equal effect. This is erroneous because in penetrating to the blood forming organs, neutrons are more strongly attenuated than gamma rays. Consequently the RBE must be higher than the kerma ratio.

There has been much confusion concerning the meaning of the term "dose" when it has been employed to characterize radiation fields "free in air." The absorbed dose at some point in air can only be energy absorbed per unit mass of air at that point. It should be noted that even this quantity need not be solely determined by the interaction between radiation and air since, for instance, structural material in the vicinity might be a source of secondary charged particles that reach the point of interest. It follows that the absorbed dose depends not only on the atomic composition of the material at the point of interest, but also on the composition and geometrical arrangement of the matter surrounding it. Attempts to define a "tissue dose in air" or "first collision dose" in terms of the absorbed dose in a "small" mass of tissue located at the point of interest are therefore ambiguous unless the magnitude of the mass is specified. In general as this tissue mass is increased the charged particle spectrum traversing its center will change from initial values characteristic of the surroundings to those characteristic of tissue, but in addition the primary radiation will also be attenuated. This subject is analyzed in some detail in Part II of ICRU Report 19. (1)

Kerma and exposure can often be effectively utilized to characterize ambient levels of indirectly ionizing radiations since they are defined in terms of a vanishing interacting mass. These quantities can be useful at radiation energies below a few MeV's because they are sometimes not too different from the maximum dose in a human body located at the point of interest. What is commonly termed the "dose" registered by an area monitor is frequently either tissue kerma or tissue kerma multiplied by some back-scattering factor which might again be uncertain, since its value must depend on the degree of isotropy of the radiation. At energies in excess of hundreds of MeV neither kerma nor exposure are meaningful because of the very large range of secondary charged particles and because of the complexity of interactions (particularly nuclear interactions).

It is of course, always possible to characterize the radiation field in terms of the distribution of particle fluence with respect to three variables which are (a) the nature of the particles, (b) the energy and (c) the direction; however, this information is usually very difficult to obtain and its practical applications are often involved.

In summary:

- 1) The only single valued (as opposed to spectral) quantities that can be meaningfully employed in radiation protection to characterize ambient radiation levels in "free air" are exposure for x- and gamma rays, and kerma for all indirectly ionizing radiations. The utility of these quantities tends to lessen at higher radiation energies, but even in the lower energy range further information (such as the angular distribution of the incident radiation) is desirable.
- 2) There exists no suitable quantity for directly ionizing radiations.

A few years ago the International Commission on Radiation Units and Measurements (ICRU) having recognized the lack of suitable quantities defined (1) what have become known as the "index" quantities, which meet the following requirements:

- 1) They are closely related to the maximum absorbed dose or the maximum dose equivalent in a human body if it were centered at the point of interest. In the majority of practical cases it is these quantities which need to be determined. There are, of course, instances where other doses or dose equivalents are of greater interest as for

instance in local irradiation of extremities.

2) Their value is unambiguously defined regardless of directional properties of the radiation field.

The absorbed dose index,  $D_1$ , and the dose equivalent index,  $H_1$ , are respectively the maximum values of absorbed dose and of dose equivalent in a 30 cm diameter sphere which is centered at the point of interest and consists of tissue equivalent material of specified composition. Except for highly unusual situations (such as traversal parallel to the body axis by a beam of extremely energetic heavy ions) the maximum dose or dose equivalent in a 30 cm sphere is not less than that in a human body at the same location.

The ICRU has omitted the definition of the time rate of change of these quantities but such an extension follows quite naturally. Thus, one might quantify the hazards to personnel in a fallout field in terms of a map containing information on the absorbed dose index rate. The statement that at the control console of an accelerator the dose equivalent index rate is X millirem per hour implies that if an operator were to be located there, a dose equivalent rate of X millirem per hour will not be exceeded in any part of his body. If the accelerator building is surrounded by a fence that is located along a line which is defined by an annual value of the dose equivalent index of 500 millirem, compliance with current radiation protection regulations is ensured regardless of occupancy factors.

In situations illustrated by these examples the term "dose" has been employed with the same meaning as that of the index quantities. However, as explained above it has also been used to indicate tissue kerma in free air or other less clearly defined concepts and this has led to uncertainties. If one is dealing with a unidirectional beam of 1 MeV neutrons the absorbed dose index is 50% larger than tissue kerma in free air. In the case of isotropic incidence of low energy  $\gamma$  radiation the magnitude of either index can be less than half as large as the kerma.

Because they are defined in terms of maximum values in an extended region centered at the point of interest, the index quantities have somewhat unusual characteristics. For example, the maximum absorbed dose in the sphere will usually be near its surface rather than at its center and outside of the limits of a sharply collimated beam the value of either index can be large at positions (i.e. locations of the center of the sphere) where there is very little radiation. Also, the index quantities are not defined for locations that are less than 15 cm from solid structures (e.g. a shielding wall). The reason is, of course, that there would be no room for a real or conceptual sphere of tissue equivalent material. However, placement of a human body in such locations would be similarly impossible. Finally, it should be pointed out that for mixtures of different radiations the maximum absorbed doses or maximum dose equivalents occur at different locations in the sphere with the result that summation of the indexes for the components of the mixture results in a conservative upper limit. It should be stressed that the objective of defining the index quantities was to provide a clear and unambiguous specification of the radiation field for practical purposes. They are neither intended nor suitable as a basis for the formulation of radiation protection standards.

Although  $D_1$  and  $H_1$  are in principle determined by exploration of a spherical phantom of 30 cm diameter, such a procedure should rarely be necessary in practice. Information on the radiation field will usually be adequate to assess the value of these quantities on the basis of measurements involving conventional ionization chambers or other dosimeters with reasonable corrections made for such factors as backscattering or wall attenuation.

It is my hope that this presentation has adequately indicated the need for the formulation of the index quantities and that they will be accepted as useful adjuncts to the other quantities employed in radiation protection.

<sup>1</sup>Radiation Quantities and Units. Report 19 of the International Commission on Radiation Units and Measurements.

## LES UNITES SI DANS LES RAYONNEMENTS IONISANTS

A. ALLISY

Professeur au Conservatoire National des Arts et Métiers, Paris,  
et Bureau International des Poids et Mesures,  
Pavillon de Breteuil, 92310 SEVRES, France

### Abstract

The present positions of the Comité International des Poids et Mesures and the International Commission on Radiation Units and Measurements regarding the use of the International System of Units (SI) are given. Physical quantities and units are briefly considered with special reference to coherence. A few simple rules are given concerning the symbols and units.

### Introduction

On se propose dans cette introduction de résumer brièvement les positions officielles sur l'utilisation du Système International d'Unités (SI) dans le domaine des rayonnements ionisants.

#### Les tâches de l'I.C.R.U.

Depuis sa création en 1925 sous les auspices du Congrès International de Radiologie, la Commission Internationale des Unités et Mesures de Rayonnements (I.C.R.U.) a, entre autres, la responsabilité du développement d'un système de grandeurs et d'unités dans le domaine des rayonnements ionisants. L'I.C.R.U. s'est toujours efforcée de produire un tel système de manière qu'il puisse avoir le champ d'application le plus large possible.

Lorsqu'on parcourt les rapports <sup>1 à 7</sup> publiés entre 1950 et 1971, on ne peut pas manquer de noter la tendance de plus en plus nette à utiliser en dosimétrie les grandeurs physiques et leur formalisme, en même temps qu'une rigueur de plus en plus grande était introduite. Dans le rapport de 1962 <sup>5</sup> on lit: "Le but du présent réexamen des concepts à utiliser en dosimétrie était principalement de renforcer le degré de rigueur. S'il est nécessaire, dans ce processus, de donner aux définitions une forme plus stricte pour éliminer les ambiguïtés prévisibles, ceci doit être accepté".

## Historique succinct du Système International d'Unités (SI)

En 1948<sup>8</sup>, la 9<sup>e</sup> Conférence Générale des Poids et Mesures (C.G.P.M.) a chargé le Comité International des Poids et Mesures (C.I.P.M.) "d'étudier l'établissement d'une réglementation complète des unités de mesure", "d'ouvrir à cet effet une enquête officielle sur l'opinion des milieux scientifiques, techniques et pédagogiques de tous les pays" et "d'émettre des recommandations concernant l'établissement d'un système pratique d'unités de mesure, susceptible d'être adopté par tous les pays signataires de la Convention du Mètre".

La 10<sup>e</sup> C.G.P.M. (1954)<sup>8</sup> et la 14<sup>e</sup> C.G.P.M. (1971)<sup>8</sup> décidèrent d'adopter les sept unités de base de ce "système pratique d'unités". La 11<sup>e</sup> C.G.P.M. (1960)<sup>8</sup> adopta le nom Système International d'Unités, avec l'abréviation internationale SI, et donna des règles pour les préfixes, les unités dérivées et les unités supplémentaires, et d'autres indications, établissant ainsi une réglementation d'ensemble pour les unités de mesure.

En 1964<sup>9</sup>, le C.I.P.M. transforma sa "Commission du Système d'Unités" en Comité Consultatif des Unités (C.C.U.) dont le rôle le plus important est "d'assurer une meilleure coordination des travaux des diverses organisations internationales travaillant dans le domaine des définitions des unités et de leurs symboles".

En 1970, puis en 1973<sup>8</sup>, le Bureau International des Poids et Mesures (B.I.P.M.) a publié un document intitulé "Le Système International d'Unités (SI)", dans lequel est rassemblé dans un ordre systématique le contenu des Résolutions et des Recommandations de la C.G.P.M. et du C.I.P.M. concernant le SI. On y trouve également des commentaires explicatifs et des règles d'utilisation pratique extraites des Recommandations d'usage général adoptées par l'Organisation Internationale de Normalisation (I.S.O.). Ce texte a été traduit en plusieurs langues.

### L'I.C.R.U. et le SI

L'I.C.R.U., qui est un des membres du C.C.U., a participé aux réunions de ce Comité en 1967<sup>10</sup>, 1969<sup>11</sup> et 1971<sup>12</sup>. En 1968<sup>6</sup>, elle a adopté le SI pour toutes ses publications, mais a maintenu, de même qu'elle le fait aujourd'hui, les unités spéciales rad, röntgen et curie. Dès 1962<sup>5</sup>, la Commission avait estimé que l'addition d'autres unités spéciales n'était pas désirable dans le domaine de la dosimétrie des rayonnements.

En 1969<sup>13</sup>, le C.I.P.M., suivant une proposition du C.C.U. de 1969<sup>11</sup>, a précisé le classement des unités en dehors du SI; le rad, le röntgen et le curie sont alors mis dans la classe B ainsi définie: "Unités en dehors du SI dont l'emploi pourra être évité, mais qu'il semble préférable de maintenir temporairement pour qu'elles soient employées conjointement avec les unités du SI, en raison de la force des usages existants". Le vocable "temporairement" n'est accompagné d'aucune date.

Compte tenu des avantages de l'utilisation quasi universelle d'un même système d'unités, l'I.C.R.U. a chargé en 1973<sup>14</sup> son secrétaire scientifique, K. Lidén, d'inviter, par une lettre envoyée aux revues spécialisées, les scientifiques concernés à envoyer leurs commentaires et suggestions sur le problème du remplacement des unités spéciales rad, röntgen et curie, par des unités dérivées SI.

## Grandeurs physiques, Unités, Cohérence

Afin d'apporter quelques éléments d'appréciation sur les avantages du SI, on donne ici de brefs rappels sur les grandeurs physiques et les unités, en insistant sur la notion de cohérence. Le lecteur intéressé par ce sujet consultera avec profit R. Fleischmann<sup>15</sup>, U. Stille<sup>16</sup> et J. de Boer<sup>17</sup>.

L'ensemble des grandeurs physiques peut être subdivisé en sous-ensembles de grandeurs physiques de même nature. Ainsi, par exemple, le diamètre intérieur d'une chambre d'ionisation sphérique, la distance interréticulaire d'un cristal ou la longueur d'onde dans le vide d'une radiation définie sont des grandeurs physiques de même nature et appartiennent au sous-ensemble des longueurs.

Les physiciens expérimentalistes qui effectuent des mesures sur des objets physiques, et les théoriciens qui considèrent les grandeurs physiques comme des objets mathématiques qu'ils font correspondre aux objets physiques, expriment les grandeurs physiques sous la forme suivante:

$$\boxed{\text{Grandeur physique} = \text{valeur numérique} \times \text{unité}}$$

où la valeur numérique est un nombre réel et l'unité une grandeur physique de même nature que celle considérée et dont la valeur numérique est 1 par convention.

Une grandeur physique est indépendante des unités choisies: la longueur d'une table ne varie pas selon qu'on l'exprime en mètres, en inches ou en mille marins; seule la valeur numérique change.

Soit  $G$  une grandeur physique,  $\{G\}_a$  et  $\{G\}_b$  ses valeurs numériques dans deux systèmes d'unités notés "a" et "b",  $[G]_a$  et  $[G]_b$  les unités de cette grandeur physique dans les systèmes respectifs. L'invariance de la grandeur physique dans tout changement d'unité s'écrit alors

$$G = \{G\}_a \cdot [G]_a = \{G\}_b \cdot [G]_b . \quad (1)$$

La relation (1) permet de résoudre les problèmes de changement d'unités si l'on connaît le rapport entre deux unités des systèmes "a" et "b" donné par une équation aux unités

$$[G]_b = \alpha [G]_a , \quad \text{où } \alpha \text{ est un nombre réel.} \quad (2)$$

On a, en effet, d'après (1) et (2)

$$\{G\}_b = \{G\}_a \frac{[G]_a}{[G]_b} = \frac{1}{\alpha} \{G\}_a ,$$

qui donne la valeur numérique de  $G$  dans le système "b" en fonction de celle dans le système "a" et de  $\alpha$ .

Exemple - Envisageons les trois grandeurs physiques quantité d'électricité  $Q$ , masse  $m$  et exposition  $X$ , et deux systèmes d'unités définis par

$$\begin{aligned} [Q]_a &= C & [Q]_b &= C \\ [m]_a &= \text{kg} & [m]_b &= \text{kg} \\ [X]_a &= C \cdot \text{kg}^{-1} & [X]_b &= R \end{aligned} \quad (3)$$

L'équation aux unités  $[X]_b = \alpha [X]_a$  s'écrit alors

$$1 R = 2,58 \cdot 10^{-4} C \cdot \text{kg}^{-1} \quad (\text{exactement}),$$

donc  $\alpha = 2,58 \cdot 10^{-4}$ .

Une exposition  $X = 10^{-2} C \cdot \text{kg}^{-1}$ , soit  $\{X\}_a = 10^{-2}$ , aura dans le système "b" la valeur numérique

$$\{X\}_b = \frac{10^{-2}}{2,58 \cdot 10^{-4}} \approx 38,76,$$

donc

$$X \approx 38,76 R.$$

### Les équations aux grandeurs

La structure algébrique du calcul sur les grandeurs physiques a été analysée récemment par R. Fleischmann<sup>15</sup> et W. Quade<sup>18</sup>. Rappelons quelques règles élémentaires:

- N'importe quel élément, différent de 0, de l'ensemble des grandeurs physiques de même nature peut être utilisé comme unité.
- On ne peut additionner ou soustraire que des grandeurs physiques de même nature.
- La multiplication (ou la division) de grandeurs physiques de même nature ou de natures différentes donne une nouvelle grandeur physique.

Les relations entre grandeurs physiques que l'on peut obtenir conformément à la règle (c) s'appellent des équations aux grandeurs. Elles ne comprennent que des grandeurs physiques et sont donc invariantes dans tout changement d'unités. Ces équations sont, en particulier, utilisées pour définir des nouvelles grandeurs physiques à partir de grandeurs précédemment établies. Par exemple, si  $\epsilon$  est l'énergie communiquée par des rayonnements ionisants à la masse  $m$  de matière,



on définit l'énergie massique par la relation  $Z = \epsilon/m$ . On conçoit qu'en procédant de cette manière il est possible de définir des grandeurs physiques dérivées à partir d'un noyau restreint de grandeurs physiques de base.

Le choix des grandeurs physiques de base est en principe arbitraire, mais il est dicté par des considérations de commodité et de précision des mesures. Le nombre de grandeurs physiques de base est égal à la différence entre le nombre total de grandeurs physiques envisagées et le nombre d'équations indépendantes reliant ces grandeurs. Ces équations sont soit des équations de définition de nouvelles grandeurs, soit des relations entre grandeurs correspondant à des lois expérimentales de la physique.

### Systèmes d'unités, Cohérence

A l'ensemble des grandeurs physiques formé des grandeurs de base et des grandeurs dérivées on fait correspondre un ensemble d'unités (qui sont aussi des grandeurs physiques).

Les unités de base correspondent aux grandeurs physiques de base. A chaque grandeur dérivée correspond une unité dérivée qui est définie, à une constante réelle près, par une relation analogue à l'équation aux grandeurs définissant la grandeur dérivée.

Exemple - L'équation aux grandeurs définissant l'exposition X est

$$X = \frac{Q}{m} \quad . \quad (4)$$

En reprenant les systèmes d'unités "a" et "b" définis par les relations (3), on a

$$[X]_a = \frac{[Q]_a}{[m]_a} \quad \text{et} \quad [X]_b = \alpha \frac{[Q]_b}{[m]_b} \quad .$$

Les deux unités  $[X]_a$  et  $[X]_b$  sont définies correctement. L'unité  $[X]_a$  pour laquelle la constante réelle est égale à l'unité est appelée l'unité cohérente d'exposition dans le système "a".

La cohérence d'un système d'unités est une qualité primordiale qui simplifie les calculs et soulage la mémoire qui n'est plus encombrée de coefficients numériques inutiles. Le tableau 1 met en évidence ces avantages en montrant que dans un système d'unités cohérent les équations aux valeurs numériques et les équations aux unités sont identiques à l'équation aux grandeurs correspondantes.

Tableau 1

Systèmes d'unités	"a" cohérent	"b" non cohérent
Equation aux grandeurs	$X = \frac{Q}{m}$	$X = \frac{Q}{m}$
Equation aux grandeurs explicitée	$\{X\}_a [X]_a = \frac{\{Q\}_a [Q]_a}{\{m\}_a [m]_a}$	$\{X\}_b [X]_b = \frac{\{Q\}_b [Q]_b}{\{m\}_b [m]_b}$
Equations aux unités	$[X]_a = \frac{[Q]_a}{[m]_a}$	$[X]_b = \alpha \frac{[Q]_b}{[m]_b}$ $1 R = 2,58 \cdot 10^{-4} C \cdot kg^{-1}$
Equations aux valeurs numériques	$\{X\}_a = \frac{\{Q\}_a}{\{m\}_a}$	$\{X\}_b = \frac{\{Q\}_b}{\{m\}_b} \cdot \frac{[Q]_b}{[m]_b [X]_b}$ $= \frac{\{Q\}_b}{\{m\}_b} \cdot \frac{1}{\alpha}$

## Le SI

Le Système International d'unités<sup>8</sup> est un système cohérent fondé sur les sept unités de base des grandeurs physiques données dans le tableau 2.

Tableau 2

<u>Grandeur physique</u>	<u>Nom de l'unité</u>	<u>Symbole</u>
longueur	mètre	m
masse	kilogramme	kg
temps	seconde	s
intensité de courant électrique	ampère	A
température thermodynamique	kelvin	K
quantité de matière	mole	mol
intensité lumineuse	candela	cd

Les unités dérivées sont données à partir des unités de base par des expressions algébriques en utilisant les symboles mathématiques de multiplication et de division. Plusieurs de ces unités dérivées ont reçu un nom spécial et un symbole particulier, lesquels peuvent être utilisés à leur tour pour exprimer d'autres unités dérivées d'une façon plus simple qu'à partir des unités de base. Dans ce cas, une unité dérivée peut s'exprimer de plusieurs façons équivalentes en utilisant des noms d'unités de base et des noms spéciaux d'unités dérivées, mais il est clair qu'elle ne s'exprime d'une manière, et d'une seule, qu'en fonction des unités de base.

Exemple - La grandeur physique puissance qui s'exprime en unités de base mètre carré kilogramme par seconde cube a reçu le nom spécial watt, mais peut aussi s'exprimer en joule par seconde ou en volt ampère. On utilise en général la dénomination la plus commode. Ainsi, un débit de dose absorbée peut s'exprimer en joule par kilogramme seconde, de manière à faire apparaître le temps explicitement.

Il est possible de former des multiples et des sous-multiples décimaux des unités SI en utilisant des préfixes dont les noms et les symboles ont été fixés pour quelques facteurs allant de  $10^{-18}$  à  $10^{12}$ . Il est bien clair que ces multiples et sous-multiples ne font pas partie du système cohérent puisqu'ils introduisent des facteurs numériques différents de l'unité.

## Qu'implique l'introduction du SI?

Il est clair que l'utilisation des unités SI entraînera un changement substantiel dans les valeurs numériques des grandeurs physiques utilisées dans la pratique journalière<sup>14</sup>. Etant donné qu'en radiothérapie, en radiodiagnostic et en radioprotection des erreurs dues à une modification des unités sont susceptibles de mettre en danger la vie d'êtres humains, il est indispensable de prévoir une période de transition pendant laquelle les deux systèmes d'unités seront utilisés conjointement. La décision du C.I.P.M. de 1969<sup>14</sup> de maintenir temporairement le rad, le röntgen et le curie est donc une décision sage. Il faut néanmoins noter que les rapports entre ces unités spéciales et les unités SI sont très différents de l'unité, ce qui amoindrit les risques d'erreurs.

L'introduction des unités SI entraîne logiquement l'adoption de règles de "grammaire" simples (mais que l'on a intérêt à maintenir strictes)<sup>8</sup>. En voici deux exemples:

- Les symboles des unités sont définis de manière rigoureuse; lors de leur utilisation, ils ne doivent être affectés d'aucun qualificatif supplémentaire.

Commentaires: Dans l'expression d'une grandeur physique une erreur sur le symbole de l'unité est aussi grave qu'une erreur sur la valeur numérique.

Une unité sert à spécifier la grandeur physique (donc sa nature) dont la valeur numérique est 1 et elle ne sert qu'à cela. S'il est nécessaire de préciser des situations particulières par des qualificatifs appropriés, ces derniers seront affectés au nom de la grandeur physique. On dira, par exemple, que la dose absorbée dans le tissu est  $x \text{ J} \cdot \text{kg}^{-1}$  ou  $y \text{ rad}$ , et non pas la dose absorbée est  $x \text{ "J} \cdot \text{kg}^{-1} \text{ tissu"}$  ou  $y \text{ "rad tissu"}$ . Souvent, d'ailleurs, le nom même de la grandeur physique implique une série de conditions conformément à une définition. Ainsi lorsque nous parlons d'une exposition de  $x \text{ C} \cdot \text{kg}^{-1}$ , nous savons qu'il s'agit d'une quantité d'électricité massique libérée dans l'air par des photons et qui doit être mesurée dans des conditions très précises. L'idée que toutes ces connotations sont comprises dans le nom de l'unité röntgen est sans doute encore assez répandue, mais cette manière de voir n'est pas conforme aux règles généralement observées en physique.

- Les symboles des unités ne sont pas suivis d'un point; ils restent invariables ou pluriel. Lorsqu'un préfixe est placé devant un symbole, la combinaison du préfixe et du symbole doit être considérée comme un nouveau symbole qui peut être élevé à une puissance positive ou négative sans utiliser de parenthèses.

L'utilisation de préfixes doubles doit être évitée.

Exemples -

$$1 \text{ cm}^3 = [10^{-2} \text{ m}]^3 = 10^{-6} \text{ m}^3$$

$$1 \text{ Ms}^{-1} = [10^6 \text{ s}]^{-1} = \frac{1}{10^6 \text{ s}}$$

## Conclusion

On a tenté de mettre en évidence quelques avantages qui résultent de l'utilisation du SI dans le domaine des rayonnements ionisants. Ces avantages découlent principalement de la cohérence du système. Mais quel physicien peut rester insensible au fait que le SI est en train de devenir un langage universel utilisé et compris par tous? Parlant du SI, J. Terrien dit en 1965<sup>19</sup>: "Bien sûr, ce système peut être critiqué comme toute création humaine; sa supériorité ne peut être prouvée par l'expérience puisqu'il est avant tout affaire de convention, et la qualité primordiale d'un langage conventionnel c'est d'être universellement agréé, même s'il n'est pas parfait. En fait, ce système a reçu l'approbation d'une majorité importante se fondant sur des raisons solides. Il épargne beaucoup de temps et d'efforts à l'étudiant dans la conversion des unités d'un système à l'autre et évite des erreurs attachées à ce genre de calculs dans la pratique journalière".

## Références

- 1 Recommendations of the International Commission on Radiological Protection and of the International Commission on Radiological Units, National Bureau of Standards Handbook 47 (1950).
- 2 Recommendations of the International Commission on Radiological Units, 1953, Brit. Journ. Rad. XXVII, 316, 243 (1954).
- 3 Report of the International Commission on Radiological Units and Measurements, National Bureau of Standards Handbook 62 (1956).
- 4 Report of the International Commission on Radiological Units and Measurements, National Bureau of Standards Handbook 78 (1959).
- 5 Radiation Quantities and Units, ICRU Report 10a, National Bureau of Standards Handbook 84 (1962).
- 6 Radiation Quantities and Units, ICRU Report 11 (1968), ICRU Publications, P.O. Box 30165, Washington, D.C. 20014.
- 7 Radiation Quantities and Units, ICRU Report 19 (1971), ICRU Publications, P.O. Box 30165, Washington, D.C. 20014.
- 8 Le Système International d'Unités (SI), 2<sup>e</sup> édition (1973), Offilib, 48 rue Gay-Lussac, 75005 Paris.
- 9 Comité International des Poids et Mesures, Procès-Verbaux, 53<sup>e</sup> session, 32 (1964).
- 10 Comité Consultatif des Unités, 1<sup>re</sup> session (1967).

- 11 Comité Consultatif des Unités, 2<sup>e</sup> session (1969).
- 12 Comité Consultatif des Unités, 3<sup>e</sup> session (1971).
- 13 Comité International des Poids et Mesures, Procès-Verbaux, 58<sup>e</sup> session, 37 (1969).
- 14 K. LIDÉN, SI Units in Radiology and Radiation Measurement, Health Phys. 25, 2, 199 (1973).
- 15 R. FLEISCHMANN, Physikalisches Begriffssystem und Dimensionen, Physikal. Blätter 9, 7, 301 (1953).
- 16 U. STILLE, Messen und Rechnen in der Physik, Vieweg, Braunschweig (1961).
- 17 J. DE BOER, Some General Aspects of the International System of Units, De Ingenieur 27, A 550 (1970).
- 18 W. QUADE, Über die algebraische Struktur des Grössenkalküls der Physik, Abh. Braunschweig Wiss. Ges. 13, 24 (1961).
- 19 J. TERRIEN, Scientific Metrology on the International Plane and the Bureau International des Poids et Mesures, Metrologia 1, 1, 15 (1965).

# RADIOECOLOGY

## DISTRIBUTION OF ENVIRONMENTAL PLUTONIUM IN THE TRINITY SITE ECOSYSTEM AFTER 27 YEARS

Thomas E. Hakonson and LaMar J. Johnson  
Los Alamos Scientific Laboratory  
Los Alamos, New Mexico

### Abstract

The results are presented for a radioecological survey of the Trinity Site environs, where the world's first (July 1945) atomic bomb was detonated. The temporal behavior of the low environmental levels of the plutonium produced by this detonation are discussed. The data from this study were compared with similar data obtained in the Trinity Site environs nearly 20 years ago. The major change which was observed was an increased migration of Pu into the soils. Concentrations of Pu in vegetation and rodents were too low to make valid comparisons.

### Introduction

An ecological investigation of plutonium was initiated in the fallout pathway of Trinity, the first nuclear detonation, which occurred on July 16, 1945 in southern New Mexico. Trinity Site was especially interesting as a study area because of the "aged" nature of the radioactive debris distributed in the area. In addition, it was of interest to gather ecological data on plutonium in the xeric Trinity Site environs to compare with similar data being gathered in several semi-mesic ecosystems at the Los Alamos Scientific Laboratory in northern New Mexico.<sup>1</sup>

The data presented in this paper were obtained from samples gathered during one sampling period in the Trinity Site environs on September 27-28, 1972. The primary objectives of this effort were to survey the plutonium content of a few ecosystem components and to measure the field gamma radiation intensity as a function of distance from Ground Zero (GZ) some 27 years after the detonation to facilitate the design of more intensive studies.

### Methods and Materials

Trinity Site, a fenced area immediately around GZ, is located in the semi-arid northern portion of the Tularosa Basin about 40 miles SW of Socorro, New Mexico, on the White Sands Missile Range (Fig. 1). A general description of the physiography, climate, vegetation and mammals of the area around Trinity Site can be found in various references.<sup>2,3,4</sup>

A transect was established along the reported fallout pathway of the nuclear debris from the detonation (Fig. 1), utilizing maps constructed by University of California scientists in 1948.<sup>5</sup> Nine sampling locations were established on the transect, two in the GZ area, and the remainder at 8 km increments to a distance of about 56 km from GZ.

One soil core was taken at each location with a disposable 30 cm section of 2.4 cm diameter polyvinyl chloride pipe. The pipe and contained core from

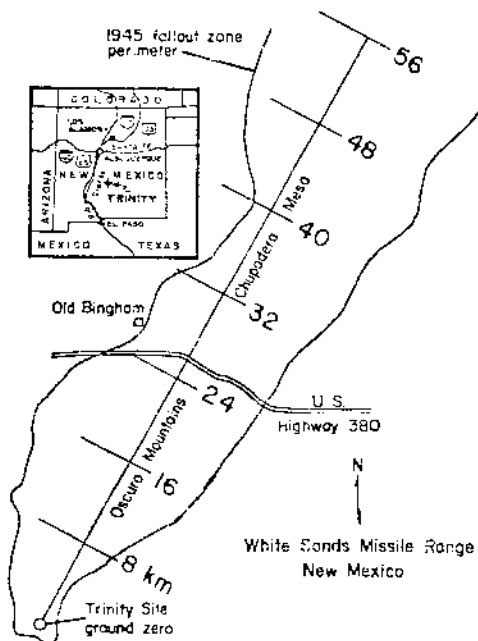


Fig. 1. Sampling transect utilized for the radioecological resurvey of Trinity.

each station was sealed in a plastic bag, frozen on return to the laboratory and sectioned into a 0-2.5 cm, 2.5-7.5 cm and 7.5-30 cm segment.

A sample of the most abundant forb, grass, and shrub/tree species was collected where possible and was individually sealed in plastic bags. Samples included the above-ground portions of the grasses and forbs and the terminal leaves and stems of the shrub/tree species. Dust on the plant surfaces was not removed prior to analysis.

Rodents were collected with peanut butter baited snap traps and were bagged and frozen for later dissection. Tissues analyzed for plutonium included lungs, liver, hide, and carcass (skeleton and skeletal muscle). Care was taken during the dissection to avoid cross-contaminating the soft tissues with hair from the pelt.

Trinitite, the fused soil material formed by the intense heat produced by the detonation, was also collected to determine its plutonium content and to identify the gamma emitters present.

Analytical procedures for plutonium included a combination of wet-dry ashing techniques utilizing a muffle furnace and  $\text{HNO}_3$ -HF solutions followed by ion exchange column separation of plutonium, electrodeposition and alpha ray spectroscopy for both  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$ . The minimum sensitivity of the alpha counting system based upon background counts during a 24 hour period was 0.03 pCi  $^{238}\text{Pu}$  or  $^{239}\text{Pu}$ /sample ( $\alpha = 0.05$ ).

All soil and vegetation samples contained sufficient Pu and/or were of sufficient mass to reduce the relative counting standard deviation on each sample to less than 25 percent (1  $\sigma$ ). However, the generally low Pu content and small mass of certain rodent tissues resulted in relative counting standard deviations of as much as 100 percent. The standard errors associated with the rodent data presented later, reflect this fact.



Direct measurement of radiation in the field was accomplished with a Ludlum Model 12S Count Rate Meter, which utilizes a NaI (Tl) scintillation detector, for in situ environmental gamma radiation measurements. This instrument's readout was calibrated to give a proper "µR/h" reading with <sup>60</sup>Co gamma rays. During the survey, the instrument was held at about 3 feet above the ground surface and the observed rate noted and recorded at the respective locations. Because of an inherent photon energy-dependent response, all readings obtained with the Ludlum were normalized using 10 Los Alamos Scientific Laboratory environmental radiation dosimetry stations which utilize LiF thermoluminescent dosimeters (TLD) for background radiation measurements.<sup>6</sup> Dosimetric values obtained from these TLD materials have been shown to be essentially independent of radiation energy and, therefore, provided a basis for the correction or normalization of the Ludlum Model 12S meter readings. The normalization assumed uniform photon spectral distribution. The observed average ratio of TLD-determined exposure rates to the survey meter measurements was 0.70.

### Results and Discussion

The Pu content (<sup>239</sup>Pu and <sup>238</sup>Pu) of all sample types as a function of distance from GZ is presented in Table 1.

Table 1. The <sup>238-239</sup>Pu content of some ecosystem components collected in the fallout zone of the Trinity detonation.

	Kilometers From Ground Zero (GZ) Along Fallout Pathway								
	GZ	0.1	6.1	16.1	26.1	32.2	40.3	48.3	56.4
<b>Soils</b> (fCi/g dry)									
0-2.5 cm	254812	36	434	311	125	295	668	278	1442
2.5-7.5 cm	262720	36	0	331	746	116	344	20	89
7.5-30 cm	61858	23	6.6	213	175	3.4	62	10	35
<b>Vegetation</b> (fCi/g wet) *									
Grasses	768	169	73	34	5.1	15	19	11	28
Forbs				3.0	0.60	2.5	4.7	5.4	26
Shrubs/trees	6.2	2.4	9.5		1.9	4.1	6.5	0.60	2.2
<b>Rodents</b> (fCi/g wet) *									
Liver		12 (11)**	4.4 (2.2)	1.3 (0.8)	1.7	2.3 (1.5)	4.5 (2.7)	6.2	
Lungs		25 (19)	11 (8.1)	4.1 (3.9)	3.3	48 (27)	50 (45)	0	
Hide		4.8 (0.96)	5.6 (2.3)	2.0 (2.0)	0.75	3.4 (1.2)	2.8 (1.1)	3.8	
Carcass ***		3.2 (1.2)	15 (8.0)	4.2 (3.0)	0.84	30 (29)	4.7 (1.6)	2.5	
No. of samples		4	5	3	1	5	4	1	

\* Species comprising the vegetation and rodent samples are given in the text.

\*\* Parenthetic value represents the standard error of the determination.

$$\text{Standard Error} = \frac{\text{standard deviation}}{\sqrt{\text{No. of samples}}}$$

\*\*\* Carcass includes skeleton and skeletal muscle.

The Pu data for vegetation (fCi/g wet) were summarized according to the type of plant (grass, forb or shrub/tree) to provide some basis for viewing Pu concentration gradients with distances from GZ since none of the plants were found at every sampling station. Grass species included Tridens pulchellus, Sporobolus Nealleyi and Bouteloua eriopoda while the forb category included Mirabilis multiflora, Conyza Coulteri, Dithyrea Wislizeni, Aphanostephus humilis and Melilotus albus. All the grass and forb species were generally less than 60 cm tall; the shrub/tree species which included Atriplex canescena,

Larrea tridentata, Lycium Andersonii, Juniperus monosperma and Rhus macrophylla were generally greater than 60 cm tall.

The Pu data for rodent tissues (fCi/g wet) were also summarized without regard to species because the inadequate number of samples did not permit a species comparison and because the species composition of the catch changed with distance from GZ. Species caught included Perognathus flavus, Citellus spilosoma, Peromyscus maniculatus, P. truei, Onychomys leucogaster, Neotoma mexicana and Dipodomys ordi.

The Pu concentrations in many of the soil core segments (Table 1) were significantly above background. Levels in GZ soils were as much as  $10^4$  times higher than the 10-100 fCi Pu/g which has been reported for several New Mexico area soils.<sup>7</sup> A maximum of about 260,000 fCi Pu/g was observed at GZ in both the 0-2.5 cm and the 2.5-7.5 cm core segments. The maximum concentration in non-GZ soil (1442 fCi/g) was measured in the 0-2.5 cm segment from the 56.4 km sampling station.

The Pu data for soils from the GZ and 0.1 km stations cannot be compared with the data for the remainder of the transect because the area around GZ was mechanically disturbed shortly after the detonation in an effort to reduce surface radionuclide contamination. Ground Zero, for example, was covered with at least 15 cm of uncontaminated soil and the area around the 0.1 km station was scraped to remove the Trinitite lying on the ground surface. The high Pu concentration in the 0-2.5 cm segment of the GZ soil sample (Table 1) indicates either 1) the overburdening was not successful; 2) that the covered Pu had migrated to the soil surface; or 3) that the overburden had blown away over the last 27 years, thus exposing the contaminated soil.

The Pu concentrations in the 0-2.5 cm segment generally increased toward the distal end of the sampling transect and reached a maximum at the 56.4 km station. Olafson et al.<sup>8</sup>, during efforts to map the fallout zone from Trinity also noted that the highest Pu concentration in soils, vegetation and small mammals outside the CZ area occurred about 45 km from GZ.

The vertical distribution of Pu was relatively uniform in most of the core samples from GZ to the 24.1 km station. This suggested that Pu which was initially deposited on the soil surface as much as 27 years ago had migrated at least 30 cm into the soil profile. On the other hand, the Pu in soils from distances greater than 24.1 km was increasingly concentrated in the upper 2.5 cm. Olafson et al.<sup>8</sup> and Olafson and Larson<sup>9</sup> reported that the Pu in Trinity area soils about 20 years ago was almost exclusively confined to the top 2.5 cm of soil.

Many factors could account for a difference in the rate of vertical migration of Pu in soils, including differences in the chemical and physical form of the Pu and/or differences in the chemical, physical, and biological makeup of the environment.<sup>10</sup>

The Pu concentrations in grasses were consistently elevated with respect to similar measurements in other areas of New Mexico.<sup>1,9</sup> On the other hand, the Pu concentrations in forb and shrub/tree samples were generally indistinguishable from worldwide fallout Pu levels in New Mexico vegetation which measure about 1-5 fCi/g wet sample.<sup>1,7</sup> The Pu data for grasses as a function of distance from GZ generally followed the pattern which was observed for the 0-2.5 cm soil core segment (Table 1). The maximum Pu concentration in grasses (768 fCi/g wet) was observed at GZ and decreased to a minimum of 5.1 fCi/g at the 24.1 km station. Pu concentrations in grasses then generally increases with distance.

The Pu data for rodent tissues in Table 1 show a considerable variability. Sources of this variability would include among other things, species differences, the low Pu content of the tissues yielding generally poor counting statistics, and an insufficient number of samples.

In general, rodent lungs had the highest mean Pu concentrations and exhibited a pattern with distance from GZ that was similar to the 0-2.5 cm layer of soil and the grass. Lung deposition of Pu suggested that resuspension of soil may be an important mechanism in the biological redistribution of Pu. Other investigators have noted high lung concentrations in small free-roaming mammals.<sup>11,12</sup>

The activity ratios  $^{239}\text{Pu}/^{238}\text{Pu}$  for all sample types are summarized in Table 2. The ratios for the 0-2.5 cm and 2.5-7.5 cm core segments averaged 19 and 18, respectively, while the 7.5-30 cm segment averaged 9. The mean values for vegetation were about 8-12 and about 0.5-2 for rodent tissues. The significance of the decreasing  $^{239}\text{Pu}/^{238}\text{Pu}$  ratio from soils to vegetation to rodent tissues is not clear at this time. It may indicate that  $^{238}\text{Pu}$  in the Trinity environs is more mobile than  $^{239}\text{Pu}$ .

Table 2. The  $^{239}\text{Pu}/^{238}\text{Pu}$  ratio in some ecosystem components collected in the fallout zone of the Trinity detonation.

Type Sample	$^{239}\text{Pu}/^{238}\text{Pu}$		No. Samples
	X	S.E.*	
<u>Soils</u>			
0-2.5 cm	19	3.2	9
2.5-7.5 cm	18	1.7	7
Remainder	9.0	2.3	6
<u>Vegetation</u>			
Grasses	12	2.0	13
Forbs	7.6	1.9	10
Shrubs	8.0	1.6	9
<u>Rodents</u>			
Liver	0.44	0.10	5
Lungs	1.0	0.28	8
Hide	1.8	0.89	16
Carcass	1.7	0.65	20

\* S.E. =  $\frac{\text{standard deviation}}{\sqrt{\text{number of samples}}}$

The Pu content of three samples of Trinitite from GZ measured 3.2 nCi/g, 1.5 nCi/g and 1.2 nCi/g with an average  $^{239}\text{Pu}/^{238}\text{Pu}$  ratio of  $21 \pm 0.8$  ( $1\sigma$ ). These Pu concentrations are about an order of magnitude higher than the Pu concentrations in GZ soils. The  $^{241}\text{Am}$  concentrations of the Trinitite samples measured 0.5 nCi/g, 0.024 nCi/g and 0.033 nCi/g. Preliminary data from radioecological studies at Los Alamos<sup>1</sup> indicated that  $^{241}\text{Am}$  may enter biological systems to a greater degree than Pu and, consequently, may be of equal or greater importance than Pu as a contaminant in natural ecosystems.

A wide variety of additional radionuclides were identified in Trinitite, including  $^{133}\text{Ba}$ ,  $^{152}\text{Eu}$ ,  $^{155}\text{Eu}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ - $^{90}\text{Y}$ .

The average gross gamma radiation measurements obtained outside the GZ area were not significantly different from the measurements obtained at remote locations or what could be considered to be the natural background radiation levels for the central New Mexico area.<sup>7</sup> Measured values of radiation also fall within

the range of 12-20  $\mu$ R/h suggested by Cowan<sup>13</sup> as being normal for the northern White Sands Missile Range elevation depending on the geological composition of the earth's crust. The measured values within GZ were significantly above background levels and approached a maximum of one mR/h under the measurement circumstances noted previously.

Results of this preliminary investigation indicated that the general pattern of Pu distribution in soils, vegetation, and rodents as a function of distance from GZ was similar to the findings of Leitch<sup>5</sup> and Olafson et al.<sup>8</sup> However, there has been an increased migration of Pu into the soils since the last measurements were made about 20 years ago. Concentrations of Pu in all sample types of the present study were generally similar to the findings of 20 years ago at Trinity Site. However, the limited number of samples analyzed in the present study does not allow an adequate comparison, and more refined studies are currently in progress.

#### Literature Cited

1. Hakonson, T. E., J. W. Nyhan, L. J. Johnson and K. V. Bostick. 1973. Ecological investigation of radioactive materials in waste discharge areas at Los Alamos. Los Alamos Scientific Laboratory Report LA-5282-MS.
2. Blair, W. F. 1943. Ecological distribution of mammals in the Tularosa Basin, New Mexico. *Contr. Lab. Vert. Biol. Univ. of Michigan* 20: 1-24.
3. Larson, K. H., J. L. Leitch, W. F. Dunn, J. W. Neel, J. H. Olafson, E. E. Held, J. Taylor, W. J. Cross, and A. W. Bellamy. 1951. Alpha activity due to the 1945 Atomic bomb detonation at Trinity, Alamogordo, New Mexico. Univ. of California Report UCLA-108.
4. Shields, Lora M. 1956. Zonation of vegetation within the Tularosa Basin, New Mexico. *The Southwest Naturalist* 1(2): 49-680.
5. Leitch, J. L. 1951. Summary of the radiological findings in animals from the biological surveys of 1947, 1948, 1949 and 1950. Univ. of California Report UCLA-111.
6. Herceg, J. E. 1972. Environmental monitoring in the vicinity of the Los Alamos Scientific Laboratory. Los Alamos Scientific Laboratory Report LA-4970.
7. Johnson, L. J. 1972. Los Alamos land areas environmental radiation, 1972. Los Alamos Scientific Laboratory Report LA-5097-MS.
8. Olafson, J. H., H. Nishita and K. H. Larson. 1957. The distribution of plutonium in the soils of central and northeastern New Mexico as a result of the atomic bomb test of July 16, 1945. Univ. of California Report UCLA-406.
9. Olafson, J. H. and K. H. Larson. 1961. Plutonium, its biology and environmental persistence. Univ. of California Report UCLA-501.
10. Francis, C. W. 1973. Plutonium mobility in soils and uptake in plants: a review. *J. Environ. Quality*, 2(1): 67-70.
11. Larson, K. H. 1958. Unpublished data. Cited in reference 9.
12. Whicker, F. W. 1973. Radioecology of some natural organisms and systems in Colorado. Eleventh Annual Progress Report on Atomic Energy Commission Contract AT(11-1)-1156.
13. Cowan, F. P. 1959. Natural radiation background. In *Radiation Hygiene Handbook*, McGraw-Hill Book Co., N.Y.

## THE DISTRIBUTION OF PLUTONIUM IN LIQUID WASTE DISPOSAL AREAS AT LOS ALAMOS

T. E. Hakonson, L. J. Johnson and W. D. Purtymun  
Los Alamos Scientific Laboratory  
Los Alamos, New Mexico 87544

### Abstract

This paper describes an ecological investigation of plutonium in the Los Alamos Scientific Laboratory environs. Data are presented on the distribution of plutonium in the alluvial sediments, water, vegetation and rodents from Mortandad Canyon, an area which has been used for liquid waste disposal for 10 years.

### Introduction

A survey was initiated during 1972 to determine the concentrations of liquid effluent-associated radionuclides in the alluvial sediments, water, and some of the natural biota in waste discharge areas. This paper will summarize preliminary findings on the distribution of  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  in Mortandad Canyon, an area which has been used as a liquid effluent disposal area since 1963. The data were obtained during a one-week sampling period in October 1972.

### Methods and Materials

Mortandad Canyon originates in the western portion of the Laboratory property at an elevation of about 2225 meters above sea level, and terminates about 15 km from its origin in the Rio Grande River on the eastern edge of the Laboratory property at an elevation of about 1700 meters.

Radionuclide-bearing liquid effluents from a waste treatment plant located on a mesa adjacent to Mortandad Canyon enter the stream channel at an elevation of 2200 meters near the origin of the canyon. The input of waste water over the last 10 years has been relatively constant at about 200 kiloliters per day. The effluent water along with a continuous supply of uncontaminated water ( $\approx 50$  kl/day) from a steam plant situated at the head of the canyon, moves as surface water over thin alluvial deposits ( $<30$  cm deep) for a distance of from about 500 - 1300 meters below the effluent outfall (post-outfall). The effluent disappears into the alluvium and the remainder of the stream channel is dry at distances beyond 1300 meters post outfall, where the canyon and stream channel widen with a corresponding increase in alluvium depth ( $>30$  cm).

It was estimated that about 40 mCi of  $^{238, 239}\text{Pu}$  was released into Mortandad Canyon from 1963 to 1973; and furthermore, that since 1970 at least 80 percent of the plutonium activity was  $^{238}\text{Pu}$  ( $^{238}\text{Pu}/^{239}\text{Pu} = 4$ ).

A permanent sampling network was established in the canyon during the summer of 1972 at points 100 and 200 m above the waste discharge outfall (pre-outfall) to serve as a source of "background" samples and also at 0, 20, 40, 80, 160, 320, 640, 1280, 2560, 5120, and 10,240 m below the outfall (post-outfall).

Surface and ground water, sediment, vegetation and rodents were collected from the stream channel area in the canyon using collection techniques which have been described elsewhere.<sup>1</sup>

Samples were subjected to a hydrofluoric-nitric acid leach, an ion exchange separation, electrodeposition, and alpha-ray spectroscopy corrected for yield by use of tracer quantities of  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  to quantify the plutonium content.

The plutonium content of all sample materials except rodents was sufficient to reduce the relative standard deviations of the determination to less than 30 percent (based on counting statistics). However, the generally low levels of plutonium in rodent tissues in combination with the small sample masses resulted in relative standard deviations usually greater than 30 percent. The minimum detectable amount of  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  based on a 23 hour count was 0.03 pCi/sample ( $\alpha = 0.05$ ).

### Results and Discussion

The  $^{238}\text{Pu}$  content of water, vegetation, and the 0-2.5 cm layer of the alluvial sediments as a function of distance from the effluent outfall in Mortandad Canyon is presented in Fig. 1. The data for  $^{239}\text{Pu}$  which behaved similar to that shown for  $^{238}\text{Pu}$  in Fig. 1, can be inferred from  $^{238}\text{Pu}/^{239}\text{Pu}$  activity ratios presented later in Table 2. The data for vegetation were grouped according to growth form (grasses, shrubs, and trees) and the grouped data were plotted as a function of distance post-outfall. All of the grass samples analyzed were of the genus Poa with the exception of the 5,120 meter post-outfall sample, which was Bouteloua gracilis. The shrub category consisted of Artemisia tridentata, Berberis fendleri, Chrysothamnus parryihowardi, Quercus gambelli, Prunus virginiana, Salix spp. and Rhus trilobata. Tree samples included Acer negundo, Juniperus monosperma, Pinus ponderosa, Pinus flexilis and Pseudotsuga taxifolia.

It is apparent from the data in Fig. 1 that the chronic input of low level radioactive liquid wastes into Mortandad Canyon over the last 10 years has resulted in  $^{238}\text{Pu}$  (and  $^{239}\text{Pu}$ ) concentrations in some post-outfall samples which are two to three orders of magnitude higher than corresponding pre-outfall samples. Maximum concentrations of plutonium in all samples occurred within 160 meters post-outfall and concentrations then declined steadily with distance to near pre-outfall levels at the 5,120 and 10,240 meter post-outfall sampling stations.

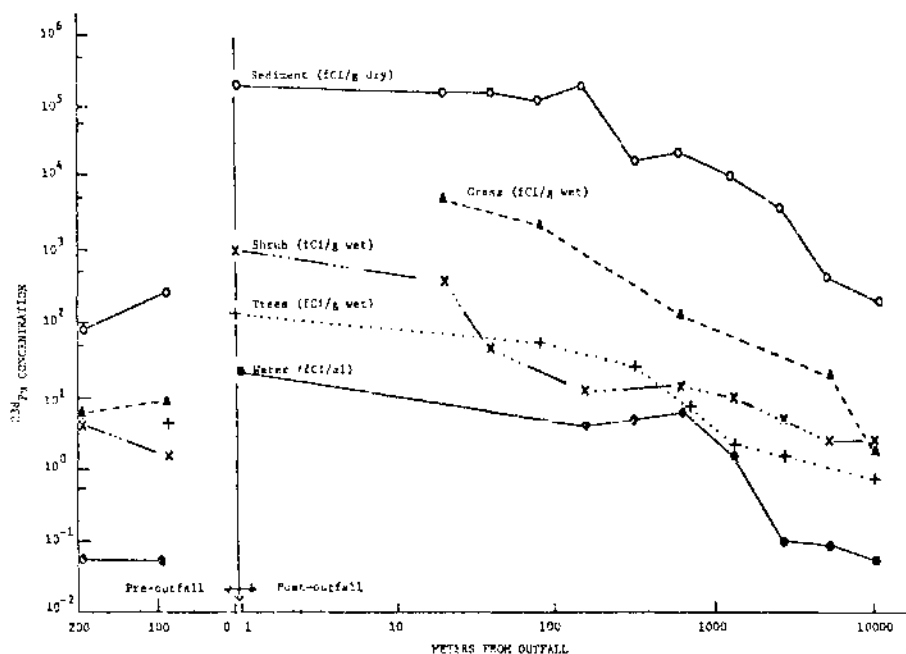


Figure 1. The  $^{238}\text{Pu}$  concentrations in sediment (0-2.5 cm layer), vegetation and water from Mortandad Canyon in October 1977.

Stream channel sediments clearly are the major reservoir of the waste plutonium (Fig. 1). Levels of both isotopes increased from less than 0.5 pCi/g dry at the pre-outfall stations to over 300 pCi/g in post-outfall samples. The concentrations of both  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  were relatively uniform to sampling depths of 30 cm from the effluent outfall to the 1,280 meter post-outfall sampling station, where surface water exists for at least part of the year. At distances greater than 1,280 meters post-outfall, the plutonium was increasingly concentrated in the top 2.5 cm of sediment.

The degree of vertical mixing of plutonium in the alluvial sediments in the canyon appears to be associated with the presence or absence of surface water. Whether the water physically mixes the sediments or acts as a medium for diffusion of plutonium is unknown at this time.

There is some evidence, as reviewed by Francis<sup>2</sup>, that plutonium does migrate downward in soils after extended exposure to the natural environment and may become more available to vegetation with time because of an enhanced root contact with the isotope. Studies at Trinity Site, which are described in a paper at this symposium demonstrated that there has been a marked penetration of Pu into the soils since 1950, when similar measurements were last made.

There was an apparent distributional relationship between the  $^{137}\text{Cs}$  and plutonium content of the alluvial soils. A log-log plot of the  $^{137}\text{Cs}$  concentrations in the 0-2.5 cm layer of post-outfall sediments versus the corresponding data for  $^{238}\text{Pu}$  (and  $^{239}\text{Pu}$ ) were linearly related. The correlation coefficients ( $r$ ) for the respective linear regressions were 0.97 ( $^{137}\text{Cs}$  vs  $^{238}\text{Pu}$ ) and 0.88 ( $^{137}\text{Cs}$  vs  $^{239}\text{Pu}$ ). The interpretation of this observation is not clear at this time, but may indicate that the distributive mechanism for these two radionuclides in Mortandad Canyon sediments may be similar.

The plutonium concentration in surface and ground water in the stream channel paralleled the data for sediments but at a much lower level (Fig. 1). Maxima of 29 fCi  $^{238}\text{Pu}$ /ml and about 1 fCi  $^{239}\text{Pu}$ /ml were observed in surface water at the effluent outfall and concentrations of both isotopes decreased to less than 0.1 fCi/ml in ground water at the 5,120 m post-outfall sampling station.

There appeared to be a relationship between the proximity of vegetation to the ground surface and the plutonium content of the plant material. In general, grass species which were <50 cm tall, contained the highest levels of both  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$ , whereas shrubs and trees (>1 m tall) contained relatively moderate to low amounts of plutonium. Perhaps the rooting zone of the grasses contained higher Pu concentrations than the shrubs or trees or possibly the low growth form of the grasses increased their susceptibility to exterior surface contamination by Pu contaminated materials.

If the plutonium measured in vegetation was contained within the plant, then calculated grass/sediment activity ratios of  $2.3 \times 10^{-2}$  and  $7.8 \times 10^{-3}$  (or about  $8 \times 10^{-2}$  and  $3 \times 10^{-2}$  on a dry weight basis) for  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  were relatively high compared to values of  $10^{-6}$  -  $10^{-3}$  reported by other investigators for the root uptake of Pu from plant-soil systems.<sup>3,4,5,6</sup> However, whether the plutonium was distributed within, or on, the plant is unknown at this time.

The plutonium concentrations in the liver, lungs, hide, and carcass of rodents (*Peromyscus maniculatus*, *P. truei*, *Reithrodontomys megalotis*) collected on the stream channel in Mortandad Canyon (Table 1) varied by as much as three orders of magnitude in samples from the same collection location. Some of this variation was undoubtedly due to the large uncertainties associated with the counting data and to species variation. There were insufficient samples at each station to permit any species comparison.

Mean plutonium concentrations were highest in the lung and hide samples from each collection location which suggested that inhalation of resuspended sediments may be the main route of Pu entry into these small, ground-dwelling rodents. Post-outfall concentrations varied from a maximum of about 8000 fCi  $^{238}\text{Pu}$ /g wet in the lungs of one rodent down to levels which were indistinguishable from background. Concentrations of  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  in rodent tissues from pre-outfall and other areas<sup>1</sup> on site measured 10 fCi/g or less. Liver and carcass samples, in general averaged from 0.1-0.01 times the plutonium concentrations of hide and lungs.

The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios for the various sample types, which are presented in Table 2, demonstrate that nearly all of the samples contained a preponderance of  $^{238}\text{Pu}$  on an activity basis. The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios which exceed one in some of the post-outfall remainder (>12.5 cm depths) sediment core sections evidence the fact that complete vertical mixing of  $^{238}\text{Pu}$  has occurred over the last three years.

The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios greater than unity in sediments from the 5,120 and 10,240 meter stations may indicate that some of the plutonium has moved a considerable distance down Mortandad Canyon, despite the low sediment concentrations measured at these sites.



Table 1. The  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  content of rodents from Mortandad Canyon in October 1972.

Location (elevation)	$^{238}\text{Pu}$				$^{239}\text{Pu}$			
	Liver	Lung	Hfnc	Carcass	Liver	Lung	Hfnc	Carcass
100 and 260 (pre-outfall)	2.8 (2.8)	9.9 (21)*	-1.0 (1.4)	0.0 (3.0)	0.42 (2.8)	3.3 (6.6)	4.1 (1.7)	0.0 (3.0)
$\bar{x} \pm 1 \text{ S.D.}^{**}$	2.8	5 $\pm$ 7	0.0	0.0	0.92	5.7 $\pm$ 3.4	9.6 $\pm$ 7.1	0.0
0 (post-outfall)	26 (10)	248. (111)	1303. (228)	88. (6.1)	4.2 (3.1)	76. (74)	185. (28)	11. (12)
$\bar{x} \pm 1 \text{ S.D.}$	25 $\pm$ 16	128 $\pm$ 533	529 $\pm$ 471	43 $\pm$ 43	8.2 $\pm$ 8.6	157 $\pm$ 182	103 $\pm$ 98	6.5 $\pm$ 6.8
2560	19. (13)	563. (1367)	569. (172)	0.86 (0.71)	7.4 (6.5)	501. (364)	198. (81)	1.0 (0.71)
$\bar{x} \pm 1 \text{ S.D.}$	6.4 $\pm$ 7.3	361 $\pm$ 520	75 $\pm$ 176	2.7 $\pm$ 4.3	4.3 $\pm$ 4.7	80 $\pm$ 130	29 $\pm$ 65	0.11 $\pm$ 0.62
10240	20. (11)	457. (133)	38. (22)	5.6 (1.9)	2.5 (3.8)	58. (58)	0.0 (23)	0.93 (1.9)
$\bar{x} \pm 1 \text{ S.D.}$	11 $\pm$ 25	780 $\pm$ 2278	22 $\pm$ 49	7.9 $\pm$ 21	13 $\pm$ 34	135 $\pm$ 1153	477 $\pm$ 1694	1.9 $\pm$ 4.0

\* (d)g wet  $\pm 1 \text{ S.D.}$  based on counting statistics.

\*\* The mean was calculated by assigning a 0.0 to any negative value and by disregarding any less than (4) values.

Vegetation and rodent tissue reflected the enhanced  $^{238}\text{Pu}$  content of water and sediment in the canyon but not in any readily identifiable pattern. There was a tendency for the ratio in grasses to exceed the ratio in shrubs and trees.

The release of low level plutonium wastes in liquid effluents has resulted in significant plutonium concentrations in several of Mortandad Canyons ecological components. The presence of water in the stream channel appeared

Table 2. The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios in water, sediment, vegetation, and rodents from Mortandad Canyon in October 1972.

Distance From Outfall	Water	Sediment			Vegetation			Rodents				
		0-2.5	2.5-7.5	7.5-12.5	>12.5	Grass	Shrub	Trees	Liver	Lung	Hfnc	Carcass
200	0.14	0.02	0.07	--	4.6	2.1						
100	0.83	0.19	0.08	0.06		1.7	1.3	3.0	3.0			
0	29	7.9	--	--	--	4.2	1.6	3.5	1.4	7.0	7.8	
20		0.46	3.8	10.	--	14.	7.0					
40		3.9	2.2	1.9	--		2.2					
80		5.3	3.4	0.83	0.33	10.		8.3				
160	8.4	0.82	--	5.1	11.3		1.1					
320	12.	3.4	5.1	5.1	8.1			1.4				
640	9.5	4.0	2.6	5.6	3.6	2.8	1.9	1.1				
1280	8.2	4.4	6.5	3.0	1.9		1.3	3.3				
2560	1.8	4.1	3.6	3.5	0.92		1.9	1.2	2.3	6.8	3.1	3.2
5120	>1.6	1.4	--	1.5	0.70	5.7	2.6					
10240		1.4	1.2	2.2	--	1.8	1.0	0.32	2.3	4.5	6.5	3.8

to be correlated with the rate and degree of vertical mixing of plutonium in the alluvial sediments. The mechanisms involved are not understood but may include the mixing action of the flowing water and/or the water may serve as a medium for the vertical diffusion of plutonium. Data on low growing grass species showed that the plutonium concentration ratios for plant/sediment are on the order of  $3 \times 10^{-2}$  to  $8 \times 10^{-2}$ , which was about an order of magnitude higher than that reported by others for root uptake of plutonium from soils. However, in the present study we cannot rule out the possibility of externally deposited plutonium on the plant materials. There appeared to be a relationship between growth form and the plutonium content of the plant. Lower growth forms contained higher plutonium concentrations than higher growth forms.

The highest mean  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  concentrations in the lung and hide of rodents from the canyon suggested that resuspension of sediment-bound plutonium may be a prime mechanism in the contamination of rodents. The appreciable variation in the plutonium data for rodent tissues indicated that the contamination of the small mammal populations living near the stream channel is heterogeneous, with many individuals receiving minute quantities of plutonium and others receiving relatively large amounts.

The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios calculated from the data in the present study provided assessment of the vertical and horizontal movement of the effluent-associated plutonium. It appeared that in the mezzic portion of the canyon, the vertical mixing to the depths sampled was completed within a three year period. The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios in vegetation and rodents reflected the ratio in water and sediments but in a manner which has yet to be defined.

#### Literature Cited

1. Hakonson, T. E., J. W. Nyhan, L. J. Johnson and K. V. Bostick. 1973. Ecological investigation of radioactive materials in waste discharge areas at Los Alamos. Los Alamos Scientific Laboratory Report LA-5282-MS.
2. Francis, C. W. 1973. Plutonium mobility in soils and uptake in plants: a review. *J. Environ. Quality* 2(1): 67-70.
3. Newbould, P. 1963. Absorption of plutonium-239 by plants. In Annual report on Radiobiology ARCRL 10, Gr. Brit. Agr. Res. Council, Radiobiological Lab., Wantage, Berks, England. p. 86.
4. Rediske, J. H., J. F. Cline, and A. A. Selders. 1955. The adsorption of fission products by plants. USAEC Report IIW-36734, p. 1-17.
5. Wilson, D. O. and J. F. Cline. 1966. Removal of plutonium-239, tungsten-185, and lead-210 from soils. *Nature* 209: 941-942.
6. Cummings, S. L. and L. Roberts. 1971. The uptake of Ce-144, Promethium, and Plutonium-238 by oat plants from soils. *Rad. Health Data Rep. No. 2*, 12: 83-85.

РАДИАЦИОННО-ГИГИЕНИЧЕСКАЯ ОЦЕНКА ГЕОХИМИЧЕСКИХ ПРОВИНЦИЙ  
С ПОВЫШЕННОЙ МИГРАЦИЕЙ ЦЕЗИЯ-137

А. Н. Марей, Р. М. Бархударов, Н. Я. Новикова  
Институт биофизики Министерства здравоохранения СССР  
Москва, Союз Советских Социалистических Республик

Аннотация

This report gives the results of persistent studies on Cs-137 transfer through the food chain.

The areas with increased intake of Cs-137 from the soil into the plants and food (milk, meat) have been identified.

Cs-137 content in these foods exceeds by 10-100 times the average value for the Soviet Union. Accordingly Cs-137 contained in the bodies of rural population is also higher. The average internal radiation doses for humans of various ages ranged from 11 to 19 mrem/year. Radiation exposure has been predicted. The possibilities of leukemia occurrence has been considered. It is shown that probability for leukemia development during 70 years is  $1.2 \times 10^{-3}\%$ .

Широкие перспективы развития атомной энергетики требуют всестороннего изучения особенностей поведения искусственных радионуклидов в зонах потенциального распространения радиоактивных отходов. В частности это касается миграции цезия-137 по пищевым цепям.

В плане изучения гигиенических аспектов проблемы цезия-137 как потенциального источника внутреннего облучения населения наибольший интерес представляют сельскохозяйственные районы, где производятся пищевые продукты массового потребления, способные стать критическими в отношении цезия-137. Задачи работы включали в себя:

- выявление на территории Европейской части СССР зон с повышенной миграцией;

- установление причин повышенной миграции, констант, характеризующих переход его из некоторых типов почв в растения; выявление критических продуктов;

- гигиеническая оценка доз, получаемых сельским населением за счет инкорпорированного цезия-137.

В основу настоящего сообщения положены материалы исследований, проведенных авторами на протяжении 5 лет (1967-1972 гг.) в 20 районах на территории 12 областей, входящих в состав трех республик. Характеристика методов и организация работ содержится в ранних публикациях /1, 2/.

Наибольший интерес с точки зрения миграции цезия-137 из почвы в растительность представляют так называемые полесья<sup>†</sup>.

В Европейской части Советского Союза полесские низины находятся преимущественно в южной части нечерноземной зоны, начиная от границ Польши и до Урала. В западной части этого пояса расположено наиболее крупное Белорусско-Украинское полесье, в центральном районе — Мещера и Верхневолжская низменность, к востоку — цепь низин Лесного Подесья и ряд других. Площадь всех этих низин превышает 0,2 млн км<sup>2</sup>.

Почвенный покров этих провинций представлен разновидностями дерново-подзолистых (лесных) и торфяно-болотных песчаных, супесчаных и суглинистых почв различной степени заболоченности и отлеивности. Для них характерны высокая кислотность, низкое содержание обменных форм кальция, калия, натрия, что обусловлено их значительным увлажнением, а местами — интенсивной промывкой грунтовыми водами.

Основная часть территории полесий осушена и занята под посевы сельскохозяйственных культур. Частично заболоченные участки и земли, не удобные для пахоты, сформированные преимущественно болотными и дерновоподзолистыми песчаными почвами, используются под сенокосы и выносы, что особенно характерно для Белорусско-Украинского полесья, где преобладает животноводческое (молочно-мясное) направление сельского хозяйства. В восточных районах под выпасы молочного скота используют угодья с преобладанием суглинистых почв.

Содержание цезия-137 в почвах всех изучаемых районов примерно одинаково, составляя приблизительно 80 мкюри/км<sup>2</sup>. При этом на целинных почвах 80% его находится в верхнем десятисантиметровом слое.

Из числа полесских низин наибольший интерес в санитарном отношении представляет Белорусско-Украинское полесье, где явления миграции цезия-137 выражены наиболее ярко. Максимальное поступление цезия-137 в растения наблюдается на торфяно-болотных почвах, подстилаемых песками. По мере изменения механического состава почв от песчаных к суглинистым происходит постепенное снижение степени миграции цезия-137. Как видно из табл. I, наибольшие коэффициенты накопления наблюдаются на легких песчаных почвах. На супесчаных и суглинистых почвах коэффициент накопления значительно ниже. Высокий  $K_n$  цезия-137 для почв Белорусско-Украинского полесья объясняется особенностями минералогического состава почв и более низким содержанием доступного растениям калия.

Кроме того, как показали наблюдения, существенное влияние на степень миграции цезия-137 из почвы в растения оказывает уровень грунтовых вод. Так, при понижении уровня грунтовых вод на 0,7–1,0 м, как это наблюдалось в засушливое лето 1972 г., концентрация цезия-137 в растительности и  $K_n$  снизились в 2–3 раза. Подобное явление наблюдалось также на осушенных почвах. Наибольший коэффициент накопления цезия-137 растительностью наблюдается в западных районах ( $K_n=5,3$ ), а в центральных и восточных, где отмечается наличие глинистых минералов в илстой фракции и относительно высокое содержание подвижного калия,  $K_n$  заметно снижается (2,5–1,9). Мелиорация почвы, внесение в нее калийных удобрений приводят к значительному снижению уровня поступления цезия-137 в растения. Так, поступление цезия-137 в фуражные культуры из пахотных почв в 3–10 раз ниже, чем из аналогичных целинных почв в пастбищную растительность.

Таблица 1

Показатели перехода цезия-137 из почвы  
в пастбищную растительность

Тип почв	Белорусско-Украинское попесье		Мещерская низменность	
	Содержание цезия-137 в растительно- сти, пкюри/кг	$K_H$	Содержание цезия-137 в растительно- сти, пкюри/кг	$K_H$
Разновидности торфяно-болотных и дерново-подзолистых песчаных почв	2370±900	5,3 (1,6-1,4)	950±760	2,5 (7,0-0,7)
Разновидности дерново-подзолистых супесчаных и суглинистых почв	450±300	0,5 (1,5-0,22)	90±20	0,2 (0,3-0,1)

ПРИМЕЧАНИЕ: В скобках указан диапазон измерений.

Содержание цезия-137 в пищевых продуктах растительного происхождения, выращенных на пахотных почвах, существенно ниже, чем в пастбищных травах с целинных участков. Наибольшее загрязнение пищевых продуктов цезием-137 наблюдается в ряде районов Белорусско-Украинского попесья со специфическими типами почв, показателем чего могут служить коэффициенты накопления ( $K_H$ ).

Таблица 2

## Содержание цезия-137 в овощах и корнеплодах

Тип почв	Капуста		Картофель		Свекла	
	пкюри кг	$K_H$	пкюри кг	$K_H$	пкюри кг	$K_H$
Окультуренные торфяно-глеевые, подзолисто-глеевые песчаные	110±100	0,29	143±130	0,34	62±49	0,15
Окультуренные подзолисто-глеевые супесчаные	34±22	0,07	60±20	0,13	13	0,03

Заслуживает внимания то обстоятельство, что даже при самом высоком уровне поступления цезия-137 в овощи и корнеплоды  $K_n$  остается меньше 1, тогда как при миграции цезия-137 из подобного типа целинных почв в пастбищные травы он существенно выше, составляя для первого типа почв в среднем 5,3, для второго 0,5.

Максимальные концентрации цезия-137 в молоке и мясе коров наблюдаются в период пребывания скота на подножном корму, минимальные — зимой при стойловом содержании. В тех случаях, когда выпасы находятся на торфяно-болотных и дерново-подзолистых песчаных почвах, содержание цезия-137 в молоке составляет  $1050 \pm 420$  пкюри/л. При наличии дерново-подзолистых суглинистых почв концентрации изотопа в молоке не превышают  $70 \pm 30$  пкюри/л. Во всех случаях коэффициент накопления цезия-137 в молоке находится в пределах 0,34-0,39. В мясе крупного рогатого скота данной зоны концентрации цезия-137 в говядине примерно в 4 раза, в телятине до 5 раз выше, чем в молоке. В свинине концентрации изотопа в 2-3 раза ниже, чем в говядине.

Суммарное содержание цезия-137 в рационе сельского населения Белорусско-Украинского полесья и удельный вклад изотопа с различными пищевыми продуктами характеризуется следующими величинами (см. табл. 3).

Таблица 3

Поступление цезия-137 с суточным рационом сельскому населению Полесья

Наименование продуктов	пкюри	% от содержания в рационе
Хлеб и хлебобродуцкы	16	1,3
Молоко	920	71,3
Мясо	138	10,9
Рыба (пресноводная)	33	2,7
Картофель	126	10,0
Овощи	12	1,0
Фрукты	1	0,1
Грибы	33	2,7
ВСЕГО	1279	100

Как следует из таблицы, молоко является для данной зоны критическим продуктом.

Следствием повышенного содержания цезия-137 в пищевых продуктах, производимых и потребляемых на территории изучаемых районов, является более высокое по сравнению с характерными для средних широт величинами накопление его в организме местных сельских жителей. Измерение цезия-137 в организме человека проводилось в 15 населенных пунктах, и обследовано около 1000 человек, начиная с 5-летнего возраста. Общая картина распределения людей по содержанию в их организме цезия-137 дана в табл. 4.

Таблица 4

Содержание цезия-137 в организме жителей  
некоторых районов Полесья

Контингент	Общая численность	Диапазон величин, кюри/организм					
		$5 \cdot 10^{-9}$ - $1 \cdot 10^{-8}$	$10^{-8}$ - $5 \cdot 10^{-8}$	$5 \cdot 10^{-8}$ - $1 \cdot 10^{-7}$	$10^{-7}$	Максимум	Среднее
Мужчины	232		33 (14,7)	68 (29,3)	131 (56)	$6,4 \cdot 10^{-7}$	$1,12 \cdot 10^{-7}$
Женщины	307	3 (1,0)	98 (32,0)	147 (47,8)	59 (19,2)	$1,9 \cdot 10^{-7}$	$0,68 \cdot 10^{-7}$
Дети 5-10 лет	271	9 (3,3)	198 (72,9)	57 (21,2)	7 (2,6)	$2,0 \cdot 10^{-7}$	$0,45 \cdot 10^{-7}$
Дети 11-16 лет	180	9 (5)	82 (45,6)	82 (45,6)	7 (3,8)	$1,4 \cdot 10^{-7}$	$0,5 \cdot 10^{-7}$

ПРИМЕЧАНИЕ: В скобках указан процент от общего числа обследованных лиц.

В области наибольшего содержания (более  $10^{-7}$  кюри/организм) наиболее многочисленна, как и следовало ожидать, группа мужчин, в области наименьших - дети до 10 лет.

Несколько иная картина наблюдается, если рассматривать удельные концентрации изотопа в организме и соответствующие им величины поглощенных доз (см. табл. 5).

Таблица 5

Среднегодовые дозы внутреннего облучения  
населения некоторых пунктов Полесья,  
мрад/год

Контингент	Средняя	Максимальная	Контингент	Средняя	Максимальная
Мужчины	19	43	7-8 лет	15	39
Женщины	11	29	9-10 лет	16	62
4-16 лет	15	41	11-16 лет	12	43
4-6 лет	16	30			

Здесь нет столь заметной разницы между взрослыми и детьми, более того, облучение детей лишь немногим меньше облучения взрослых мужчин, а учитывая большую радиочувствительность

детского организма, детский контингент следует признать критическим. Оценка содержания цезия-137 в организме всех жителей Полесья, проведенная косвенным методом, и расчет на этой основе средневзвешенных доз дали следующие результаты: сельское население Белорусско-Украинского полесья - 4,4 мрад/год, все население Полесья - 2,9 мрад/год. Предполагаемая популяционная доза, рассчитанная из условий, что очищение почвы от цезия-137 происходит со скоростью радиоактивного распада и что сельское население Полесья является демографически замкнутой группой, составляет 600 мрад. Для всего населения обследованного района - 180 мрад. Эти величины соответственно в 30 и 9 раз выше средних показателей для населения СССР / 3/. Однако даже в этом случае вероятность возникновения злокачественных новообразований у жителей Полесья настолько низка, что не может быть обнаружена на фоне спонтанных случаев. Так, например, если принять, в соответствии с рекомендациями МКРЗ /4/, что доза в 1 рад на индивидуум дает 20 случаев лейкемий на  $10^6$  человек, то степень риска по этому тесту для сельского населения Полесья составит лишь  $1,2 \cdot 10^{-3}\%$  за всю жизнь.

Исходя из существующих данных, можно предполагать, что и по другим видам радиологических последствий степень риска будет столь же низкой.

Таким образом, внутреннее облучение местных жителей, по-видимому, не представляет опасности для здоровья, но масштабность явления заслуживает серьезного внимания.

---

+ Под полесьем понимают обширную заболоченную низину, покрытую лесом.

#### Литература

1. Marey A. N., Barchudarov R. M., Novikova N. Y. Effect of natural factors on cesium-137. Health Physics, 1972, v. 22, pp. 9-15.
2. Марей А. Н., Р. М. Бархударов, В. К. Чумак и др. Особенности поступления глобальных цезия-137 и стронция-90 по пищевым путям в Полесье. Гигиена и санитария, 1970, № 1, 61-65.
3. Ушакова А. П., И. А. Лихтарев, А. А. Моисеев. Популяционная доза облучения населения СССР от глобального цезия-137. Гигиена и санитария, 1970, № 7, 54-59.
4. Recommendations of the International Commission on Radiological Protection (Adopted September 17, 1965). ICRP Publication 9. Oxford-London, Pergamon Press, 1967.



## МИГРАЦИЯ РАДИОАКТИВНЫХ НУКЛИДОВ В РАЗЛИЧНЫХ ГИДРОГЕОЛОГИЧЕСКИХ УСЛОВИЯХ

А. С. Белицкий, Е. И. Орлова, Ю. А. Жаков,  
Р. А. Челышева, В. А. Смиренная, В. П. Круглов  
Институт биофизики Министерства здравоохранения СССР  
Москва, Союз Советских Социалистических Республик

### Аннотация

The report presents various methods for storage of liquid wastes from atomic power stations. The conditions for underground transport of radioactive contamination as a result of unsealing of radioactive waste reservoirs have been considered. The assessment of radioactive wastes' movement in different hydrological conditions has been given.

Работа современных атомных электростанций неизбежно связана с возникновением радиоактивных отходов. Из них жидкие отходы потенциально наиболее опасны для загрязнения внешней среды, так как содержат основное количество долгоживущих изотопов и по своему физическому состоянию хорошо мигрируют в объектах внешней среды.

В интересах охраны природных ресурсов необходимо стремиться к полной локализации продуктов деления и наведенной активности в жидких отходах в пределах небольших участков на территориях АЭС или вблизи их, достаточно изолированных от мест проживания и активной деятельности людей, т. е. эти станции должны быть по отношению к объектам внешней среды практически "безотходными" предприятиями.

Указанный принцип может обеспечиваться выбором благоприятных природных и санитарных условий для создания могильников и хранилищ отходов, а также инженерными мероприятиями. Эти два фактора взаимосвязаны: чем хуже природные условия, тем сложнее и дороже осуществить инженерные мероприятия. Следует еще отметить, что по сравнению с инженерными мероприятиями природные условия, как правило, являются более устойчивыми во времени, поэтому при выборе мест захоронения радиоактивных отходов им следует отдавать предпочтение.

По действующим в СССР санитарным нормативам могильники и хранилища радиоактивных отходов должны быть связаны с площадками размещения атомных электростанций. Поэтому одним из критериев санитарно-радиационной оценки указанных площадок также должны являться элементы природных условий, которые ограничивают миграцию радиоактивных и химических загрязнений из могильников и хранилищ в объекты внешней среды.

Современные методы обработки жидких радиоактивных отходов на атомных электростанциях позволяют их сконцентрировать путем выпарки и ионного обмена в относительно небольших объемах.

Кубовые остатки и отработанные ионообменные смолы и шламы направляются в специальные бетонные емкости, облицованные нержавеющей сталью, иногда по типу "банка в банке". В СССР начато освоение отверждения жидких и полужидких среднеактивных отходов методом битумирования, но пока большая часть этих отходов хранится в указанных емкостях.

В тех случаях, когда позволяют гидрогеологические и санитарные условия, возможно непосредственное удаление жидких радиоактивных отходов в поглощающие горизонты горных пород, как это осуществляется уже в течение 6 лет на Ульяновской АЭС /1/. При захоронении жидких радиоактивных отходов в недра Земли не всегда требуется проводить концентрирование нетехнологических слабо- и среднеактивных стоков, а можно непосредственно удалять их в поглощающие горизонты.

Считают, что гидроизоляция железобетонных емкостей для кубовых остатков, отработанных смол и шламов не является гарантированной во времени. По условиям проектирования, строительства и эксплуатации этих емкостей надежность изоляции указанных отходов в них составляет 25-30 лет. Не исключается аварийное нарушение герметичности емкостей и в более ранние сроки.

При разгерметизации емкостей (могильников) жидких отходов степень опасности радиоактивного загрязнения объектов внешней среды определяется способностью радиоизотопов мигрировать в горных породах.

Миграция радиоизотопов в породах зависит в значительной степени от сорбционной способности пород и химического состояния изотопов фильтрующихся растворов. Сорбционная способность пород очень разнообразна и экспериментально определяется в каждом конкретном случае для образцов пород, отобранных в районе расположения могильников АЭС.

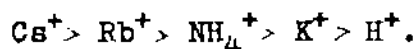
Форма нахождения радиоизотопов зависит в основном от химического состава инфильтрирующихся вод. При разгерметизации емкостей, как правило, расположенных выше уровня грунтовых вод, в почвы и подстилающие породы зоны аэрации будут поступать жидкие отходы, поэтому химическое состояние радиоизотопов в почвенных растворах будет определяться составом этих отходов.

Химический состав жидких отходов в емкостях был определен на примере отходов Нововоронежской АЭС с реакторами водо-водяного типа. Выполненные анализы показали, что жидкие отходы имеют сильно щелочную реакцию и высокую окисляемость (от 4 до 12 г  $O_2$ /л). Сухой остаток отходов колеблется от 90 до 150 г/л. Их состав представлен в основном различными солями натрия, при этом количество боратов колеблется от 2 до 20 г/л, гидрокарбонатов - от 12 до 80 г/л, оксалатов - от 1,5 до 4 г/л, сульфатов - от 1 до 5 г/л, хлоридов - от 1 до 3 г/л. Кроме того, жидкие отходы содержат значительное количество мощных средств, а именно: ОП-7 - до 9 г/л и порошка "Новость" - до 7 г/л.

Радиоактивность жидких отходов почти во всех емкостях составляла  $\approx 10^{-3}$  кюри/л и только в одной емкости была равна  $4,7 \cdot 10^{-5}$  кюри/л. При этом радиоактивность жидкой части отходов была обусловлена в основном цезием-137. Кроме этого радиоизотопа, были обнаружены цезий-134, рутений-106, сурьма-125 и кобальт-60. Цезий-134, рутений-106 и сурьма-125 присутствовали, примерно, в 10 раз меньших количествах по сравнению с цезием-137, а кобальта-60 было в 100 раз меньше, чем цезия-137. Наиболее токсичный среди долгоживущих радиоизотопов стронций-90 не обнаружен в жидкой фазе отходов, что, вероятно, связано с его соосаждением с солями жесткости и практически полным переходом его в твердую фазу отходов.

Как показали исследования, проведенные с помощью ионного обмена и электрофореза, по своему химическому поведению в жидких отходах обнаруженные радиоизотопы резко отличаются друг от друга. Так, радиоизотопы цезия практически полностью находятся в катионной форме, рутений-106 и 50% кобальта-60 – анионной форме, и остаток кобальта-60, а также сурьма-125 присутствуют в виде нейтральных молекул. Следовательно, при загрязнении жидкими отходами почв и подстилающих пород радиоизотопы рутения, сурьмы и кобальта будут мигрировать со скоростью фильтрации растворов, и только радиоизотопы цезия будут поглощаться горными породами.

Миграция цезия будет определяться механизмами сорбции его породами. Цезий сорбируется частично ионообменно на поверхности глинистой и илистой фракций почв и пород и частично фиксируется глинистыми и слюдястыми минералами в межкристаллических пустотах /1, 2, 3, 4 и 5/. Миграция радиоизотопов цезия определяется соотношением между количествами его, сорбируемыми по этим двум типам поглощения. При фиксации значительных количеств цезия в межкристаллических пустотах он задерживается в основном слоями почвы или подстилающих пород, залегающими непосредственно под емкостями. При этом величина поглощения его зависит от количества в растворе тех катионов, которые также способны внедряться в межкристаллические пустоты глинистых и слюдястых минералов. По данным Ю. Л. Кокотова и др. /4/, эти катионы по их способности уменьшать величину сорбции цезия можно расположить в ряд:



Для расчета опасности загрязнения цезием пород и подземных вод необходимо было оценить роль каждого из механизмов сорбции. Проведение такой оценки в экспериментах по сорбции цезия породами из жидких отходов представляет большие трудности. Поэтому она была выполнена в эксперименте по десорбции цезия  $0,1$  в нейтральном растворе хлористого аммония из предварительно загрязненных образцов суглинков, обладающих различной катионообменной емкостью (от 12 до 43 мг-экв/г), имеющих большое количество слоистых материалов. На таких образцах должна преобладать сорбция путем фиксации цезия в межкристаллических пустотах.

Полученные данные показали, что величина десорбции фиксированного цезия примерно на два порядка ниже, чем для цезия, сорбированного ионообменно. Следовательно, фиксированный цезий будет в основном задерживаться поверхностными слоями почв и пород, а дальность распространения его будет определяться только тем количеством, которое сорбируется ионообменно. Кроме того, было обнаружено, что с течением времени десорбции фиксированный цезий постепенно переходит в ионообменный из-за нарушения кинетического равновесия.

При ионообменной сорбции миграция цезия определяется величиной катионообменной емкости пород, солевым составом и объемом жидких отходов, а также скоростью фильтрации через почву и подстилающие породы зоны аэрации. Как было показано выше, в жидких отходах, отобранных из емкостей на Нововоронежской АЭС, отсутствуют радиоизотопы стронция. Но, учитывая возможные изменения технологии очистки сточных вод и состава моющих средств, которые могут привести к появлению стронция в жидкой фазе, представляло интерес прогнозировать и его миграцию в горных породах.

Для расчета миграции радиоизотопов, поглощенных горными породами ионообменно, успешно применяются приближенные дифферен-

циальные уравнения ионообменной хроматографии /6/. Вследствие того, что радиоизотопы находятся в загрязненных поровых растворах в микроколичествах, то при постоянном химическом составе фильтрующихся жидких отходов поглощение изотопов происходит по линейной изотерме сорбции.

Применение одного из таких уравнений позволяет определить за время ( $t$ ) расстояние ( $x_{0,5}$ ), на которое распространится радиоизотоп с относительной концентрацией ( $\varphi_{0,5}$ ), равной половине от его удельного содержания в исходных отходах.

$$x_{0,5} = \frac{hu}{1+h}t, \quad (1)$$

где  $t$  - продолжительность поступления жидких отходов в породы;  $u$  - действительная скорость фильтрации;  $h$  - распределительное отношение:

$$h = \frac{\gamma p}{\delta K_p}, \quad (2)$$

где  $\gamma$  - плотность фильтрующейся через породы жидкости;  $\delta$  - объемный вес породы;  $p$  - общая пористость породы;  $K_p$  - коэффициент распределения радиоизотопа между породой и жидкостью.

Коэффициент распределения обычно определяется в лабораторных экспериментах в статических или динамических условиях для образцов пород, отобранных в районе могильников.

Другое приближенное уравнение позволяет рассчитать за определенное время ( $t$ ) расстояние  $x$ , на которое распространится радиоизотоп с любой заданной относительной концентрацией, меньшей, чем  $\varphi_{0,5}$ .

$$x = x_{0,5} + \frac{2wu}{1+h} \sqrt{\frac{ht}{\beta(1+h)}}, \quad (3)$$

где  $w$  - параметр, зависящий от заданной относительной концентрации радиоизотопа в фильтрующемся растворе. Приблизительно этот параметр может быть получен из уравнения  $\Phi(w\sqrt{2}) \approx 1 - 2$  ( $\Phi(w\sqrt{2})$  - интеграл вероятности);  $\beta$  - константа скорости сорбции. Величина  $\beta$ , имеющая размерность времени в степени минус единица  $t^{-1}$ , в условиях движения растворов в идеальных однородных сорбентах определяется скоростью сорбции. Но при движении загрязненных вод в породах, кроме времени, затрачиваемого на сорбцию, ее также учитываются и другие факторы, стремящиеся размазать фронт движения в воде сорбируемого катионита, а именно неоднородность водных, сорбционных свойств водоносных пород, неодинаковым обменом воды между породами, различной сорбционной способностью частиц, слагающих породы и т.д.

Приблизительно  $\beta$  может быть определена графически по кривым изменения концентрации радиоизотопа в воде или породе с расстоянием от места поступления жидких отходов. Такие кривые могут быть получены в лабораторных экспериментах, проводимых в динамических условиях с применением высоких разборных колонн (для песчаных пород) или специальных фильтрационных приборов, работающих под определенным давлением (для глинистых пород). На этих кривых устанавливаются ( $x_{0,5}$ ) с относительной концентрацией изотопа ( $\varphi_{0,5}$ ) и точка ( $x$ ) с  $\varphi < \varphi_{0,5}$ . Затем определяется

$\beta$  по уравнению:

$$\beta = \frac{4w^2hu^2 t}{(1+h)(x-x_{0,5})^2} \quad (4)$$

Во все приведенные выше основные уравнения входит величина  $\alpha$ , характеризующая действительную скорость движения подземных вод, определение которой в природных условиях представляет некоторые трудности. Поэтому в использованных уравнениях эта величина может быть заменена скоростью фильтрации или объемом фильтрующейся жидкости и активной пористостью пород района могильников.

С помощью приближенных уравнений и на основании коэффициентов распределения цезия и стронция, полученных в лабораторных экспериментах, проведенных с различными горными породами, были выполнены расчеты миграции этих радиоизотопов в различных геологических условиях зон аэрации. Коэффициент фильтрации, по которому может быть определена скорость фильтрации, и активная пористость исследуемых пород, определялись в полевых условиях.

Проведенные расчеты показали, что в однородных песчаных породах фронт движения радиоизотопов стронция и цезия определяется в большинстве случаев уравнением (1), т. е. участком с относительной концентрацией радиоизотопа в воде, равной половине от исходного удельного содержания его в жидких отходах.

Аналогичная зависимость распространения стронция и цезия была получена для зоны аэрации, сложенной преимущественно трещиноватыми базальтами, в толще которых залегают мощные слои крупно- и среднеобломочных вулканических пород с высокой фильтрационной способностью.

При миграции изотопов в глинистых породах наблюдается другая картина фронта их движения. Благодаря небольшой величине распределительного отношения и константы скорости сорбции изотопов стронция-90 и цезия-137 в глинистых породах, отмечается сильное размазывание фронта их распространения. Поэтому для расчета миграции этих радиоизотопов в зоне аэрации, сложенной суглинками и глинами, необходимо применять уравнение (3), так как расположение фронта с небольшой относительной концентрацией радиоизотопа будет далеко впереди от места нахождения исходной концентрации этих изотопов в жидких отходах.

Однако несмотря на значительную величину размазывания фронта движения радиоизотопа в глинистых породах, абсолютная глубина проникновения загрязнения в указанных породах на много меньше, чем в хорошо проницаемых песках и базальтах.

В районе с большим количеством атмосферных осадков возможна десорбция радиоизотопов с загрязненных пород, которая может приводить к проникновению их на большие глубины. Это явление может приводить к нежелательным санитарным последствиям, особенно на участках, сложенных с поверхности земли проницаемыми песчаными породами. При этом наибольшей подвижностью будет обладать стронций-90. В связи с этим был проведен полевой эксперимент по вымываемости стронция-90 из загрязненных песчаных пород зоны аэрации, мощность которых в исследуемом участке была равна 80 м. Наблюдение, проведенное в течение четырех лет, показало, что при количестве инфильтрующихся атмосферных осадков, равном в среднем 360 мм/год, первоначальный объем зоны, ограниченной удельной активностью песка  $1 \cdot 10^{-9}$  кюри/кг, увеличился примерно в три раза.

Проведенные эксперименты и расчеты подтверждают целесообразность учета природных условий при выборе мест расположения могильников для хранения жидких радиоактивных отходов. Указанные могильники следует размещать выше уровня грунтовых вод, преимущественно в слабо проницаемых глинистых

породах. В аридных областях, где испарение значительно превышает количество атмосферных осадков, могильники следует располагать в участках, сложенных мощной зоной аэрации.

Отсутствие в районе расположения АЭС благоприятных гидрогеологических условий для хранения жидких радиоактивных отходов в емкостях требует осуществление дополнительных сложных инженерных устройств. На АЭС, построенных в таких районах, следует в первую очередь организовать отверждение этих отходов.

Непосредственное удаление жидких радиоактивных отходов в глубоко залегающие поглощающие горизонты возможно лишь при наличии пригодной для этого гидрогеологической структуры, что должно быть обосновано проведением широкого комплекса гидрогеологических, геохимических, санитарных и других исследований.

#### Литература

1. Юдин Ф. П., Пименов М. К., Назаров А. И. и др. Атомная энергия, 25, вып. 2, 1968.
2. Alphlett C. B. Ground Disposal of Radioactive Wastes. Atomic and Nuclear Energy, 9, 6, 1958.
3. Tamura T., Jacobs D. G. Health Physics, 2, 4, 1960.
4. Кокотов Ю. А., Попова Р. Ф. и др. Радиохимия, т. III, вып. 2, 1961.
5. Титлянова А. А. Почвоведение, № 3, 1962.
6. Белицкий А. С., Орлова Е. И. Охрана подземных вод от радиоактивных загрязнений. Изд-во "Медицина", М., 1968.

АНАЛИЗ ГЕОГРАФИЧЕСКИХ РАЗЛИЧИЙ СКОРОСТИ ОЧИЩЕНИЯ  
ЦЕПОЧКИ ЛИШАЙНИК-ОЛЕНЬ-ЧЕЛОВЕК ОТ ЦЕЗИЯ -137

М. И. Троицкая, М. С. Ибатуллин, Б. Л. Литвер,  
А. А. Моисеев, А. И. Нижников, П. В. Рамзаев, Л. А. Тенлих  
Институт радиационной гигиены, Ленинград, СССР

ABSTRACT.

The experiments carried out by a number of research workers in the past 10-15 years have proved the food chain lichen-reindeer-man in the Arctic areas to have the highest  $^{137}\text{Cs}$  concentration of the global fallout. The authors of this paper have investigated  $^{137}\text{Cs}$  clearance dynamics of this chain in the years 1961-1973 along the Soviet coastline of the Arctic Ocean stretching for many thousands kilometers from Alaska to Finland. The relevant data have been obtained from 5 regions for all the links of the chain and compared with other similar chains. The northern chain has been found to clear about 2 times slower than other chains. The clearance rate varies in the range of 2 or more times in different geographical regions.

Реферат

Работами ряда исследователей, выполненными в последние 10-15 лет, было показано, что пищевая цепочка лишайник-олень-человек в Арктических районах представляет собой пример наибольшего концентрирования цезия-137 от глобальных выпадений. Авторы данного сообщения проводили исследование динамики очищения этой цепочки от цезия-137 с 1961 по 1973 гг. на всем советском побережье Северного Ледовитого океана, простирающемся на многие тысячи километров от Аляски на востоке до Финляндии на западе. Полученные материалы приводятся по пяти районам по всем звеньям цепочки в сопоставлении с другими аналогичными цепочками. Показано, что северная цепочка очищалась примерно в 2 раза медленнее, чем другие цепочки. Географические вариации ее очищения также лежат в пределах двухкратных (а иногда и более) отклонений.

Введение

Работами Хансона, Пальмера, Грифина на Аляске<sup>1</sup>, Маэттинена и Рахола по Финляндии<sup>2</sup>, Лидена, Густафсона<sup>3</sup>, Линдела и Мэги<sup>4</sup> по Швеции, Аарпрога<sup>5</sup> по Гренландии, Хвиндена и Лиллгравека<sup>6</sup> по Норвегии, нашим работам по СССР<sup>7</sup> продемонстрированы необычно высокие концентрационные свойства северной цепочки лишайник-олень-человек в отношении ряда долгоживущих радионуклидов и, особенно, цезия-137. Звенья этой цепочки оказались нагруженными цезием-137 в 10-100 раз больше, чем аналогичные звенья других наземных цепочек. Факторы и условия, способствующие такому концентрированию, как и закономерности очищения цепочки от цезия-137, уже неоднократно обсуждались. Мнения ученых особенно вариировали по оценке параметра

прогноза загрязнения, каким является скорость очищения цепочки, выраженная через  $T_{1/2}$ .

В своих исследованиях мы ставили задачу по натурному определению этой скорости и ее возможных вариаций на обширной протяженности Советской Арктики, измеряемой десятком тысяч километров.

### Методы и материалы исследования

Пробы лишайников, оленьины и суточных выделений пастухов-оленьеводов отбирались авторами во время ежегодных экспедиций в обследуемые районы, как правило, в зимние периоды года. Без какой-либо обработки в полиэтиленовых упаковках они доставлялись в Ленинградский институт радиационной гигиены и здесь производилось определение цезия-137. Для оленьины и выделений использовалась гамма-спектрометрия в натуральных пробах. Цезий-137 в лишайниках определялся после предварительного радиохимического его выделения в виде висмут-йодида цезия. Содержание цезия-137 в организме оленеводов измерялось в местах их проживания прижизненным методом портативным гамма-счетчиком для всего тела. Ошибки измерений во всех случаях оцениваются не более 10%.

Каждый раз в обследуемом районе отбиралось: в среднем по 5 проб лишайников (кладонии и цетрарии) из-под снега или из заготовленных осенью запасов; по 10 проб оленьины (мышцу от шейного зареза) во время массового забоя или со складов; пробы суточных выделений - от 5-10 оленеводов. Одновременно на счетчике всего тела измерялось 10-20 оленеводов, как правило, (но не всегда) ежегодно одни и те же лица.

Районами систематических наблюдений служили: 1) Мурманская область (Кольский полуостров), граничащая с Финляндией на западе, 2) Чукотка, примыкающая на востоке к Алтске, 3) Коми АССР, 4) Таймыр и 5) Якутия, занимающие промежуточное положение между двумя предыдущими районами.

### Результаты исследования

Отдаленность обследуемых районов от Ленинграда создавала немалые трудности в одновременном охвате наблюдением всех районов и в получении материалов для воссоздания стройной динамики цезия-137 по всем звеньям цепочки. Многих "точек" в наших данных (см. таблицы 1, 2, 3) явно не хватает, чтобы быть слишком категоричными при их анализе в отношении географических вариаций скорости очищения цепочки в целом и ее отдельных звеньев.

Лишайники. Являющиеся основным кормом северных оленей в течение 7-8 месяцев, лишайники, как первое звено цепочки, в конечном итоге определяют (хотя и не полностью) динамику концентрации цезия-137 в оленьине и в организме человека.

В сопоставимые годы концентрация цезия-137 в лишайниках варьирует от района к району не более чем в 2,7 раза. По-видимому, в Мурманской области и, возможно, Коми АССР загрязненность лишайников была более высокой, чем в других (восточных) районах. Это согласуется с двухкратными различиями в количестве атмосферных осадков. Их было в 1963-64 гг. в годы интенсивных выпадений цезия-137 по Мурманской области (500 мм/год) в 2 раза больше, чем в восточных районах. Уверенный анализ очищения лишайников от цезия-137 по нашим материалам не может быть выполнен и мы ограничимся здесь лишь грубой схемой. В концентрации изотопа в лишайниках за 1961-72 гг. условно следует выделить три периода: 1) подъем с 1961 г. по 1964 г. в 3-6 раз вслед за испытаниями ядерного оружия в



Таблица I

Цезий-137 в лишайниках (кладонии и цетрарии) нкюри/кг сухого веса. Приведены средние значения со стандартными ошибками по 5 пробам.

Дата отбора проб	Р а й о н				
	Мурман- ская область	Коми АССР	Таймыр	Якутия	Чукотка
1961	26 ± 7	11 ± 1	-	-	-
У-63	48 ± 10	-	-	-	-
У1-64	-	74 ± 6	-	-	-
Ш-65	50 ± 1	50 ± 1	-	-	-
ХП-65	27 ± 4	-	-	24 ± 7	-
УУ-66	34 ± 4	-	-	-	-
ХП-66-П-67	33 ± 7	-	14 ± 1	21 ± 3	5 ± 1
Ш-68	27 ± 3	12 ± 2	13 ± 3	12 ± 5	10 ± 1
П-У-69	13 ± 2	9 ± 1	9 ± 3	-	-
П-70	14 ± 1	-	-	-	-
П-У-71	16 ± 2	8 ± 1	8 ± 1	-	14 ± 1
Ш-72	-	-	5 ± 1	11 ± 1	-

1961-62 гг. 2) Трех-шести-кратное снижение с 1965 г. до 1968 г.  
3) Стабилизация концентрации, начиная с 1969 г., когда о скорости очищения нельзя сказать что-либо определенно. Эта периодизация в основном проявляется по Мурманской области и Коми АССР. Другие районы не укладываются в данную схему. По Чукотке, например, с 1967 по 1971 г. зарегистрирован даже трехкратный "подъем", на Таймыре - "снижение" отмечено с 1969 по 1972. Такие подъемы и снижения мы берем в "кавычки", поскольку не уверены в их представительности, судя по динамике изотопа в оленине.

Есть два обстоятельства, осложняющие и снижающие значимость анализа очищения лишайников для прогноза загрязнения последующих звеньев:

- ненадежность стандартизации отбора проб,
- возможность существенных вариаций лишайников в корме оленей различных районов.

Дело в том, что сами лишайники по высоте загрязнены цезием-137 весьма неравномерно. Верхушка растения имеет в 5 раз более высокую концентрацию, чем его основание. Различные соотношения отдельных частей растения в пробе, не поддающиеся контролю, могут привести к флуктуации данных, не связанных с истинной динамикой концентрации изотопа. При отборе проб лишайников также вряд ли можно рассчитывать на воспроизведение этим отбором самого процесса поедания лишайников оленями и по данным анализа растения точно прогнозировать концентрацию цезия-137 в оленине.

Северные олени. Три четверти мирового поголовья оленей (2300 тысяч голов) выпасается на территории СССР. В среднем в году по нашим оценкам оленеводы потребляют оленину в количестве эквивалентном по цезию-137 около 230 г мышц в сутки. В одно и то же время географические вариации цезия-137 в оленине оказались более выраженными, чем в лишайниках. Оленина из Мурманской области была во все годы наблюдений загрязненной цезием-137 в 5-7 раз больше, чем в Якутии. Этот факт может быть объяснен не только большей концентрацией изотопа в лишайниках Мурманской области (в 2,7 раза), но и большей (не менее 2 раз) долей лишайников в корме мурманских оленей.

Таблица 2

Цезий-137 в мышцах северных оленей, нкюри/кг сырого веса. Приведены средние значения со стандартными ошибками по 5-10 пробам.

Дата забоя оленей	Район выпаса				
	Мурманская область	Коми АССР	Таймыр	Якутия	Чукотка
XI-61	24(I)	21± 1	8(I)	-	-
У-У1-62	33± 2	16± 2	4± 1	-	8± 1
XI-62-У-63	48± 7	38± 2	13± 3	-	-
I-64-У-64	80± 3	61± 8	-	-	-
XII-64-П-65	96± 8	81± 7	36± 4	17± 3	25± 2
X-65-У-66	79± 4	-	-	13± 1	18± 3
XI-66-П-67	57± 2	30± 3	20± 3	-	24± 4
I-68	45± 2	20± 2	18± 2	8± 1	18± 1
XII-68	45± 2	22± 1	17± 2	6± 1	-
XII-69	38± 2	-	-	-	15± 1
XII-70	37± 3	17± 1	17± 2	-	15± 1
XII-71-III-72	42± 5	-	19± 2	7± 1	12± 4
XI-72-I-73	29± 8	14± 2	11± 1	6± 1	13± 1

В динамике концентрации цезия-137 в оленине также можно выделить три этапа: подъем (4-кратный) с 1961 г. до зимы 1964-65 гг. быстрый спад по  $T_{1/2}=2,5$  года в 1965-66 гг. и медленный спад с 1968 г. по  $T_{1/2}=7$  лет. В районах Якутии и Таймыра с 1966 г. по 1971-72 гг. убедительных изменений не произошло. Это, конечно, не означает, что  $T_{1/2}$  оленины здесь близок к бесконечности. Он во всяком случае мене  $T_{1/2}=30$  лет. Вероятно, существенные неконтролируемые и случайные флуктуации данных еще слишком велики, а время наблюдения мало, чтоб выявить имеющее место очищение цепочки. Некоторое значение могут иметь и продолжающиеся выпадения цезия-137, ролью которых мы пренебрегаем в своем анализе.

Сравнение динамики концентрации изотопа в оленине, с одной стороны, и в мясе других сельскохозяйственных животных средних широт, с другой, показывает, что оленина за весь наблюдаемый период имела почти в 100 раз более высокую концентрацию - очищение говядины, например, шло значительно более быстрым темпом, чем оленины.  $T_{1/2}$  для говядины по имеющимся данным<sup>8</sup> в 1965-67 гг. был равен 1,6 году. В целом с 1964 г по 1967 г снижение уровней цезия-137 в говядине было почти восьмикратным, в то время как для оленины в разных районах оно колебалось от 1 до 2 раз.

Оленеводы. Поступление цезия-137 в организм оленеводов, судя по содержанию изотопа в выделениях, достигло максимума зимой 1965-66 гг., когда оно в Мурманской области почти в 100 раз превышало поступление изотопа у жителей средних широт северного полушария<sup>9</sup>. Минимальное поступление отмечено у оленеводов Якутии, где оно оказалось в отдельные годы в 4-25 раз меньше, чем у оленеводов Мурманской области.

На основании данных по мурманским оленеводам можно говорить о 2,0-2,7-кратном снижении у них поступления изотопа в 1966-1968 гг. В последующие годы заметных изменений не наблюдалось. По районам Таймыра, Якутии и Чукотки определенно говорить о таком снижении не имеет смысла. Здесь, к сожалению, отсутствуют измерения в тот период (начало 1966 г.), когда мог быть максимум поступления.

Поступление изотопа с рационом жителей северных широт, не связанных с лишайниковой цепочкой<sup>8</sup>, снизилось к 1968 г. после достижения максимума (1964 г.) в 5-8 раз.

Таблица 3

Цезий-137 в организме пастухов оленеводов, нкери/кг тела. Приведены средние значения со стандартными ошибками для 10-20 человек.

Дата измерения	Район жительства				
	Мурман- ская область	Коми АССР	Таймыр	Якутия	Чукотка
У1-62	20 <sup>±</sup> 7	4 <sup>±</sup> 1	5 <sup>±</sup> 1		
ХП-62	22 <sup>±</sup> 5	5 <sup>±</sup> 2	4 <sup>±</sup> 1		
П-64	31 <sup>±</sup> 3	24 <sup>±</sup> 2			
УШ-64	24 <sup>±</sup> 3				
1-Ш-65	45 <sup>±</sup> 3	-	13 <sup>±</sup> 1	5 <sup>±</sup> 1	-
XI-65	27 <sup>±</sup> 2			5 <sup>±</sup> 1	
IУ-66	51 <sup>±</sup> 3				
I-67	35 <sup>±</sup> 3	21 <sup>±</sup> 2	12 <sup>±</sup> 1	8 <sup>±</sup> 1	25 <sup>±</sup> 2
П-У-68	33 <sup>±</sup> 2	24 <sup>±</sup> 2	13 <sup>±</sup> 1	6 <sup>±</sup> 1	17 <sup>±</sup> 2
П-У-69	31 <sup>±</sup> 3	19 <sup>±</sup> 1	18 <sup>±</sup> 2	8 <sup>±</sup> 1	
П-70	25 <sup>±</sup> 2				
П-IУ-71	23 <sup>±</sup> 2	8 <sup>±</sup> 1	8 <sup>±</sup> 1	-	13 <sup>±</sup> 1
Ш-72	18 <sup>±</sup> 1		5 <sup>±</sup> 1	5 <sup>±</sup> 1	12 <sup>±</sup> 1
IУ-73		6 <sup>±</sup> 1	5 <sup>±</sup> 1	5 <sup>±</sup> 1	5 <sup>±</sup> 1

В организме оленеводов максимальное содержание цезия-137 было достигнуто весной 1966 г. т.е., три года спустя после прохождения максимума выпадений (1963 г.) и через 1,5 года после максимума в организме людей из средних широт (конец 1964 г.)<sup>10</sup>. Концентрация изотопа в организме оленеводов при этом была в 20-120 раз выше, чем для остальных жителей, не потреблявших оленину в пищу. Географические вариации здесь в основном отражают аналогичные вариации уровня цезия-137 в оленине: то есть, максимум - в Мурманской области и минимум - в Якутии (при 5-кратных различиях). В очищении организма оленеводов можно выделить один период  $T_{1/2}$  - 4 года для Мурманских оленеводов с 1968 по 1972 г.; по Коми  $T_{1/2}$  АССР - 3 года, по Таймыру - 4 года, по Якутии - 8 лет, по Чукотке - 2,5 года. Для жителей средних широт этот период с 1964 г. по 1968 г. не превышал 2,3 года.

### Заключение

Приведенные материалы, требующие дополнительного более углубленного анализа и дальнейших исследований, еще не позволяют окончательно сформулировать закономерности очищения звеньев северной цепочки от цезия-137. Логическое предположение о тесной связи в этом процессе всех звеньев и ведущей роли лишайников в последующем очищении оленей и оленеводов далеко не всегда подтверждается фактическими данными. Создается впечатление, что на скорость очищения последующих звеньев цепочки, кроме очищения "материнского" звена оказывают влияние и другие факторы, в ряду которых могут быть изменения со временем роли лишайников в корме оленей и роли оленины в питании оленеводов. В результате таких изменений скорости очищения последовательных звеньев цепочки оказываются

сильно варьирующими величинами в различных географических районах.

С несомненностью доказывается лишь, что северная цепочка по сравнению с другими наземными цепочками очищается с более медленной скоростью и эта цепочка еще долгое время останется наиболее загрязненной цезием-137.

### Литература

1. Hanson J.C., Palmer H.H., Griffin B.I.  
Radioactivity in northern Alaskan Eskimos.  
Health Physics, Vol.10, 1964, pp.421-429.
2. Rahola E. and Miettinen J.K.  
Accumulation of  $^{137}\text{Cs}$  in Finnish Lapps. Radioactive food-chains in subarctic environment. Principal investigator Jorma K.Miettinen, Helsinki, 1972.
3. Lidén K., Gustafsson K.  
Relationship and seasonal variations of  $^{137}\text{Cs}$  in lichen, reindeer and man in Northern Sweden 1961-1965.  
Radioecological Concentration Processes, Oxford, 1967, 193-208.
4. Lindell B. and Magi A.  
Observed levels of  $^{137}\text{Cs}$  in Swedish reindeer meat.  
Radioecological Concentration Processes, Oxford, 1967, 217-219.
5. Aarkrog A. and Lippert J.  
Environmental radioactivity in Greenland in 1968.  
Riso Report, No.203.
6. Hvinden E. and Billegraven J.  
Cesium-137 and strontium-90 in precipitation, soil and animals in Norway.  
Nature, Vol.192, No.4808, 1961, pp.1144-46.
7. Рамзаев П.В., Моисеев А.А., Троицкая М.Н., Ибатуллин М.С., Теплых Л.А.  
Основные итоги радиационно-гигиенических исследований миграции глобальных выпадений в приарктических районах СССР.  
М., Атомиздат, 1967.
8. Magi A., Snihls J. and Swedjemark G.  
Some measurements on radioactivity in Sweden caused by nuclear test explosions.  
Radiological Health Data, Vol.11, No.10, 1970.
9. Report on the UNSCEAR, New York, 1966.
10. Прокофьев О.Н., Невструева М.А. и др.  
Цезий-137 глобальных выпадений в продуктах питания и организме человека.  
Атомиздат, М., 1969.

HIGH RADIOACTIVITY IN  
DRINKING WATER AND GROUND WATER  
IN SOUTH TEXAS

by

Martin C. Wukasch, P.E.\*  
and  
Lewis M. Cook†

Abstract

Radioactivity measurements in South Texas made by the Texas State Department of Health have demonstrated that high gross alpha, gross beta and radium 226 levels exist in natural ground waters. Levels of radium 226 over 50 times recommended drinking water limits have been found. Data on some water wells sampled are presented, and correlation of radioactivity versus water quality and well depth is attempted. Body burden estimates are presented for some individuals.

Introduction

Uranium and its radioactive daughter products in the earth are responsible for part of the natural background exposure of people through several mechanisms. These include:

- a. Contributions to the gamma-ray dose from the earth;
- b. Release of Radon 222 into the atmosphere with the resulting alpha, beta, and gamma exposure;
- c. Uptake in plants and animals and introduction into the food chain; and
- d. Solution of radioactive minerals in drinking water.

This paper will treat the last of these as it occurs in South Texas.

Uranium was discovered in South Texas in 1950 along a 300 mile belt paralleling the Gulf Coast about fifty miles inland. The trend has been adequately described by others.<sup>1</sup> Economically recoverable deposits were discovered in Karnes and Live Oak Counties, and by 1971 numerous open-pit mines had been stripped, mined, and deserted.

---

\*Director, Division of Occupational Health and Radiation Control, Texas State Department of Health, Austin, Texas

†Supervisor, Environmental Surveillance and Data Processing, Radiation Control Program, Division of Occupational Health and Radiation Control Texas State Department of Health, Austin, Texas

Investigation of a complaint about water discharged from a Live Oak County mine in 1971 led to the discovery that ground water, seeping into mines which were dug below the level of the water table, contained concentrations of radium 226 as high as 192 pCi/l, well in excess of maximum permissible concentrations allowable for discharges.<sup>2,3</sup> The State Health Department had conducted sampling of ground waters in Karnes County in the 1960's and had found levels of radium 226 to be less than six picocuries per liter (pCi/l).

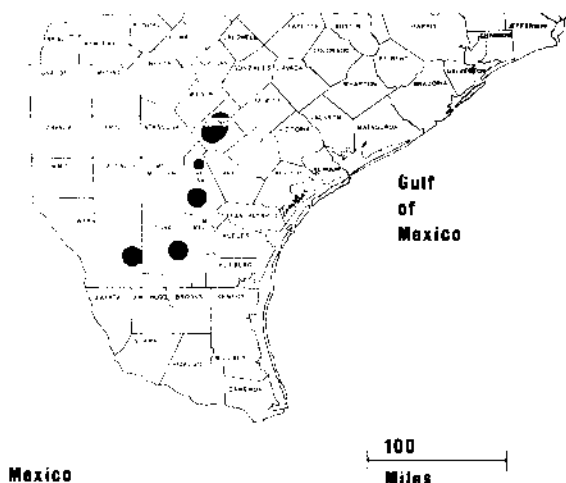


Figure 1 - Map of South Texas Showing County Boundaries and Areas of Commercial Uranium Deposits

Alpha activities in excess of 3,000 pCi/l were found in a uranium exploration well drilled into an ore zone. Radium accounted for 3.6 pCi/l of this, the remainder being uranium.

A sampling of less than twenty wells showed that, with the exception of two exploration wells, none contained water exceeding 9 pCi/l gross alpha activity. Also, none contained concentrations of radium 226 greater than 3 pCi/l, the recommended public drinking water limit.<sup>4</sup> Due to the small number of wells sampled until 1971, the 1971 sampling was our first indication of high levels of radium occurring in local ground waters.

#### Description of Sampling

Samples to determine radioactivity content of water supplies, both public and private, have been collected from wells and distribution systems in the uranium mining area over the past ten years, although most samples have been collected within the past three years. Most of the supplies have wells as their source(s) but a few municipal supplies use water from surface reservoirs. That data is included in this work where appropriate. Initial sampling of wells in the area consisted of selecting the wells closest to a uranium mill in Karnes County to determine if the mill was contributing to the radioactivity in the wells. More recent sampling has been conducted within five miles of known uranium deposits containing ore with more than 0.15%  $U_3O_8$ , municipal supplies in the region, and in areas where uranium mineralization is likely. In one area, seven miles in diameter, all fifty-six known water wells were sampled.

The data examined in this work is not considered as being representative of all well water in South Texas, but as being representative of well water within the vicinity of the significant uranium deposits. The breakdown of number of supplies sampled by county is presented in Table I., and is indicative of this deliberate bias. No other known bias in the data is present.

Table I.  
Number of Supplies Sampled by County

Atascosa	2
Bee	2
Brooks	1
Cameron	1
Duval	5
Gonzales	2
Harris	1
Jim Hogg	3
Jim Wells	2
Live Oak	66
Karnes	44
Nueces	1
Webb	4
Uncertain	3

#### Analysis of the Data

The individual data for all 137 supplies sampled containing the county, total dissolved solids, sulfate, bicarbonate, gross alpha, gross beta, and radium 226 concentrations, when known, is too lengthy to be presented here, but is available from the authors upon request.

Radioactivity data was obtained from two sources: gross alpha and beta and some of the radium data are from the Texas State Department of Health Laboratory Section, other radium 226 data are from the Environmental Protection Agency and Public Health Service. Texas radium data is total radium as determined by precipitation with a barium carrier and counting in an internal proportional counter. Data from the Federal laboratories were reviewed using the radon emanation technique. Gross alpha and gross beta results are obtained by evaporating up to 150 milliliters of the sample (depending upon total dissolved solids content) on a planchet and counting in internal proportional and low-beta anti-coincidence counters. Radium 226 and strontium 90 sources are used as standards. The results were corrected for sample self-absorption.

The data, which covers almost ten years of sporadic sampling on a few supplies, has shown no discernable trend with time on the more frequently sampled supplies, although there is not enough older data to completely refute this possibility in all supplies. Most supplies have been sampled only once.

The data for those supplies with highest alpha activities (over twenty picocuries per liter) is presented here in Table II. All of these supplies are wells. In Figure 2, Distribution of Gross Alpha Values, the number of supplies having a gross alpha value within the range indicated on the abscissa is plotted against gross alpha activity.

Table 11.  
Radioactivity of Wells with Highest Alpha Activity

Well	Gross Alpha (pCi/l)	Gross Beta (pCi/l)	County
1	43	50	Live Oak
23	23	33	Live Oak
26	31	68	Live Oak
45	320	580	Live Oak
59	35	57	Karnes
61	33	60	Gonzales
64	42	82	Live Oak
67	68	135	Gonzales
86	21	56	Karnes
90	23	71	Karnes
95	603	3000	Unknown
96	3111	1572	Unknown
97	144	146	Karnes
100	111	115	Live Oak
102	54	121	Karnes
107	22	29	Karnes

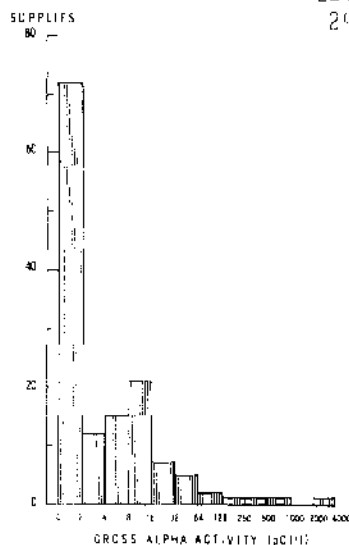


Figure 2 - Distribution of Gross Alpha Values

A modified geometrical progression was used in Figure 2 because of the range in values of the data. Figure 3, Distribution of Gross Beta Values, is also presented in this manner. Neither plot indicates the data came from a single statistical population.

The theory of the uranium deposition in the area requires several factors to be present for uranium to concentrate in the earth. One of these is a change in soil chemistry such as occurs due to oxidation of the surface layers.<sup>1</sup> The boundary between oxidized layers on the surface of the earth and lower reduced layers of the ground in this part of the State occurs typically between 50 and 200 feet. This fact may be a significant factor in increasing the radioactivity of the water. An evaluation of this possibility is beyond the scope of this investigation, but should be pursued by other investigators.



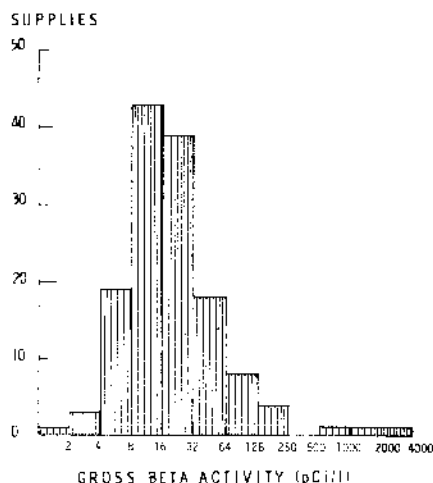


Figure 3 - Distribution of Gross Beta Values

Of 133 wells, the depth was known for 29, partial chemical water quality data for 28, and radium 226 (or total radium) for 15. Plots of radium 226 concentration, gross alpha and gross beta activities of water supplies versus several parameters were examined. There was no correlation detected for activity (alpha, beta, or radium) versus total dissolved solids, sulfates, or bicarbonate. These plots, available upon request, are not presented here. There was, however, a trend noted between gross alpha and a similar, though less distinct trend for gross beta versus depth. No well deeper than 400 feet had a gross alpha activity greater than 3 pCi/l, and the percentage of wells with gross alpha values greater than 3 pCi/l decreases with increasing well depth (see Figure 4).

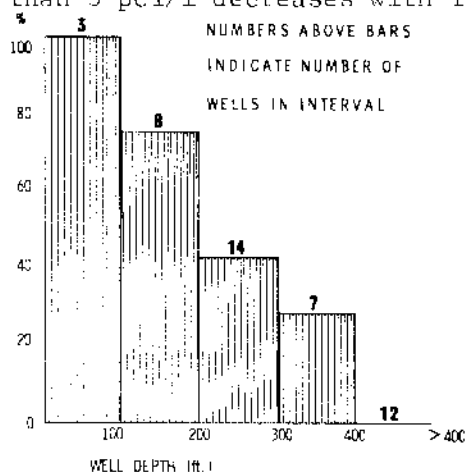


Figure 4 - Percent of Wells with Gross Alpha Activity Greater Than 3 pCi/l Versus Well Depth.

Radium 226 data on wells which are used for human consumption are presented in Table III. Included in this table are estimated body burdens of adults who have been consuming the water from these wells for extended periods.

The Federal Radiation Council estimations in their Report No. 2, namely that the body burden of people exposed to constant levels of radium 226 is at most fifty times their daily consumption, have been used to calculate these body burdens.

Table III.  
Radium 226 Data for Wells Used for Human Consumption

<u>Well No.</u>	<u>226Ra (pCi/l)</u>	<u>Estimated Body Burden (pCi)</u>	<u>% of RPG</u>	<u>County</u>
1	110	12100	404	Live Oak
3	0.8	88	3	Live Oak
27	0.2	22	1	Live Oak
32	4	440	15	Live Oak
45	5	550	18	Live Oak
50	0.5	55	2	Live Oak
78	2.4	264	9	Live Oak
85	0.4	44	1	Karnes
87	2.1	231	8	Karnes
100	153	16800	560	Live Oak

Two wells produce water with radium concentrations resulting in individuals exceeding the Radiation Protection Guide (RPG). The owners of these wells have been notified of this situation and have been advised to quit consuming the water.

#### Summary and Conclusions

High radium 226 concentrations in well waters in South Texas were discovered in 1971 and a considerable amount of data on the ground waters in the vicinity of the uranium deposits has been collected. In most wells, alpha and beta radioactivity is low, however in 25 wells shallower than 300 feet, 15 had alpha concentrations in excess of 3 pCi/l.

In two wells, used for drinking water supplies for humans high levels of radium 226 were found. These high concentrations, well above those allowable in licensed industrial discharges, and very much in excess of recommended drinking water standards have resulted in individual exposures probably exceeding the RPG and raise concern for other users of ground water in the area. While less than two percent of the supplies have radium concentrations in excess of occupational exposure limits, it is our opinion that no one should use water of this character routinely. Attempts are being made to screen supplies in the area most likely to contain elevated radioactivity. Work in progress in the area includes sampling every source of public water supplies and selected private wells used as the principle source of drinking water for humans.

Uranium is known to occur in other parts of Texas and in many other regions of the world, with a potential impact on human exposure to radiation. Uranium producing areas should be evaluated by the responsible health agency in sufficient detail to exclude the possibility of radiation exposures beyond acceptable standards.

#### Acknowledgements

Numerous persons in the uranium industry, State, local, and Federal governments, have given valuable assistance in the collection of data and information on radioactivity in the waters of South Texas. Especially worthy of comment are personnel of the Conquista Project, Dalco Oil Company, Susquehanna-Western, Inc., Texas State Department of Health Laboratory, and Texas State Department of Health Public Health Region 10. The authors would like to take this opportunity to thank them for their help and advice.

### References

- 1) Eargle, D. H., Hinds, G. W., and Weeks, A. M. D., 1971, Uranium Geology and Mines, South Texas, Guidebook 12, Bureau of Economic Geology, University of Texas at Austin.
- 2) Wukasch, M. C., and Cook, L. M., "Environmental Surveillance in South Texas", The Natural Radiation Environment II Proceedings, in press. Houston, Texas.
- 3) Texas State Department of Health, 1972, Texas Regulations for Control of Radiation, Division of Occupational Health and Radiation Control.
- 4) U. S. Department of Health, Education, and Welfare, 1962, "Public Health Service Drinking Water Standards", Revised 1962, Public Health Service Publication No. 956, United States Government Printing Office.
- 5) Federal Radiation Council, "Background Material for the Development of Radiation Protection Standards", Report No. 2, September 1961.

137Cs AND 134Cs DISTRIBUTION IN SEDIMENT, WATER,  
AND BIOTA OF THE LOWER HUDSON RIVER AND THEIR  
DOSIMETRIC IMPLICATIONS FOR MAN

M. E. Wrenn, S. M. Jinks, N. Cohen, and L. M. Hairr  
Institute of Environmental Medicine  
New York University Medical Center  
New York, New York

Abstract

Since 1964, the magnitudes and distributions of natural, fallout, and reactor-produced radionuclides present in the Lower Hudson River Estuary have been under study.

Concentrations of the major radionuclides released to the estuary in liquid waste from a pressurized light water reactor have been measured in samples of water, sediment, and biota. These measurements have been combined with information related to site characteristics and human use of the environment to obtain estimates of the magnitudes and pathways of human radiation exposure from routine power reactor operation.

137Cs and 134Cs are presently the "critical" radionuclides in the lower estuary, and consumption of fish is the most important route of exposure for man. Based on measurements to date, releases from a PWR of 20 Ci/yr each of 137Cs and 134Cs would result in an annual total body dose to an avid local fish eater of about 0.2 mrem/yr.

Introduction

The radioecology of the Lower Hudson River Estuary has been under investigation by the Institute of Environmental Medicine since 1964. This research has been directed toward the identification of the types, sources, and magnitudes of radioactive materials present in the abiotic and biotic components of the estuary.

The region of the river which has been most intensively studied is centered about Indian Point, which is located 42 miles above the southern end of Manhattan and is the site of Consolidated Edison's Indian Point Nuclear Power Station (Fig. 1). Frequent upstream sampling is also conducted at Chelsea and Hyde Park, and selected samples have been collected as far south as the George Washington Bridge. Sample types include water, sediments, and the more abundant biota including fish, rooted plants, and plankton. Analysis for radionuclide content has been conducted by several techniques including gamma spectrometry, beta proportional counting, and liquid scintillation counting. Details of the analytical procedures employed are reported elsewhere.<sup>1-4</sup>

The spectrometric system employed for the quantitation of gamma emitting radionuclides consists of a 10 cm x 10 cm NaI(Tl) well geometry crystal coupled to a 512 channel pulse height analyzer (Nuclear Data). Estimates of the activities of gamma-emitting radionuclides in the sample are obtained by a computerized least squares fitting of standard reference spectra to the sample spectrum. Sensitivities obtainable with this analytical system have been found to be superior to those obtainable with typical large GeLi systems for all sample media except sediment.<sup>2</sup>

## Radioactivity in the Hudson

Radionuclides which have been identified in the lower Hudson occur from natural sources, from fallout of weapons testing debris, and from liquid waste releases at a pressurized light water reactor (Indian Point # 1).

The predominant natural radionuclides are  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Ra}$ . Potassium-40, which is present as a fixed proportion of all potassium in the environment, accounts for a major portion of the radioactivity in all sample types (Fig. 2). Radium-226 and  $^{228}\text{Ra}$  occur in their highest levels in bottom sediments, where they are each found at a concentration of about 1.0 pCi/g dry weight.

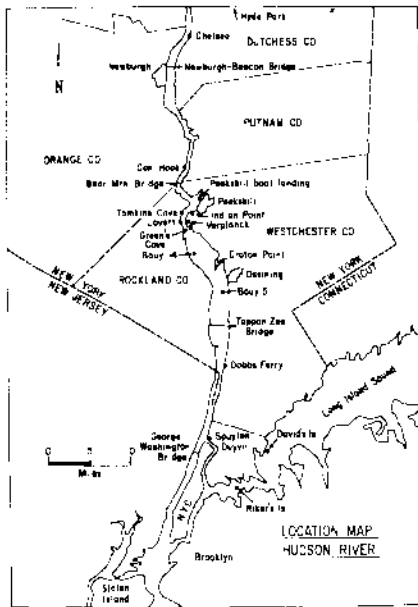


Fig. 1 Hudson River and Sampling Locations.

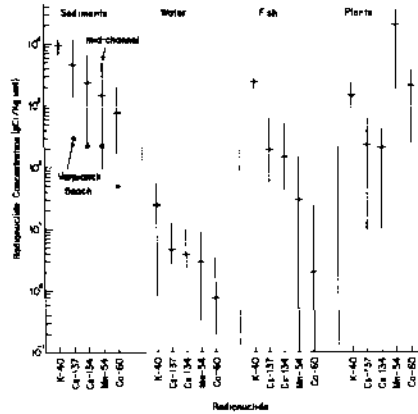


Fig. 2 Means and Ranges of Radionuclide Concentrations Observed in Sediment, Water and Biota at Indian Point (1971-1972).

Major introductions of manmade radionuclides to the Hudson River occurred as a result of the large scale weapons testing by the U. S. and U. S. S. R. during 1961-1962. Radionuclides which were introduced in significant quantities from this source include  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^3\text{H}$ ,  $^{144}\text{Ce}$ ,  $^{141}\text{Ce}$ ,  $^{106}\text{Ru}$ ,  $^{103}\text{Ru}$ ,  $^{131}\text{I}$ ,  $^{95}\text{ZrNb}$ ,  $^{125}\text{Sb}$ ,  $^{140}\text{Ba}$ ,  $^{140}\text{La}$ ,  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{65}\text{Zn}$ , and  $^{55}\text{Fe}$ . Due to the Nuclear Test Ban Treaty, fallout levels have greatly decreased since 1963. As a result, those radionuclides with relatively short half-lives are now, in most cases, below detectable levels in samples collected from the river, and at present the major fallout components are  $^{137}\text{Cs}$  and  $^3\text{H}$ .

The major intermediate to long-lived activity discharged in liquid waste from the light water reactor at Indian Point consists of fission products ( $^{137}\text{Cs}$ ,  $^3\text{H}$ ), and nuclides produced by activation ( $^{134}\text{Cs}$ ,  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{58}\text{Co}$ ). The relative activities of these radionuclides in liquid reactor waste during 1971 indicate that the two cesium isotopes,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ , comprise the major portion of the non-tritium activity released.<sup>5</sup> Cesium-134 has not been a significant component of fallout, and its occurrence in the Hudson River environment is, therefore, indicative of the presence of radioactivity discharged from Indian Point # 1. During 1971, a total of 22.5 Ci of  $^{137}\text{Cs}$  and 16.5 Ci of  $^{134}\text{Cs}$  were released to the Hudson in liquid waste discharges from the reactor.

### Evaluation of the Radiation Dose to Man from Indian Point # 1 Aquatic Releases

Observed environmental distributions of radionuclides in the Hudson have

been combined with information relating to specific site characteristics and human use of the environment in order to assess the important pathways and magnitudes of human radiation exposure attendant to low level liquid waste releases to an estuary from an operating light water reactor. Based upon measurements of the radionuclide content of environmental media in the vicinity of Indian Point, estimates of the whole-body doses resulting from reactor discharges and from fallout are presented for the pathways in Table 1. The pathways can generally be divided into those resulting in internal exposure through consumption of food and water, and external exposures from radionuclides present in water and shoreline soils.

Table 1. Summary of Estimated Doses to Man from Reactor\* and Fallout Radionuclides in the Hudson River (1971-1972)

Pathway	Annual Whole-Body Dose (mrem/yr)		% Reactor Dose from Radiocesium
	Reactor	Fallout	
Fish Consumption 30 g/day	0.19	0.01	98%
Sunbathing 200 hr/yr	0.06	0.01	80%
Swimming 200 hr/yr	0.01	0.001	65%
Drinking Water (2.2 l/day)	0.003	0.08	90%

\*Indian Point # 1 Nuclear Power Station

The radiation doses to man in the Indian Point area resulting from natural sources has also been determined. These doses are based on 19 measurements with a pressurized ion chamber at a number of locations in the vicinity of Indian Point. They can be categorized into those resulting from external exposure to cosmic and terrestrial radiations (~94 mrad/yr), and those caused by radioactivity occurring naturally within the body (~20 mrad/yr). The total mean natural radiation dose amounts to more than 114 mrad/yr, which for purposes of this paper will be considered equal to 114 mrem/yr.

### Fish Consumption

Since no edible shellfish populations exist in the Hudson and the aquatic plants present are not consumed by man, fish consumption is the primary pathway by which radionuclides can be recycled to man through the aquatic food web.

Over 200 samples of Hudson River fish have been collected and analyzed during 1971 and 1972. These samples included 20 different species, ranging from anadromous fish such as the striped bass and shad to indigenous types such as white perch, sunfish, catfish, and largemouth bass.

Indigenous fish species in the Indian Point area have been observed to contain about fourfold higher concentrations of reactor-produced radionuclides than those species migrating into the river to spawn. Over 90% of the content of manmade gamma-emitting radionuclides in these fish is comprised by the two cesium isotopes,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  (Fig. 2). It is interesting to note that fish discriminate against the uptake of manganese and cobalt isotopes from vegetation upon which they feed. This phenomenon makes these nuclides insignificant to considerations of human radiation exposures by this route. Concentrations of  $^{54}\text{Mn}$  and  $^{60}\text{Co}$  in fish are 2 to 3 orders of magnitude less than the concentrations occurring in vegetation. This observation is in direct contradiction to the popular assumption that all aquatic contaminants are, in effect, concen-

trated by transfer to a higher trophic level (a process generally referred to as biomagnification). Recent scientific assessments of the existing literature indicate that, in fact, biomagnification of metals is probably an infrequent occurrence in nature.<sup>6</sup>

The dose to man from fish consumption was calculated using the mean radionuclide concentrations observed in indigenous fish at Indian Point during 1971 ( $^{137}\text{Cs} = 219 \text{ pCi/kg}$ ;  $^{134}\text{Cs} = 182 \text{ pCi/kg}$ ), and using the ingestion/dose rate model of ICRP 2. The year 1971 was the year in which the highest concentrations of reactor-produced radionuclides in fish were observed since observations began in 1964. The total variation in radiocesium content among fish collected at Indian Point was approximately a factor of 10; with maximum concentrations (3-5 times the mean value) observed in fish caught at the reactor outfall. The dose calculation assumes a continuous daily ingestion of 30 grams of fish (about 15% of total meat intake for an adult), and is considered to give a conservative estimate of the dose since year-round consumption of fish caught solely in the Indian Point area is unlikely. Of the total dose calculated to result from fish consumption (0.2 mrem/yr), about 0.08 mrem is attributable to the presence of  $^{137}\text{Cs}$  and about 0.12 mrem to  $^{134}\text{Cs}$ .

### Drinking Water

The water in the vicinity of Indian Point is sufficiently brackish throughout the year to prevent its use as a potable water supply. Estimates of the doses from drinking water have, therefore, been calculated using radionuclide concentrations measured at the Chelsea pumping station, which is located 22 miles upstream from Indian Point. This station serves as an auxiliary water supply for New York City and is only 1.5 miles north of the public water supply closest to Indian Point, the Veteran's Administration Hospital at Castle Point.

Due to the hydrological characteristics of the estuary, the concentrations of reactor-produced radionuclides in water tend to decrease exponentially with distance north of Indian Point. Thus, as shown in Table 1, the reactor contribution to radiation dose from consumption of water at upstream, freshwater sites is minute, and insignificant in relation to exposures by other pathways.

### Swimming

Although the water near the reactor site is not suitable for drinking, it is used for recreational purposes. The maximum external exposure from radionuclides present in water can be expected to occur to persons immersed while swimming.

Calculation of the swimming dose was made using the mean radionuclide concentrations measured in water samples collected continuously at Verplanck (Fig. 2), which is about one mile downstream from the reactor discharge and represents the nearest publicly used beach. Since the water samples from Verplanck are collected continuously, they are assumed to provide reasonable estimates of average radionuclide concentrations occurring at that site.

### Sunbathing

Radiocesium accumulations in near-shore submerged sediments have been determined by analysis of cores collected during 1972 in the vicinity of Indian Point. Cumulative activities ranged between 180 and 545  $\text{mCi/km}^2$  for  $^{137}\text{Cs}$  and between 40 and 405  $\text{mCi/km}^2$  for  $^{134}\text{Cs}$ .

Estimates of the total accumulations of reactor radionuclides in sediments at the river shoreline have also been obtained by analysis of core samples collected at six selected sites north and south of the reactor outfall.

Cumulative depositions of radiocesium per unit release from Indian Point # 1 have been calculated on the basis of activity measured in submerged and shoreline sediments 1-2 miles south of the reactor outfall. For submerged sediment approximately 15-20  $\text{mCi/km}^2$  was accumulated per Ci released, while for

shoreline sediments the value was only about 2 mCi/km<sup>2</sup>-Ci.

The dose to a sunbather at the nearest public beach was calculated using the observed accumulations and depth distributions of radionuclides in shoreline sediment at this site. Since the emissions from radionuclides accumulated in submerged sediments are effectively shielded by the overlying water, calculations of the dose to man were based on measurements of the shoreline accumulations.

### Radiation Dose to Biota

Radiation standards for the release of radioactive materials to the environment are based upon limiting the radiation dose to man. This has been presumed to be a reasonable philosophy in view of the fact that man is one of the most radiosensitive organisms known. However, it is relevant to ask whether or not biota are being exposed to higher doses than man, and whether or not these doses are radiobiologically significant.

Accordingly, from the measurements of radionuclides in biota, water, and sediment, it is possible to calculate the radiation dose being received by several classes of aquatic biota present in the vicinity of Indian Point. The results of such calculations for fish and benthos are shown in Table 2, where it can be seen that the largest exposure, both natural and manmade, occurs to organisms immersed in the top portions of local sediments. The additional dose rate from the reactor is about equal to that from the natural background in these sediments, about 180 mrad/yr. Accordingly, benthic organisms (including fish) might accumulate doses on the order of 0.2 of a rad/yr. Auerbach (1970) has suggested that the lowest dose rates at which observable biological effects can be noted are approximately one rad/d, or roughly 400 rads/yr.<sup>7</sup> The dose to biota in the vicinity of Indian Point is at least two thousandfold below this. Thus, the results of environmental measurements to date give good reason to believe that no perceptible deleterious effects on the biota will occur as a result of the radiological releases at Indian Point.

Table 2. Estimated Doses to Fish and Benthos from  
Reactor Releases and from Natural Potassium

Biota	Method of Exposure	Dose Rate (mrad/yr)	
		K-40	Reactor
Fish	Internal	17	3
Fish	External - water	0.2	0.2
Benthos	External - sediments	100	180

### Summary

Examination of the magnitudes of the estimated doses indicates that consumption of fish represents the "critical pathway" for radiation exposure of man due to routine aquatic discharges from a light water reactor on an estuary. Furthermore, as indicated in Table 1, the cesium (<sup>137</sup>Cs and <sup>134</sup>Cs) isotopes deliver the majority of the dose by all exposure pathways, and are especially important in determining the dose from fish consumption.

The total whole body dose to the maximally-exposed individual from an annual liquid waste discharge of 39 Ci of <sup>137</sup>Cs and <sup>134</sup>Cs (1971) is estimated to be about 0.2 mrem/yr.<sup>1</sup> This dose amounts to .04% of the internationally accepted dose limits (ICRP - 500 mrem/yr) and about 0.2% of the natural background dose in the reactor area.

Radiation doses to biota resulting from reactor liquid waste discharges to the Hudson estuary are largest for organisms inhabiting the surface layers of local sediments. These doses, which may be as high as 0.2 rad/yr, are



radiobiologically insignificant. Thus, it appears that discharge of radioactive waste to the Hudson River Estuary from a light water reactor in operation more than a decade neither presents any limitation upon human use of the estuary, nor constitutes a hazard to the river biota.

#### References

1. Wrenn, M. E., et al., Radiocesium Distribution in Water, Sediment, and Biota in the Hudson River Estuary from 1964 Through 1970, Proc. 3rd National Symposium on Radioecology, USAEC CONF-710501-P1, 334-343 (1971).
2. Wrenn, M. E., et al., Natural Activity in Hudson River Estuary Samples and its Influence on the Detection Limits for Gamma Emitting Radionuclides Using NaI Gamma Spectrometry, Proc. 2nd Internat'l Symposium on the Natural Radiation Environment, Houston, Texas (1972).
3. Lentsch, J. W., et al., Manmade Radionuclides in the Hudson River Estuary, Proc. 5th Annual HPS Mid-year Topical Symposium, Idaho Falls (1970).
4. Kneip, T. J., et al., Trace Elements, Radionuclides, and Pesticide Residues in the Hudson River, Proc. of FAO Technical Conference on Marine Pollution and its Effects on Living Resources and Fishing, Rome, Italy (1970).
5. Consolidated Edison Company of New York, Inc., Indian Point Station Semi-Annual Operations Reports, Submitted to the USAEC.
6. Kneip, T. J. and G. J. Lauer, Trace Metal Concentration Factors in Aquatic Ecosystems, Progress in Analytical Chemistry, Vol. 5, Plenum Press, New York, New York (in press, 1973).
7. Auerbach, et al., Ecological Considerations in Reactor Power Plant Siting, Proc. of the IAEA Symposium on Environmental Aspects of Nuclear Power Stations, New York (1970).

AN INVESTIGATION INTO THE REASONS FOR A LACK OF A  
TROPHIC LEVEL EFFECT FOR  $^{137}\text{Cs}$  IN FUNDULUS HETEROCLITUS  
AND ITS FOOD IN THE HUDSON RIVER ESTUARY

John Mauro\* and McDonald E. Wrenn  
Institute of Environmental Medicine  
New York University Medical Center  
New York, New York

Abstract

A detailed investigation into the behavior of radiocesium in the food chain of one species of fish (Fundulus heteroclitus) in the Hudson River Estuary revealed the probable reasons for the lack of a trophic level effect for  $^{137}\text{Cs}$  in the food chain of Fundulus and perhaps that of most fish in the Hudson River Estuary.

A series of laboratory experiments revealed that the elimination rate of  $^{137}\text{Cs}$  was 1/5 that of potassium in Fundulus, thereby creating the potential for the trophic level effect. However, since the fish were not in equilibrium with their source of  $^{137}\text{Cs}$  and the ability of the fish to assimilate  $^{137}\text{Cs}$  relative to that of potassium was markedly reduced due to the presence of sediment in the ingested foods, the trophic level effect did not occur.

Introduction

Radiocesium has been shown to be the critical radionuclide in the liquid effluent of one light water reactor located on an estuary and the ingestion of fish has been shown to be the critical pathway of exposure to man.<sup>1</sup> It is reasonable to believe that radiocesium may be the critical nuclide (in the sense of ICRP 7) for many light water reactors because of the high escape rate coefficient in relation to other radionuclides such as  $^{90}\text{Sr}$ .<sup>2</sup> Accordingly, a thorough understanding of the ecological behavior of radiocesium in aquatic ecosystems is needed in order to predictively assess the dose to man due to a given release of radiocesium to a given aquatic environment.

Many investigators have observed a systematic threefold increase in the concentration of radiocesium between successive trophic levels of aquatic food chains in freshwater environments.<sup>3-8</sup> If this trophic level effect were observed to occur under all or most circumstances, it would make the accurate prediction of the concentration of radiocesium in aquatic biota a relatively trivial matter. Some other investigators have not always observed such a trophic level effect. In particular, Nelson studying the accumulation of  $^{137}\text{Cs}$  in fish in the Clinch River, has noted that threefold trophic level enrichment of the  $^{137}\text{Cs}/\text{K}$  ratio did not occur.<sup>9</sup> Studies of radiocesium and potassium performed in the Hudson River Estuary showed that biota in this ecosystem did not

---

\*Now with Ebasco Services, Inc., New York.

These investigations have been supported by grants from the U.S. Public Health Service, the New York State Department of Health, Consolidated Ediaon Company, Inc., and is part of a center program supported by NIEHS, Grant No. ES 00260.

exhibit a trophic level effect.<sup>1,10</sup> This paper presents the results of investigations into the accumulation of radiocesium and potassium in the food chain of one species of fish, Fundulus heteroclitus, in the Hudson River Estuary, and the results are used to explain the reasons for the lack of a trophic level effect.

Based on our review of the literature, we conclude that the following set of conditions must exist in an ecosystem in order for a trophic level effect to exist:

- 1) The fish must obtain essentially all of its radiocesium and potassium from its food organisms.
- 2) The activity of radiocesium and potassium in the fish must be in equilibrium with that in the fish's food organisms.
- 3) The fractional assimilation of radiocesium and potassium by the fish from its food organisms must be the same.
- 4) The elimination rate of radiocesium from the fish must be 1/3 that of potassium.
- 5) The potassium concentration of all organisms of the fish's food chain must be similar.

A series of field and laboratory studies conducted during 1971 and 1972 were performed in order to determine which among the above conditions were responsible for the observed lack of a trophic level effect in Fundulus heteroclitus in the Hudson River Estuary.

#### Materials and Methods

##### Field Studies

Fundulus heteroclitus, aquatic plants, invertebrates, sediment and water were collected on a biweekly basis from the shore area at site 11W1 (see Fig. 1) from the Hudson River Estuary during the growing seasons of 1971 and 1972. The samples were reduced in volume and analyzed for <sup>40</sup>K and <sup>137</sup>Cs by gamma spectrometric analysis described elsewhere.<sup>1</sup>

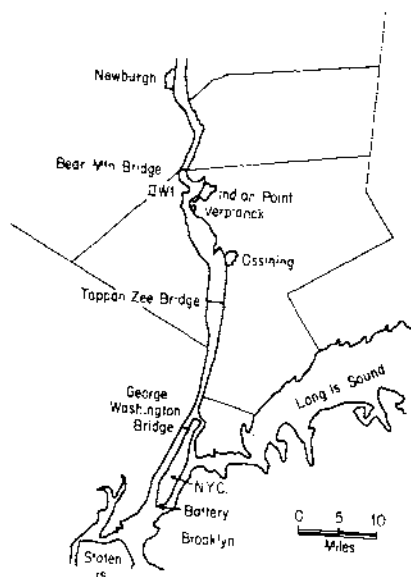


Fig. 1 The Lower Hudson River Estuary

Prior to analysis, the stomachs of the fish were removed and the contents weighed and identified in order to determine the feeding habits of the fish.

##### Laboratory Studies

###### Stomach Clearance Rate

Well-fed Fundulus held in the laboratory and also Fundulus obtained from the river were held unfed in the laboratory. The fish were then periodically sacrificed and the stomach content weight determined. This yielded a stomach clearance rate which was used, along with the stomach content analysis, to

determine the daily feeding rate of *Fundulus* in the Hudson River Estuary.

Elimination Rate of Radiocesium

*Fundulus* were fed various food items which were labeled with  $^{137}\text{Cs}$ . Immediately after labeling, the fish were live counted and placed into aquaria. These fish were then periodically live counted yielding the fractional assimilation of radiocesium by *Fundulus* from its various food items and also the biological half life of  $^{137}\text{Cs}$  in the fish. (The procedure followed was identical to that described in detail by Kevers.)<sup>4</sup>

Elimination Rate of Potassium by *Fundulus*

*Fundulus* were force fed  $^{40}\text{K}$  and then individually placed into aquaria. Periodically the aquaria water was removed and the  $^{40}\text{K}$  in it, due to excretion by the fish, was collected on ion exchange resins. The resins were then analyzed for  $^{40}\text{K}$  by gamma spectrometry. This yielded the biological half time of potassium in *Fundulus*. (This procedure is described in detail by Kolehmainen.)<sup>6</sup>

The Uptake Rate of  $^{137}\text{Cs}$  by *Fundulus* from Direct Sorption from Water

*Fundulus* were held unfed in Hudson River water of various salinities and containing  $1\ \mu\text{Ci/l}$  of  $^{137}\text{Cs}$ . The fish were periodically washed and then live counted yielding the uptake rate of radiocesium by direct sorption from water.

The above series of experiments quantified the uptake and elimination rates of  $^{137}\text{Cs}$  in *Fundulus*, thereby enabling one to identify the causes for any lack of a trophic level effect which might be found to occur.

Results and Discussion

Table 1 presents the  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activity observed in *Fundulus heteroclitus* and its food organisms collected from the Hudson River in 1971 and 1972.

Table 1. The Concentration of  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in *Fundulus heteroclitus* and its Food Organisms During 1971 and 1972 pCi/g Ash Weight

	<i>Fundulus heteroclitus</i>				<i>Gammarus</i>			
	K-40	Cs-137	Cs-137	# of Samples	K-40	Cs-137	Cs-137	# of Samples
1971	33.9±5.0	4.2	.13	10	-	7.4	-	2
1972	32.2±6.8	3.1	.09	6	25	3.2	.13	3
		Filamentous Algae				Higher Plants		
1971	40±12	22.6	.56	3	60±36	11.4	.13	6
1972	75±16	6.3	.08	8	77±35	4.2	.05	13

There is an obvious lack of a trophic level effect. The following results reveal which among the 6 conditions presented in the introduction were responsible for the absence of a trophic level effect.

Condition 1

The fish can obtain radiocesium from its foods, from the ingestion of sediment, and from direct sorption from water. The uptake rate from food is the product of the mass of food eaten per day, the  $^{137}\text{Cs}$  activity of the food and the fractional assimilation of  $^{137}\text{Cs}$  from the food by the fish.

Stomach content analyses performed on fish which were collected during the months of May through September revealed that the fish ingested 14% of its body weight per day, that 52% of the ingested food was filamentous algae, 29% was *Gammarus*, 8% was higher plants and less than 1% was sediment. Table 2 presents the average activity observed in these items during 1971 and 1972 and the fractional assimilation of  $^{137}\text{Cs}$  by *Fundulus* from these items. The assimilation of  $^{137}\text{Cs}$  was markedly reduced due to the presence of sediment along with the ingested material.

The results of the series of laboratory experiments where *Fundulus* obtained  $^{137}\text{Cs}$  solely via direct sorption from water revealed that among 12 fish ranging in size from 2 to 12 grams and held in water ranging in salinity from 1 to 140 ppm potassium the uptake rate was  $.057\pm.027$  of the concentration in the water per day.

Table 3 summarizes the above studies showing the rate of uptake of radio-

cesium by Fundulus from its various sources during 1971. Almost all of the radiocesium in Fundulus was from the ingestion of food organisms and very little was from the direct sorption from water or the ingestion of sediment. It is also believed that the potassium content of fish originated from the fish's food items.<sup>11</sup> Therefore, Condition 1 for the existence of a trophic level effect is fulfilled in Fundulus in the Hudson River Estuary.

Table 2. The Average Activity of <sup>137</sup>Cs Observed in Water and Food Items of Fundulus heteroclitus Taken from the Hudson River Estuary During 1971 and 1972 and the Fractional Assimilation of <sup>137</sup>Cs by Fundulus from These Items

Item	1971		1972		Fractional Assimilation
	pCi/Kg Wet Wt.	# of Samples	pCi/Kg Wet. Wt.	# of Samples	
Filamentous					.084 <sup>±</sup> .058*
Algae	616	3	218	8	.79 <sup>±</sup> .16
Higher Plants	340	6	131	13	.084 <sup>±</sup> .058
<u>Gammarus</u>	70	2	36	3	.834 <sup>±</sup> .077
Sediment	11452	5	10322	11	.031 <sup>±</sup> .017
Water	.78	5	.30	17	-

\*This value is the assimilation of <sup>137</sup>Cs by Fundulus from labeled filamentous algae had sediment associated with it.

Table 3. The Rate of Uptake of <sup>137</sup>Cs by Fundulus heteroclitus from its Various Sources During 1971

Source	Uptake Rate (pCi/Kg-day)	% of Total
Filamentous algae	2.4	61
Higher plants	.2	5
<u>Gammarus</u>	1.2	29
Sediment	.13	3
Water	.04	1.2

#### Condition 2

The data on the observed concentrations of <sup>137</sup>Cs in Fundulus, filamentous algae, Gammarus, and water taken at a single site in the Hudson River during 1971 and 1972 are shown in Fig. 2. It can be seen that the variation of radiocesium with time is much less in Fundulus than in any of the other media. This is reasonable in view of the relatively long half-life of <sup>137</sup>Cs in Fundulus, equivalent to about 250 days as determined from elimination rate experiments. Others have shown that filamentous algae come into equilibrium with water in days and that Gammarus equilibrates in a period of weeks.<sup>12-14</sup> In this location, <sup>137</sup>Cs in water was a result of the addition of runoff into the watershed from the accumulated <sup>137</sup>Cs in soil from fallout and from the <sup>137</sup>Cs introduced from the Indian Point power plant across the river. By the time releases from Indian Point had diffused across the river, several tidal cycles normally would have occurred and, accordingly, fluctuations in the water over any several-day period would not be too great. However, from Fig. 2 one can see that the fluctuations in water were simply greater than those observed in Fundulus. The water samples are the results of grab samples taken biweekly during this period. Because the river is tidal and the flushing time is in excess of one month at this location during the late summer, the grab samples are believed to represent a reasonable average over a period of some days. It is, accordingly, reasonable to conclude, from the data presented in Fig. 2, that the food organisms upon which Fundulus feed reached equilibrium with the activity in the water relatively rapidly. On the other hand, it was clear that Fundulus had not reached an equilibrium activity because of its continued

increase in activity during the months of August and September of 1971 when  $^{137}\text{Cs}$  concentrations in water were relatively constant. It is, therefore, clear that Fundulus had not reached equilibrium with its environment and, moreover, the environment itself was fluctuating, the water and the food at different rates, but all generally much more rapidly than the radiocesium content of Fundulus. Accordingly, Condition 2, which requires that the fish be subsisting on food which is at equilibrium with water and that equilibrium conditions be obtained in the fish, was not satisfied in this environment. Because of the seasonal fluctuations of freshwater flow in the Hudson, and the variability of reactor releases with time, it is unlikely that a constant radiocesium level in water will ever be obtained over time periods exceeding 2 half lives (or about a year) in Fundulus, so that equilibrium conditions never exist in this system.

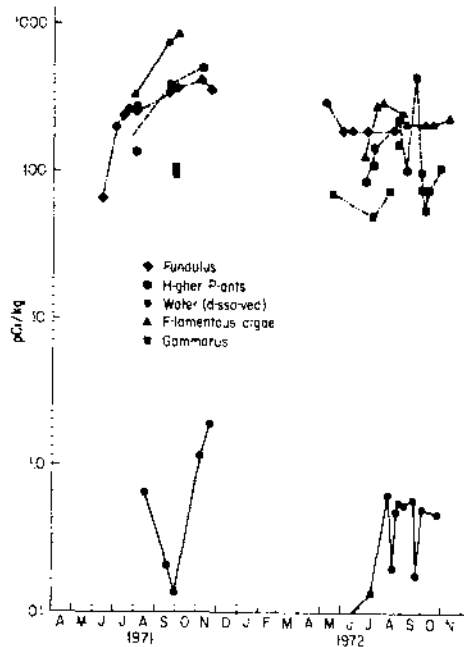


Fig. 2  $^{137}\text{Cs}$  Activity of samples collected during 1971 & 1972 from site 11W1 on the Hudson River Estuary.

### Condition 3

The fractional assimilation of radiocesium by Fundulus from plants has been found to be markedly reduced (perhaps 10-fold) due to the association of sediment with the ingested material (see Table 2). Perhaps due to the high potassium content of the sediment, the presence of sediment with the ingested plants does not reduce the assimilation of potassium from the plants. Therefore, the relative assimilation of  $^{137}\text{Cs}$  and potassium by Fundulus from its various foods is markedly different and, accordingly, Condition 3 is not fulfilled.

### Condition 4

Laboratory experiments revealed that the biological half life of  $^{137}\text{Cs}$  among 9 Fundulus ranging in weight from 2 to 12 grams and held in water containing 20 ppm potassium (salinity = 2 ppt) at a temperature of 20°C was  $265 \pm 45^*$  days. Under similar conditions, the biological half life of potassium in 12 Fundulus was  $50 \pm 18$  days. Therefore, the rate of elimination of potassium is about 5 times greater than that of cesium. Accordingly, Condition 4 is fulfilled, although the relative elimination rates differ somewhat from that assumed in the current "dogma".

\* $\pm$  1 standard deviation.

#### Condition 5

The fifth condition for existence of the trophic level effect is that the potassium concentration in the major food organisms of fish should be similar. Table 1 lists the concentrations of potassium observed in Fundulus and its major food items and in view of the fact that there exists only a two-fold difference among them, it is considered that this condition is satisfied.

#### Conclusions

Although the elimination rate of  $^{137}\text{Cs}$  was 1/5 that of potassium in Fundulus heteroclitus, the  $^{137}\text{Cs}$  was not consistent with that which would have been predicted by postulating a "trophic level effect" for the following reasons:

1) The  $^{137}\text{Cs}$  activity in Fundulus was not at equilibrium with that in its food organisms.

2) The assimilation of  $^{137}\text{Cs}$  from much of its food was markedly reduced relative to that of potassium. This was most likely due to the association of sediment with ingested plants which was effective in inhibiting uptake of radiocesium but not of potassium.

Clearly, the one phenomenon which was largely responsible for the lack of a trophic level effect in Fundulus in the Hudson River Estuary was the large reduction in the assimilation of  $^{137}\text{Cs}$  relative to that of potassium due to the association of sediment with ingested material.

#### References

1. Wrenn, M.E., et al., Radiocesium Distribution in Water, Sediment, and Biota in the Hudson River Estuary from 1964 Through 1970, Proc. 3rd Nat'l. Symp. on Radioecology, USAEC CONF-710501-P1, 334-343 (1971).
2. Eisenbud, M., Environmental Radioactivity, Second Edition (Lee, Douglas H. K.; Hewson, E. Wendel; and Okun, Daniel, Editors), Academic Press, New York (1973).
3. Gustafson, P.F., et al., Cs-137 in Edible Freshwater Fish, Nature 211: 343 (1966).
4. Kevern, N.R., Feeding Rate of Carp Estimated by a Radioisotopic Method, Transactions of the American Fisheries Society, 95 (4), 363 (1966).
5. Pendleton, R. C., Accumulation of  $^{137}\text{Cs}$  Through the Aquatic Food Web, In: Biological Problems in Water Pollution, Third Seminar (Aug. 13-17, 1962).
6. Kolehmainen, S.E., The Balance of  $^{137}\text{Cs}$ , Stable Cesium and the Feeding Rates of Bluegill (Lepomis macrochirus Raf.) in White Oak Lake, Ph.D. Thesis, Univ. of Tenn., ORNL-4445 (1969).
7. Hasanen, E. and J.K. Miettinen,  $^{137}\text{Cs}$  Content of Freshwater Fish in Finland, Nature 200: 1008 (1963).
8. Bigliocca, C., et al., Natural Cesium in Freshwater and Fishes, Octes In Symp. Internat'l. le Radioecologie, Centre D'Etudes Nucleaires de Cadarache du 8 du 12 Septembre 1969.
9. Nelson, D.J., Cesium,  $^{137}\text{Cs}$  and Potassium Concentrations in White Crappie and Other Clinch River Fish, Symp. on Radioecology, Edited by D.J. Nelson and F.C. Evans, CONF-67053 (1969).
10. Lentsch, J.W., et al., Manmade Radionuclides in the Hudson River Estuary, Presented at the 5th Ann. Health Physics Soc. Midyear Topical Symp. (1971).
11. Krough, A., Osmatic Regulation in Aquatic Animals, Dover Publications, New York (1965).
12. Harvey, R.S., Effect of Temperature on Sorption of Radionuclides on Blue-green Algae, In: Symp. of Radioecology, Ann Arbor, Mich., CONF-670503 (1967).
13. Garder, R. and O. Shulberg, On Experimental Investigation on the Accumulation of Radioisotopes by Freshwater Biota, Arch. Hydrobiol. 62: 50 (1966).
14. Kevern, N.R., et al., Biological Half Time of  $^{134}\text{Cs}$  in Carp and Two Aquatic Insects, ORNL-3697, 101 (1964).

REACTOR EXPERIENCES, WASTE MANAGEMENT  
AND ENVIRONMENTAL MONITORING

MEASUREMENTS OF THE DISPERSION OF GASEOUS RADIOACTIVE  
EFFLUENTS FROM AN OPERATING BOILING WATER REACTOR\*

William M. Carcy,<sup>2</sup> Lewis Battist, Warren E. Keene<sup>†</sup>  
American Environmental Control Systems, Inc.  
Gaithersburg, Maryland

Abstract

Measurements of off-site concentrations of the radioactive gaseous effluents from a BWR were correlated with measured meteorological parameters to define plume behaviour at a coastal site. Average ground concentrations were found to be in the  $10^{-9}$   $\mu\text{Ci/cc}$  range, while peak values were observed as high as  $10^{-7}$   $\mu\text{Ci/cc}$ . The data indicates that sea breeze fumigation and nocturnal fanning conditions significantly affect dispersion characteristics.

Introduction

The waste gas from a boiling water reactor consists primarily of the noble gases and iodine isotopes which can be determined by spectroscopic analysis of samples obtained at the steam jet air ejector and gross activity measurements in the stack. The problem thus arises as to how the effluent disperses under the given meteorological conditions prevailing at the time of release, and how to determine the average and peak concentrations at any distance from the discharge point. These data are necessary to restrict the overall population radiation exposure.

The concentration of the gaseous effluents can be discussed in terms of the effective emission height, the transport and dispersion of the effluent. The effective height is variable as wind speed, ambient temperature and lapse rate affect the height to which the effluent rises. The transport of the gaseous effluent is determined by wind direction and speed. Surface characteristics that produce turbulence as well as diurnal effects are influential in producing complex wind patterns which are difficult to describe or predict. Finally, the dispersion of the effluent is dependent on the wind speed and the horizontal and vertical eddies produced by mechanical shear and temperature differences.

Description of the Experiment

The objective of this endeavor was to provide measured values of the concentrations of radioactive gases at three locations near Millstone Point. (Ref. Table 1). The measured concentrations were correlated with meteorological data and stack emission data, to yield measured values of  $X/Q$  (concentration/emission rate,  $(\text{sec}/\text{m}^3)$ ) on a half-hourly, daily and monthly basis.

The Environmental Monitoring System, ERMS, measured the changes in the near ground concentrations of radioactive gases above the natural background. Three groups of data were obtained by the ERMS. The flow through alpha detector electrically precipitates radon and thoron decay daughter products in the atmosphere and measures subsequent alpha decays. The resulting



count rate is indicative of the changing levels of radon and its daughter products in the atmosphere.

The cosmic ray monitor is located above the beta/gamma detector. As the cosmic ray flux at sea level is primarily composed of muons, this detector is in anti-coincidence with the shielded beta/gamma detector. If a cosmic ray muon traverses both detectors, the count observed in the beta/gamma detector is negated. This cosmic ray monitor shield is also sensitive to external gammas. The count rate from the cosmic ray monitor is indicative of the cosmic ray flux and external gamma radiation above 0.4 Mev.

The third detector is the beta/gamma detector. Sampled air is continuously blown through this detector and if beta and/or low energy gamma emitters are in this air, this count rate is indicative of their concentration. Radon, thoron and the daughters of these isotopes also contribute to this count rate. This system is calibrated and capable of detecting concentrations of Kr-85 as low as  $10^{-9}$   $\mu$ Ci/cc.

Millstone Point Company personnel performed weekly analysis on primary coolant and steam jet air ejector samples to determine the composition of the gaseous effluent from the stack and provided "Q" value data.

Meteorological data was recorded at the 140 ft. tower near the stack by The Research Corporation of New England who provided all meteorological data. Wind speed and direction at the 140 ft. level were used to correlate with the measured values of near ground concentration at each site. Temperature measurements were recorded at 5', 70' and 140' in order that  $\Delta T/\Delta h$  could be determined.

#### Experimental Results

During the period of observation at the Millstone Point Boiling Water Reactor data similar to that shown in Figure 1 was collected. The alpha count rate which is proportional to the concentrations of the daughters of radon and thoron daughters in the atmosphere did not exhibit any drastic fluctuations as were observed in the beta/gamma chamber or the large enclosed anti-coincidence shield.

The observed peaks in the anti-coincidence shield were found to be due to the flux of external gamma radiation in the energy range of 0.4 Mev to 2.5 Mev and the character of these peaks strongly suggest that they are due to a plume traversing the detector. The flat region of the curve is the cosmic ray muon background.

The peaks in the beta/gamma count rate are due to the effect of this external gamma flux and the contribution of low energy photons and beta particles within the detector sample volume.

The data presented in summary form for the three sites, Figures 2 thru 4, were found to correlate with the direction of the wind as measured at the 140 foot level at the stack location and to exhibit a strong diurnal variation. The correlations are illustrated for the three sites in Figures 2 and 4.

The correlation with the "from" wind direction is more pronounced as the distance between the measurement point and the stack increases. The broadness of the histograms for Site I and Site II as well as the contributions when the wind was not in the direction of the measurement site, were found to correspond to stable meteorological conditions. This suggests, particularly for Site II data, that terrain effects are governing factors.

Table 1 summarizes the data for all sites. Peak and average concentrations at Site III were found to be lower than those at both Sites I and II. However, Site II exhibits higher peak and average concentrations than Site I. This may be explained by the occurrence of a sea breeze fumigation phenomena which will not occur frequently at the Site I distance. Average values for Sites I and II were found to be in the  $10^{-9}$  to  $10^{-7}$   $\mu\text{Ci}/\text{cc}$  range, while for Site III in the  $10^{-8}$   $\mu\text{Ci}/\text{cc}$  range.

This study though primarily concerned with a determination of the  $\chi/Q^*$  value, also measured the gamma flux dose, Table 1. The cosmic ray detector exhibits a flat response to gammas between 0.4 Mev to 2.5 Mev. The major contributors to this gamma flux are Cs-138 and Rb-88 with lesser contributions from Xe-138 and Kr-88. Considering that single scattered photons are also important, a good approximation to the dose rate is to select the mid-point of the energy range. The dose estimates shown in Table 1 could be refined to yield actual dose rates for each site.

#### Conclustions

The results of this study indicate that while long-term average concentrations and doses from the effluent of a BWR are low, short-term values appear as perturbations on a slowly varying natural background. Thus the long-term average values can be obtained by averaging easily measured peak values. This type of measurement is considered desirable as the micrometeorological aspects of a coastal site are the predominant influence on near neutrally bouyant plumes. While current technology is being employed to achieve near zero release from BWR, this type of study provides a valuable data base for the prediction of doses and airborne concentrations in an accident situation.

For a costal site during the summer months, two phenomena seem to merit important consideration. Referencing Figure 4, one observes that when the vertical temperature gradient is super adiabatic ( $\nabla T < \Gamma$ ) ; one group of data is distinguished between mid morning and noon. Also, when the vertical temperature gradient is greater than the adiabatic lapse rate, another group of data is singularized as occurring mostly in the early morning hours.

These two groupings of the data are consistent with the existence of the sea breeze fumigation and a nocturnal fanning phenomena. That is to say that the changes in the solar radiation effect the near ground temperature profiles, which, if effected to a considerable height can effect the dispersion of the gaseous effluent.

In the early morning hours, the ground is cooler than the air above, consequently the temperature increases with height to some elevation at which a neutral temperature gradient is achieved. Under these stable conditions, the effluent plume will tend to seek a level at which it is neutrally bouyant. There it can remain for several miles, dispersion being accomplished by diffusion and the weak wind variability in the stable layer. Thus, the plume fans the perimeter of the reactor site serving as a localized source of external gamma's whose flux and concentration at near ground level is determined by the height of the plume above the ground.

At sunrise the solar radiation warms the ground faster than the air. The stable condition is gradually replaced by an unstable forced convection layer with a super adiabatic temperature gradient. ( $\nabla T < \Gamma$ ) This condition persists to some height at

which a stable condition exists. This represents a capped inversion, the forced convection is due to the ground heating. A sea breeze fumigation condition exists when cool air from the sea moves inland over the heated ground. If the emission point is below this capped inversion, then the effluent is mixed downward and high ground concentrations can be observed as shown in the second group of data under discussion. The duration of this effect is strongly related to site topography. By late afternoon this situation is no longer present and a near neutral condition exists.

This data is consistent with this interpretation as that data representative of the fanning situation shows a slowly increasing external gamma flux with distance from the emission point. This is due to the fact that the plume is relatively constant in height and that the Cs-138 and Rb-88 actively increase with distance from the point of emission.

Table 1

	Site I	Site II	Site III
Period of Measurement	Jun 22 to Jul 14, 1972	Jul 14 to Aug 4, 1972	Aug 4 to Aug 30, 1972
Distance (meters)	2570	5030	8210
Direction (WRT to True North)	059° 013'	054° 021'	059° 057'
Elevation MSL (meters)	36.2	16.7	30.4
X peak half-hour average ( $10^{-9}$ $\mu$ Ci/cc)	164	226	78.6
X peak daily average ( $10^{-9}$ $\mu$ Ci/cc)	24.6	36.0	13.5
<X> period average ( $10^{-9}$ $\mu$ Ci/cc)	6.45	19.7	2.73
X/Q half-hour peak ( $10^{-8}$ sec/m <sup>3</sup> )	245	261	78.6
X/Q daily average peak ( $10^{-8}$ sec/m <sup>3</sup> )	33.8	42.2	15.1
<X/Q> period average ( $10^{-8}$ sec/m <sup>3</sup> )	8.95	30.3	2.81
Estimated average dose ( $\mu$ r/hr)	2.83	8.64	1.197

\* This study was conducted under the sponsorship of the Northeast Utilities Company.

† Ph.D. Candidates, Nuclear Science and Engineering Department, Catholic University of America.

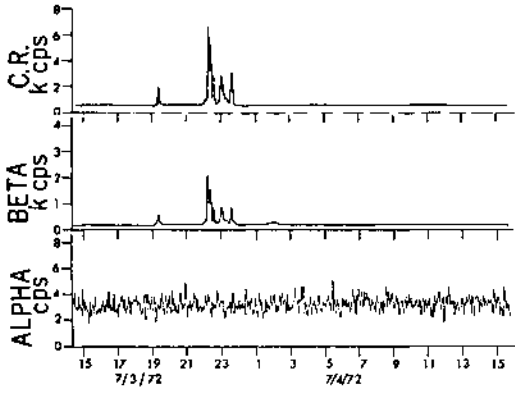


FIG.1, SITE 1

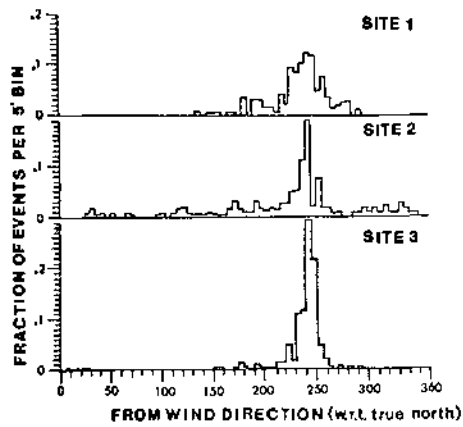


FIG.2, NORMALIZED HISTOGRAMS OF BETA/GAMMA EVENTS VERSUS WIND DIRECTION

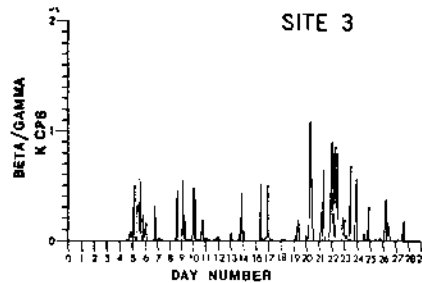
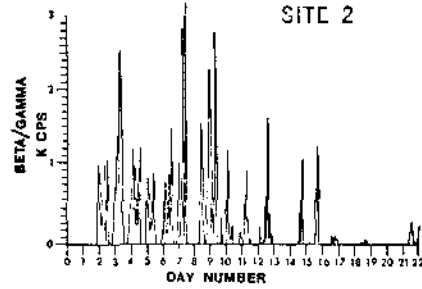
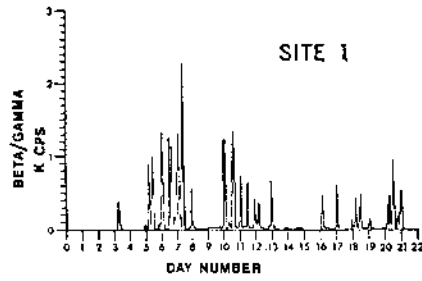


FIG.3, SUMMARY OF BETA/GAMMA DATA

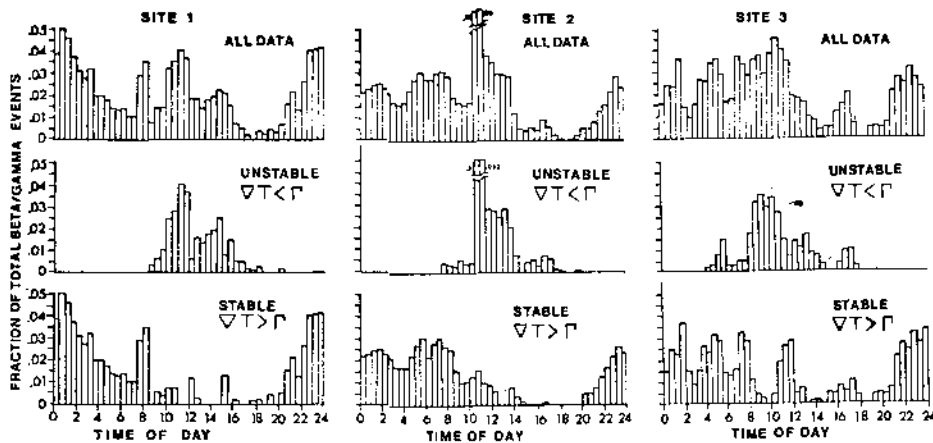


FIG.4, NORMALIZED HISTOGRAMS OF BETA/GAMMA EVENTS VERSUS TIME OF DAY.

## АТОМНЫЕ ЭЛЕКТРОСТАНЦИИ КАК ИСТОЧНИК ВЫБРОСА УГЛЕРОДА-14

Рублевский В. П., Зыкова А. С., Туркин А. Д.  
Институт биофизики Министерства здравоохранения СССР  
Москва, Союз Советских Социалистических Республик

### Abstract

$^{14}\text{C}$  is produced in nuclear reactors by neutron reactions in  $^{14}\text{N}$ ,  $^{17}\text{O}$  and  $^{13}\text{C}$ . The expanding use of nuclear reactor power production will contribute additional amounts of  $^{14}\text{C}$  to the existing background levels due to atmospheric testing of nuclear devices and natural production by neutrons in upper atmosphere.

The report considers the problem of environmental  $^{14}\text{C}$  contamination from increasing utilisation of nuclear reactors for power production. The experimental data on production of  $^{14}\text{C}$  at different types of nuclear power stations in the USSR are presented in the report.

Future production of  $^{14}\text{C}$  from nuclear power stations will be approximately 12 000 Ci/day to 2010. The  $^{14}\text{C}$  concentration in the terrestrial biosphere will reach 1050 per cent of its natural level by this year.

These estimates are compared with existing equilibrium levels of naturally occurring  $^{14}\text{C}$  and with the decreasing inventory of  $^{14}\text{C}$  from atmospheric nuclear tests.

Углерод играет существенную роль во всех формах жизни. Он участвует в большинстве биологических и биохимических процессов, происходящих на Земле, являясь структурной основой всей органической материи. Он непосредственно входит в молекулы белков и генетически значимых структур. Среднее содержание его в организме человека составляет по весу 10%, в молекулах белков - примерно 50%, в молекулах ДНК и РНК - около 37%.

Попавший во внешнюю среду углерод-14 включается в обменный цикл углерода и проникает вместе с ним во все живые организмы.

Естественный углерод-14 образуется в основном по реакции  $\text{N}^{14}(\text{n}, \text{p})\text{C}^{14}$  при взаимодействии нейтронов космического происхождения с ядрами азота атмосферы. Скорость образования, равная в среднем 2,2 ет/сек.см<sup>2</sup>, обуславливает его концентрацию в земной биосфере 14+1 расп/мин.г С /I/.

Искусственный  $\text{C}^{14}$  в заметном количестве образуется при испытании ядерного оружия и работе атомных реакторов.

Испытания ядерного оружия привели к образованию значительного количества углерода-14. В результате концентрация его в атмосфере и земной биосфере в 1962-63 гг. увеличилась в два раза.

В атомных реакторах  $C^{14}$  может образоваться в результате происходящих ядерных реакций типа  $p\alpha$ ,  $n\gamma$  и  $n\alpha$  на ядрах  $N^{14}$ ,  $O^{17}$  и  $C^{13}$ . Вклад каждой из этих реакций в образование  $C^{14}$  будет зависеть от типа и конструкции атомного реактора. Были исследованы выбросы в атмосферу углерода-14 на трех типах атомных реакторов, используемых в настоящее время в СССР для АЭС /2, 3/:

- водо-графитовых на АЭС АН СССР и I блоке Белоярской АЭС,
- водо-водяных на I блоке Нововоронежской АЭС и Ульяновской АЭС,
- реактора с органическим замедлителем-теплоносителем в атомной реакторной блочной установке.

Результаты измерений приведены в табл. I.

Таблица I

Количество углерода-14, поступающего в атмосферный воздух с газовыми отходами атомных реакторов различного типа

Тип реактора	Тепловая мощность, Мвт	Тепловая мощность в период исследования, Мвт	Выброс $C^{14}$ , мкюри/сутки
Водо-графитовый, АЭС АН СССР	30	12	$9 \pm 3$
Водо-графитовый (АМБ), БАЭС	285	210	$140 \pm 50$
Водо-водяной (ВВЭР-210), НВАЭС	760	740	$120 \pm 30$
Водо-водяной (ВК-50), УАЭС	150	90	$30 \pm 10$
С органическим замедлителем-теплоносителем (АРБУС)	5	5	$0,6 \pm 0,2$

Для изучения водо-графитовых реакторов величину выброса  $C^{14}$  можно определить из равенства:

$$Q = 0,75 M_T,$$

где  $Q$  - величина выброса  $C^{14}$ , мкюри/сутки;

$M_T$  - тепловая мощность реактора, Мвт.

Для водо-водяных реакторов -  $Q = 0,25 M_T$ .

Приведенные зависимости позволяют оценить величину выбросов проектируемых АЭС с реакторами подобного типа: ВВЭР-440, ВВЭР-I 000 (водо-водяные) и РБМ-К-I000 (водо-графитовый) /3/. В табл. 2 приведены результаты такой оценки.

Таблица 2

Предполагаемое количество  $C^{14}$ ,  
которое может поступать в атмосферный воздух  
с выбросами реакторов ВВЭР-440, ВВЭР-1000 и РБМ-К-1000

Тип реактора	Примерная тепловая мощность, МВт	Выброс углерода-14 мкюри/сутки
ВВЭР-440	1370 /7/	340
ВВЭР-100	3000 <sup>†</sup>	750
РБМ-К-1000	3200 /7/	2500

<sup>†</sup>Определено из условия, что КПД АЭС = 30%.

Исследования показали, что количество  $C^{14}$ , поступающее во внешнюю среду с выбросами действующих в СССР АЭС, увеличивает его естественную концентрацию в растительности в районах их размещения не более, чем на несколько процентов от естественного уровня.

В связи с программой широкого строительства мощных АЭС, намеченного во всех экономически развитых странах мира /3,4,5/, количество углерода-14 поступающее в тропосферу с их выбросами, будет непрерывно возрастать. При этом можно предположить, что загрязнение окружающей среды будет по своему характеру приближаться к глобальному.

В. К. Спинрад /4/ привел прогноз развития атомной энергетики до 2010 года. Если предположить, что в этот период АЭС будут строиться только с водо-графитовыми и водо-водяными реакторами, то можно рассчитать общее количество углерода-14, которое будет поступать во внешнюю среду. Уровни его накопления в биосфере рассчитаны, исходя из двухрезервуарной модели обменного бассейна углерода/6/. Результаты расчетов приведены в табл. 3.

Таблица 3

Электрическая мощность АЭС, общая величина выброса  $C^{14}$   
и накопление его в биосфере

Г о д	1970	1975	1980	1985	1990	1995	2000	2005	2010
Эл.мощн. АЭС, Гвт/4/	24	123	345	810	1610	2780	4260	6070	8290
Выброс $C^{14}$ , кюри/сут	40	180	520	840	2400	4200	6400	9100	12400
Концентрация $C^{14}$ , % от естественн.уровня	0,6	3,0	10	30	80	180	350	620	1050

Поступление  $C^{14}$  во внешнюю среду с выбросами АЭС приведет к тому, что к 2010 году концентрация его в биосфере превысит естественную примерно в 10 раз.

Максимально допустимый уровень накопления  $C^{14}$  в биосфере зависит от принятой допустимой величины генетически значимой

дозы внутреннего облучения (ГЗД<sub>в</sub>). Если исходить из ГЗД<sub>в</sub>, равной 1,5 бэр за 30 лет /7/, и суточной нормы потребления углерода - 300 г/8/, то максимально допустимая концентрация углерода-14 в земной биосфере не должна превышать 1000 расп/мин.г С или  $4,5 \cdot 10^{-10}$  кюри/г С.

Следует отметить, что особенность действия  $C^{14}$  на организм заключается в том, что радиационное действие бета-излучения усугубляется фактором химического превращения атома углерода в атом азота при радиоактивном распаде углерода-14, особенно когда он включен в молекулы генетических структур ДНК и РНК. Впервые на возможность мутагенного действия этого эффекта указал Л. Паулинг /9/. Он считал, что этот процесс вызывает около 10% всех повреждений. К. Е. Прудом /10/ исследовал этот эффект на дрозофилах и не обнаружил его. С другой стороны, А. М. Кузин с сотрудниками /11, 12/, Г. А. МакКвайд, М. Фридкин и А.А. Атчисон /13/ и др. получили положительные результаты. Величина эффекта, определяемая по различным тестам, колебалась от 2,3 до 25. Поэтому в связи с расширением контингента населения, который может подвергнуться воздействию углерода-14, необходимо предусматривать меры для уменьшения количества углерода-14 в выбросах АЭС и определить его мутагенную эффективность при превращении атома углерода в атом азота в молекулах ДНК, которая, вероятно, явится решающим фактором в определении допустимой величины генетически значимой дозы внутреннего облучения (ГЗД<sub>в</sub>) и опасности увеличения концентрации углерода-14 во внешней среде.

В связи с возможностью глобального загрязнения внешней среды углеродом-14 уже в настоящее время возникают два вопроса, которые требуют решения в ближайшем будущем.

Во-первых, расширение контингента населения, которое может подвергнуться воздействию углерода-14, требует однозначного решения задачи о роли эффекта трансмутации атома углерода при его радиоактивном распаде в молекулах генетически значимых структур (ДНК и РНК). Величина ОБЭ, связанная с этим эффектом, необходима для оценки радиационного действия углерода-14 на организм человека.

Во-вторых, необходимо предусмотреть меры для уменьшения количества углерода-14, которое будет поступать в атмосферу с выбросами АЭС.

#### Литература

1. Доклад НКДАР ООН. 1961, А/АС, 82А, 137.
2. А. М. Петросьянц. Атомная энергия, 1967, т. 23, вып. 5, 379-395.
3. А. М. Петросьянц. Атомная энергия, 1969, т. 27, вып. 4, 263-274.
4. Spinrad B. A. Symposium on environmental aspects of nuclear power stations. New-York, 1970, 10-14, SM-146/2.
5. Доклад НКДАР ООН. 25 сентября 1969 г., А/7613.
6. Harkness D. D. Walton A., Nature, 1969, v. 223, No 5212, 1216-1219.
7. Нормы радиационной безопасности, НРБ-69. М., Атомиздат, 1970.



8. Standards Physiological values for a reference man.  
ICRP/69/CJ-1/2.
9. Pauling L. Science, 1958, v. 123, No 3333, 1183-1186.
10. Доклад НКДАР ООН. 15 июля 1966 г., A/6314.
11. А. М. Кузин, Б. М. Исаев, В. В. Хвостова, В. И. Токарская,  
Ю. И. Брегадзе. АН СССР, М., 1960.
12. А. М. Кузин, Я. Л. Гембицкий, Ю. А. Лапин, Г. С. Каландо,  
Ю. И. Брегадзе, Я. В. Мамуль, Е. Н. Мясникова.  
Радиобиология, 1964, т. IV, в. 6, 804-809.
13. McQuade H. A., Fridkin M., Atchison A. A. Exper. Cell. Res.,  
1956, v. 11, 249-264.

THE SAFE DISPOSAL OF LIQUID RADIOACTIVE WASTES FROM  
THE UNITED KINGDOM NUCLEAR POWER PROGRAMME:  
A CURRENT VIEW OF THE ENVIRONMENTAL SITUATION

Dr N. T. Mitchell  
Ministry of Agriculture, Fisheries and Food  
Fisheries Radiobiological Laboratory  
Hamilton Dock  
Lowestoft  
Suffolk

Abstract

Operational experience of the nuclear power programme in the United Kingdom has shown that the principal environmental consequences arise from the disposal of liquid radioactive waste, and relate to public health rather than to direct effects on resources. The emphasis on environmental protection programmes has thus been on the limitation of public radiation exposure. Provided that this is held within the acceptable limits as defined by ICRP, then resource considerations will be adequately protected.

1 Introduction

The principal source of radioactive waste in the United Kingdom is the nuclear power programme; other sources of radioactive waste include a variety of research and development activities, industry and medicine, and though some use large quantities of radioactivity little becomes waste for disposal. The most important category of radioactive waste is liquid, the relative environmental impact of both gaseous and solid releases being of little significance. Another important generalization concerns the relative importance of human radiation exposure and the potential damage to environmental resources. Provided that disposals are so controlled that human exposure is within the dose limits recommended as acceptable by the International Commission on Radiological Protection<sup>1</sup>, the potential risk to environmental resources is minor<sup>2</sup>, reflecting the relative radiosensitivity of an animal or organism, which usually bears an inverse relationship to its evolutionary status. The emphasis in the UK has therefore been on limiting public radiation exposure whilst still maintaining a sufficiently close watch on environmental resources to ensure that they are safeguarded.

Radioactive waste is an inevitable consequence of a nuclear power programme and in its management some disposal to the environment is a practical necessity. The British approach is thus a pragmatic one, seeking the responsible utilization of environmental resources for waste disposal to an extent which will not put either the environment or public health at risk, or impose economically-crippling restrictions on the nuclear industry and thus deny the nation use of this energy source.

2 UK waste disposal policy

2.1 Radiation exposure standards

The basic objectives of UK waste disposal policy were stated in a government White Paper in 1959<sup>3</sup> and were embodied in legislation<sup>4</sup> in the following year. The first



two objectives, which are both mandatory and must be met regardless of the cost, require:

- (i) compliance with the ICRP-recommended dose limits for members of the public - covering somatic consequences of waste disposal, and
- (ii) that exposure of the public averaged over the population of the country does not exceed 1 rem per person per 30 years - designed to safeguard against genetic effects.

The third policy objective requires that, this time taking into account cost, convenience and national importance of the subject, doses shall be kept as far below these mandatory limits as is practicable.

The first of these objectives originally referred to the most highly-known exposed individual in line with the ICRP recommendations of the day. Since then the later series of ICRP recommendations<sup>1</sup>, endorsed by the Medical Research Council, have been adopted for use in the UK. In consequence and wherever practicable, control measures are now based on mean exposure of critical groups, though in most cases it has not proved possible to identify such groups, so that control has continued on the basis of known individual exposure.

## 2.2 The application of policy objectives

All radioactive waste discharges are subject to a system of prior authorization, issued by departments of central government. Authorizations are legally enforceable and specify limits on the quantity, type and rate of release of radioactivity, with further conditions, for instance relating to effluent and environmental monitoring.

In view of the mandatory nature of the first two objectives of national waste disposal policy, it is essential to know the capacity of the environment to which the waste will be released, before an authorization is issued. However, in no case has the operator of a site been allowed to discharge waste up to this limit, for, in line with the third policy objective, only disposal for which there exists a proven need is permitted, though some reasonable margin is allowed for operational flexibility and inaccuracies in forecasting waste arisings, treatment plant performance, etc. In principle, the environmental capacity may be set against either somatic or genetic dose limits, though in practice the former has generally been found to be more restrictive.

The first authorization - issued before the site is in operation - is based on an assessment of environmental capacity calculated on largely theoretical data and, because of the uncertainties in some of the assumptions made, a "safety factor" - usually 10 - is applied to the estimate of environmental capacity before it is used to decide the upper limit which can be specified in the authorization.

Once a site begins to discharge waste a system of inspection is set in being by the authorizing departments of central government, backed up by effluent and environmental monitoring. The primary aim in monitoring effluent is to check operators' statements, and whilst this is also true for environmental monitoring - the site operator must carry out sufficient monitoring to demonstrate the radiological safety of discharges in terms of public radiation exposure - the work undertaken by authorizing departments includes an element of research to provide data on which environmental capacity can be reassessed.

### 2.3 The scientific evaluation of discharge limits

The system used for assessment of discharge limits is the well-known critical path approach, which aims at achieving a basic understanding of the way in which radioactivity behaves after release to the environment, in particular the routes or "pathways" through which contamination of environmental materials and human radiation exposure occurs. The first stage is a habits survey in which the characteristics or habits of the exposed population are studied - initially a qualitative identification of the pathways in a particular environment by which radioactivity is, or may conceivably be, returning to man. This is followed by a quantitative evaluation of each pathway, in the course of which it is usually found that one or perhaps two pathways will be of so much greater importance than the others that exposure through the remainder is minor and can often be ignored. These limiting situations are termed the critical pathways and it is frequently found that almost all the exposure is due to one radionuclide or a very few - the so-called critical radionuclide(s). Because of the individual characteristics of the critical radionuclide(s), which may, for instance, lead to uneven distribution within the body, a particular organ may be said to be "critical", meaning that exposure of it is greater than of others; alternatively the whole body may be uniformly irradiated, as is often the case from external sources of contamination, and the critical organ is then quoted as "total body". Within the exposed population it may also be possible to identify a small group who are at greatest risk - the "critical" group - and on which exposure estimates may be based, particularly for purposes of waste disposal control; more often this does not prove possible because of the small number of people exposed to a significant extent, and estimates are then made on the basis of the most highly exposed person found in a sample of the exposed population - the "critical individual".

Applied to the evaluation of discharge limits, the term "environmental capacity" is now in general use; the system differs according to whether assessment is being made for the first time - that is before operation of the site has begun - or is a reassessment of the consequences of a disposal which has been in operation long enough to generate measurable environmental contamination.

2.3.1 Pre-operation The starting point for the computational model is an assumed discharge rate - e.g. 1 Ci/day - of a mixture of specified isotopic composition. A hydraulic model of the receiving water mass is needed to provide a basis for estimating equilibrium concentrations in water for each radionuclide. In the next stage, appropriate concentration factors are used to obtain estimates of the equilibrium concentrations in the critical material(s). It is then a simple step, by way of habits survey data on consumption rates, etc., to translate these concentrations into rates of intake of radioactivity, which may be compared with the values of permissible daily intake, consistent with ICRP dose limits (derived ICRP data); an actual estimate of exposure rate which would result from discharges at the assumed rate can then be made. These steps are carried out separately for each radionuclide, and the total effect of the effluent is estimated by summation. Separate estimates are made for several body organs - often the GI Tract and Bone in addition to Total Body - as a result of which the identity of the critical organ(s) and radionuclide(s) will become apparent.

This is the procedure applied to an internal exposure pathway; the method for external pathways is very similar. Translation of contamination levels into exposure is more complex than for internal exposure and requires a dosimetry model, but the rest of the sequence is simpler because the estimate of exposure can be compared directly against ICRP-recommended dose limits.

2.3.2 During operation Once routine discharges are being made a simpler system can be used unless contamination has not reached measurable levels, in which case the pre-operational system is retained. Provided that contamination is measurable in critical materials, then a direct correlation can be deduced between this and the discharges causing it, utilizing environmental and effluent monitoring data. The relationship found is then used, in the same way as in pre-operation, to estimate the extent of public radiation exposure.

In the early phase of operation of a site, when discharges are relatively small and maybe not yet at equilibrium with respect to quantity and/or isotopic composition, it may be possible to improve on the pre-operational method of assessment by use of indicator materials. A more extensive knowledge of concentration factors is required than when there is contamination of the critical material, involving perhaps several radionuclides and the indicator and (potentially) critical materials. Whilst this is a useful variation on the normal methods it is one which has found little use in practice, for although it avoids the need for hydraulic models it is not this factor but uncertainty in effluent composition that is the major source of error in pre-operational assessment.

### 3 Waste disposal and its control in practice

The ultimate test of any waste disposal policy is its use in practice and in particular the successful limitation of public radiation. However, merely reducing such exposure is not an end in itself, for given unlimited financial resources any measure of waste treatment can be achieved; in a limited economy it is necessary to balance the cost against the benefit which will be reaped. In the UK the application of control measures has become a commonsense compromise between the two extremes of unnecessary discharge on the one hand and unnecessary and financially crippling restriction to nil discharges on the other - given always that public exposure is not going to exceed the mandatory limits of UK policy, the attainment of which is the first priority.

Just how this flexible policy has worked out in practice can be seen from the nuclear power programme - examples covering a range of different environments - the salient factors being summarized in Table 1. Fuller details are contained in reference<sup>5</sup>.

#### 3.1 Fuel element manufacture

Of the two establishments involved in fuel manufacture the larger discharges are from Springfields, Lancashire, from which disposal of low-level radioactive effluent is made by pipeline into the tidal estuary of the River Ribble. Wastes are predominantly  $\beta$ -active and consist of residues of uranium and its immediate daughter products, the most prominent of which is protactinium-234m. This results in contamination of the mud banks, especially in the vicinity of the pipeline outlet, and the critical pathway is one of external exposure, the most highly exposed population being workmen who maintain the river banks and attend to navigational aids. Even in extreme cases, exposure by this means is insignificant - at most a fraction of 1% of the ICRP-recommended dose limit - and the genetically-significant dose is negligible.

#### 3.2 Nuclear power stations

With 11 sites in operation on a commercial basis, examples can be drawn from a range of types of location and three will be cited here - one discharging waste to a soft-freshwater lake (Trawsfynydd, Merioneth), one to a river estuary (Bradwell, Essex),

and the third to the open sea (Dungeness, Kent). In each case the type of reactor is the same - the "Magnox" type of carbon dioxide gas-cooled, graphite-moderated unit, and the same method of waste disposal is employed - by dilution after treatment into the cooling water outflow which provides a large measure of initial dilution.

3.2.1 Trawsfynydd Discharges are made to a small lake whose capacity is small, partly because of an unusual combination of critical nuclide and material, caesium-137/-134 and trout, which reconcentrates this radioactivity to high degree. A considerable amount of treatment plant, more than at any other power station, has been installed, designed primarily to remove radiocaesium. As a result, satisfactory control has been achieved and even the most avid consumer of trout has not been subject to as much as 5% of the ICRP-recommended dose limit. In genetic terms exposure is also very low and with a small population involved it is only around 1 man-rem per year.

3.2.2 Bradwell The critical material has been the oyster throughout the 12 years during which this power station has been in operation, though the identity of the critical nuclide has changed - initially being zinc-65, a rôle now taken over by silver-110m. Tritium apart - and as elsewhere this nuclide is of negligible radiological significance - the principal nuclides in the effluent are those of radiocaesium, minimized by means of treatment plant which also removes a large proportion of the zinc-65, and which replaced plant originally designed specifically for zinc-65. Traces of caesium-137, cobalt-60 and iron-55 are also found, though none contribute significantly compared with silver-110m and zinc-65, whose combined effect is only equivalent to a few millirem per year to the largest-known consumer of oysters. The genetic impact of discharges is also trivial, less than 0.1 man-rem/year, though the relative rôles of the radionuclides are reversed, most of this exposure being from radiocaesium.

3.2.3 Dungeness, Kent This illustrates the situation where discharges generate no detectable environmental contamination, so that it is not possible to say which is the critical pathway and it would be academic even to predict one. Two are obviously of potential importance - internal exposure through consumption of fish caught in local waters close to the discharge area, and external exposure due to occupation of the local beaches - and in the presence of effluent composition radiocaesium would be most important. Sensitivity of detection of radioactivity affords one means of estimating the maximum conceivable level of public radiation exposure, from which it is clear that no individual could possibly be subject to as much as 0.1% of the ICRP-recommended dose limit, though models used to predict dispersion suggest that the true level is some orders of magnitude lower still. Genetically significant exposure is also minute - less than 0.1 man-rem per year.

### 3.3 Fuel reprocessing

Of the two plants involved in this operation the larger is Windscale on the Cumberland coast, discharging waste by pipeline into the Irish Sea; it also provides a wider range of exposure pathways, which emphasizes the importance of setting discharge limits individually for each site.

Three exposure pathways form the present basis for control of the major components of liquid waste, two being internal, due to consumption of foodstuffs - laverbread made from the seaweed *Porphyra*, and locally-caught fish - the third being external and the result of uptake of radioactivity by mud and silt.

The Porphyra/laverbread pathway has been of prime importance since plant operation began more than 20 years ago, the critical nuclide being ruthenium-106. Unlike most situations the exposed population is not local to the discharge area but some 300 miles away in South Wales, where the foodstuff is eaten. A critical group has been identified<sup>6</sup>, so that control does not have to be exercised on the highest known rate of exposure. The remoteness of the exposed population poses further problems of monitoring and control, for during manufacture Cumberland-derived seaweed is diluted with uncontaminated seaweed from other areas. For this reason control of those components of the waste for which this pathway sets the effective limits - they include the alpha emitters and strontium-90 as well as ruthenium-106 and cerium-144 - have been related to contamination levels in locally-grown Porphyra so as to be "fail-safe". Retrospectively it is possible to calculate the received dose from analysis of the laverbread product, and in recent years the critical organ dose (the GI tract) has been found to be in the range 5 to 10% of the ICRP-recommended dose limit. This is now assessed on a daily consumption rate of 130 g of laverbread deduced for the critical group on the basis of the most recent habits assessment. In contrast, genetically-significant exposure through this pathway is very low indeed, only a few man-rems per year, a significant fraction of which comes from caesium-137 and -134, both insignificant contaminants in somatic terms.

The other important internal exposure pathway involves fish consumption, and caesium-137 and -134 are the critical nuclides - indeed the only contaminants in most of the fish caught. A large population eat it, though mean consumption rates are low. Even at the higher values of consumption rate recorded by local fishermen, exposure is only very low - a few per cent of the ICRP-recommended dose limit - total body being the critical organ. Genetically this is the most important Windscale pathway, though in absolute terms - total population gonad dose being of the order of  $10^2$  man-rem per year - it is of little significance, less than 0.01% of the national dose limit.

The external exposure pathway highlights another contrast in the size of exposed populations, which in this case is very small. The highest dose rates are found in a small estuary near to Windscale where mud and silt collects, contaminated by several  $\gamma$ -emitters but principally zirconium-95/niobium-95. Habits surveys have shown that the most highly exposed individuals are salmon fishermen, whose exposure and doses have ranged up to about 10% of the ICRP-recommended dose limit in recent years. Because of the small size of the population affected by this pathway, the total population dose is very small, not more than 1 man-rem per year.

#### 4 Conclusions

The stringent standards set to limit disposal of radioactive waste in the United Kingdom have ensured that the consequences in terms of radiation exposure of the public are very slight: not only is such exposure within the objectives of UK policy - themselves consistent with the recommendations of the International Commission on Radiological Protection - but in all disposals radiation exposure of the public is well within the prescribed dose limits and in many instances is very much less.

#### References

- 1 ANON. Recommendations of the International Commission on Radiological Protection (adopted September 17, 1965). ICRP Publication 9, Pergamon Press, Oxford (1966).

- 2 WOODHEAD, D. S., The biological effects of radioactive waste. Proc. Roy. Soc. Lond. B 177 (1971), 423.
- 3 ANON. The Disposal of Radioactive Wastes. Cmnd 884 (1959). HMSO, London.
- 4 ANON. The Radioactive Substances Act, 1960, 8 and 9 Eliz. 2 (1960) ch. 34. HMSO, London.
- 5 MITCHELL, N. T., Radioactivity in Surface and Coastal Waters of the British Isles, 1971. Ministry of Agriculture, Fisheries and Food, Fisheries Radiobiological Laboratory, Lowestoft. Technical Report FRL 9 (1973).
- 6 PRESTON, A. and JEFFRIES, D. F., The ICRP critical group concept in relation to the Windscale sea discharges. Health Physics, Vol. 16, pp. 33-46.

Table 1 Characteristics of selected disposals of liquid radioactive waste and mean radiation exposure of the public in 1970-72

Site	Pathway	Critical		Derived working limit	Exposure* (percentage of ICRP recommended dose limit)
		Material	Nuclide		
Springfields	External	Mud	$^{234m}\text{Pu}$	$1.25 \text{ mR h}^{-1}$	< 1
Trawsfynydd	Internal	Trout flesh	$^{137}\text{Cs}$	$440 \text{ pCi. g}^{-1}$	3
			$^{134}\text{Cs}$	$239 \text{ pCi. g}^{-1}$	2
Bradwell	Internal	Oyster flesh	$^{110m}\text{Ag}$	$880 \text{ pCi. g}^{-1}$	0.2
Dungeness	Internal	Plaice flesh	$^{137}\text{Cs}$	$175 \text{ pCi. g}^{-1}$	< 0.1
			$^{134}\text{Cs}$	$89 \text{ pCi. g}^{-1}$	
	External	Silt	$^{137}\text{Cs}$ $^{134}\text{Cs}$	$0.3 \text{ mR h}^{-1}$	< 0.1
Windscale	Internal	<u>Purphyra</u> /laverbread	$^{106}\text{Ru}$	$179 \text{ pCi. g}^{-1}$	15
			$^{144}\text{Ce}$	(laverbread)	
	Internal	Plaice	$^{137}\text{Cs}$	$175 \text{ pCi. g}^{-1}$	2
$^{134}\text{Cs}$			$80 \text{ pCi. g}^{-1}$		
External	Mud	$^{95}\text{Zr}/^{95}\text{Nb}$	$1.7 \text{ mR h}^{-1}$	10	

\*Most highly exposed individual, except for Windscale internal exposure pathways, where critical groups form the basis of estimation.



CLIMATOLOGICAL AND METEOROLOGICAL CONSIDERATIONS  
FOR SITING OF NUCLEAR INSTALLATIONS IN HILLY OR  
MOUNTAINOUS ENVIRONMENT

E. NAGEL

Health Physics Division  
EIR, Eidg. Institut für Reaktorforschung  
CH-5303 Würenlingen, Switzerland

Abstract

In Switzerland the geographical regions where nuclear installations can be implanted are the Midlands, situated between the Alps and the Jura mountains, where most of the population and industries are concentrated, and the broad valleys in the alpine zone.

In each region similar conditions of the general climate exist but local factors due to terrain feature and orientation make it not possible to deduce the characteristic data for a specific site from those for another location. Local, even short time investigations are necessary to permit to derive data on distribution and persistence of critical wind directions.

\* \* \*

Reliable data of the frequency of wind directions and atmospheric diffusion conditions is necessary in order to estimate the radiation dose to which the population in the environment of a nuclear installation could be exposed.

In mainly flat regions with homogeneous climatic conditions, it is possible to use the data for a typical location to characterize the situation at any site in this region.

The situation becomes different in regions where similar conditions of the general climate exist but where local factors due to topography such as mountains, hills and valleys become important. This is the case in the Midlands of Switzerland which is economically the most important region of the country, where most of the population and of the industries are concentrated. This relatively small region is situated between the two other important regions of the country, the Alps and the mountains of the Jura (cf. Fig. 1). Most of the sites of the erected and planned nuclear installations are located here along the largest river, the Aare, or next to the Rhine in a valley which is situated between the Jura and the Black-Forest in the north of the country, but where the climate is very similar to that existing in the Midlands.

The Midlands themselves are composed of a succession of relatively broad valleys which are separated by hills of moderate altitude (in general 100 - 300 m above the bottom of the valleys). This topographic configuration and the situation of some sites next to the higher mountains of the Jura do not allow to deduce all the characteristic data for a specific site from those of another typical location even if the climate over the whole region is very similar.

So for instance the similarity of the main wind directions due to the general orientation of the Midlands between the Alps and the Jura is evident, but it is only valid for advective winds with moderate or great velocity and adiabatic temperature gradient. In such situations the winds blow mostly over the hills even if their direction is perpendicular to them; they develop a relative greater velocity above the top of the ridges than above the bottom of the valleys. The "small-shot" model of air in motion hitting the slope of a hill cannot be applied here; this fact has been demonstrated by many no-lift balloon flights in the region of Würenlingen (cf. Fig. 1: W). During local inversion conditions due to heat radiation of the ground in the night, local winds with low velocity prevail, they blow from directions which are dictated in the majority of the cases by the local topographic configuration. Their persistence will be very variable. Due to the fact that local inversions are frequent, investigations become necessary to determine local winds which may become critical.

Observations made at Würenlingen (cf. Fig. 1: W) during many years show the following figures:

	Adiabatic Lapse Rate	Local Inversion
Frequency (% of total time)	39 %	41 %
Average wind velocity	3.5 m.sec <sup>-1</sup>	1.8 m.sec <sup>-1</sup>
Frequency of periods with variable winds	-	75 %
The average frequency of wind velocities <1 m.sec <sup>-1</sup> at Würenlingen amounts to 12 % of total time.		

These figures which do not include the distribution of wind directions, can be considered as generally valid for the greatest part of the Midlands. They prove also the importance of the local weak winds mentioned above, which are very often connected with unfavourable diffusion conditions. This fact is illustrated in Fig. 2: The frequency of wind velocities (hourly averages: u) recorded at Würenlingen 70 m above ground level are represented in diagram A. By multiplying these values by the corresponding factor 1/u (in diffusion formulae, u is always to be found in the denominator) one obtains a so called "degree of risk" for the different velocities, represented in diagram B. Fig. 2 shows that most of the observed wind velocities range between 0.7 and 2.0 m.sec<sup>-1</sup> (A) and that the maximal "degree of risk" appears with velocities near 0.5 m.sec<sup>-1</sup>; it decreases rapidly with velocities above and below this value (B). Calms of one hour duration have not been observed.

Investigations carried out at sites located next to higher mountains, the Jura for instance where the difference of altitude between the mountain and the Midlands is several hundreds to one thousand and more meters, showed a very characteristic distribution of the directions of slow winds; the prevailing winds of this type blow from the mountains to the Midlands (so called "mountain winds"). So the distribution becomes completely different from that observed during the day or as an average over the year, when all velocities and weather conditions are considered and when the prevailing wind directions are parallel to the general orientation of the Midlands. For instance, such a specific "mountain wind" has been recorded at a site next to the Jura (cf. Fig. 1: G) during a few years with an anemometer placed 30 m above ground level. This wind blows from the north and has an average velocity <1 m.sec<sup>-1</sup>; it can be observed practically only during the night (13% of total time between 1800 and 0600; 3% during the day only); it is more frequent between March and October (15-17 % of total night-time) than in winter. The distance up to which this wind can be observed in the Midlands has not been investigated and the maximum altitude up to which it blows is also unknown.

The connection of weak winds with stable atmospheric conditions is of great importance too, because particular diffusion conditions may appear during inversions. The occurrence of fog for instance is locally very different in the Midlands. It is most frequent in the northern parts of them, next to the Jura. Normally fog banks do not extend very high above ground so that they do not always reach the edges of the valleys. They mostly appear during the night at the end of an inversion and last a few hours. Very unfavourable dilution conditions, as fumigation, prevail in fog. Local investigations have to be carried out at the sites concerned, because the existing statistics concerning this element are incomplete.

In Würenlingen, for instance, where such studies have been performed, 190 (or 16 %) out of 1150 local inversions, recorded during 5 years, were observed which ended with fog, and approximately in 10 % of total time the atmospheric diffusion conditions (including local inversions) were favourable to fumigation. Even if these figures are specific for this location, they show the frequency of such situations which has not to be neglected. For instance it could become important to know the average vertical extension of the air masses concerned; in many cases already an increased height of the point of release (stack) of a nuclear installation could help to evacuate probable pollutants over the critical heights if thermal lift would not be sufficient.

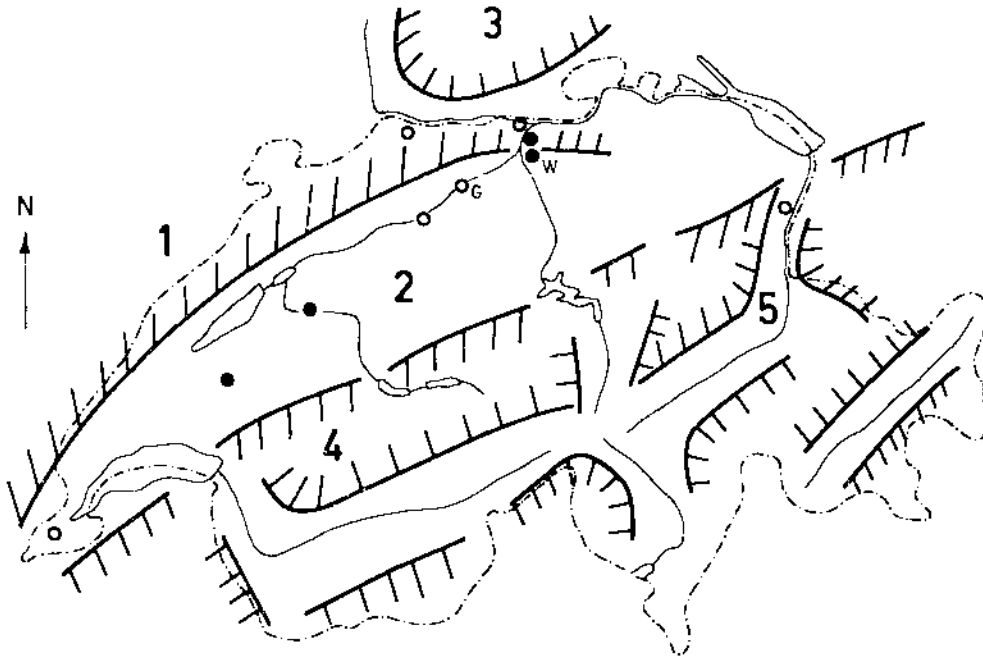
Regions other than the Midlands, where nuclear installations, power stations for instance, could be implanted are the geographically well situated (favourable communications) broad transversal and longitudinal valleys in the alpine zone (cf. Fig. 1).

The climate of these valleys is in many points different from that encountered in the Midlands. The wind regime for instance, which is similar in all the valleys considered, shows a very effective canalisation due to the topography (high and mostly steep slopes) and a pronounced diurnal periodicity of wind directions (moderate, sometimes strong winds blowing up the valleys in day-time, weak winds blowing down the valleys during the night). In addition each geographical unit shows some particularities due to its orientation; for instance there will be an increased frequency of advective winds in valleys oriented to north (or to south), which may appear as strong turbulent downwinds (e.g. Föhn). On the other hand the same valleys are sheltered from the advective westerlies or south-westerlies which are so frequent in the Midlands.

This local differences will make local, even short-time investigations necessary, as they are now being performed in the valley of the upper Rhine at the site of a planned nuclear power station (cf. Fig. 1: 5). Exact data are not yet available.

Some difficulties could arise during the measurement of the vertical gradient of temperature in such a valley namely if they are performed in meteorological huts on steep slopes; according to their orientation (exposition to sun), erroneous data may be obtained. To determine the vertical extension of local inversions which seem to be frequently confined to a few hundred meters above the bottom of the valleys, the use of captive balloons might be indicated.

Fig. 1



LOCATION OF THE ERECTED AND PLANNED NUCLEAR INSTALLATIONS IN SWITZERLAND

Facilities ● erected  
○ planned

Midlands 2

Alps 4

Jura mountains 1

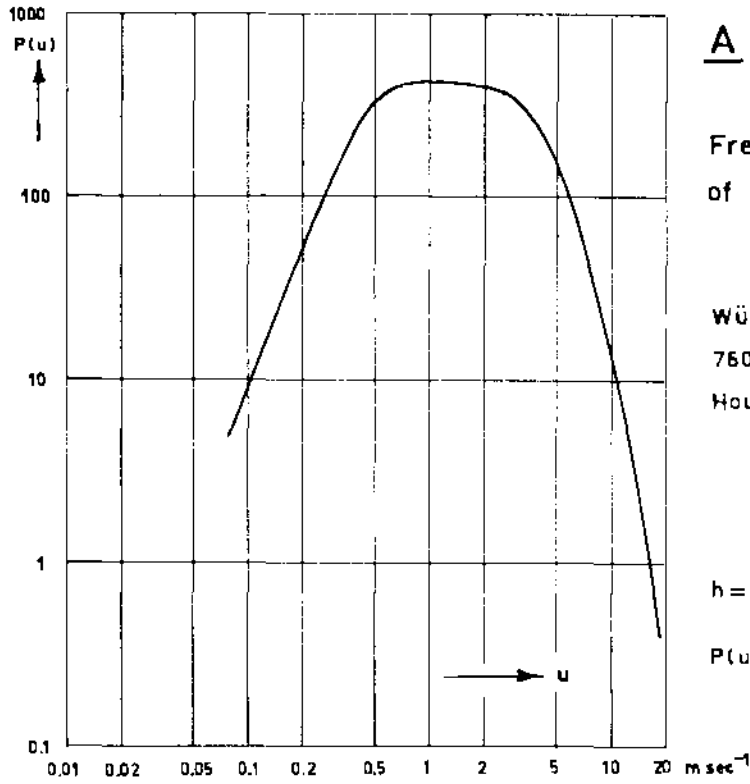
Black-Forest 3

Valley of the upper Rhine 5

Most of the facilities are situated in the Midlands along the river Aare and next to the Jura, or along the Rhine between the Jura and the Black-Forest (Germany).

Only one facility has been planned in the alpine region, in the transversal valley of the upper Rhine.

Fig. 2



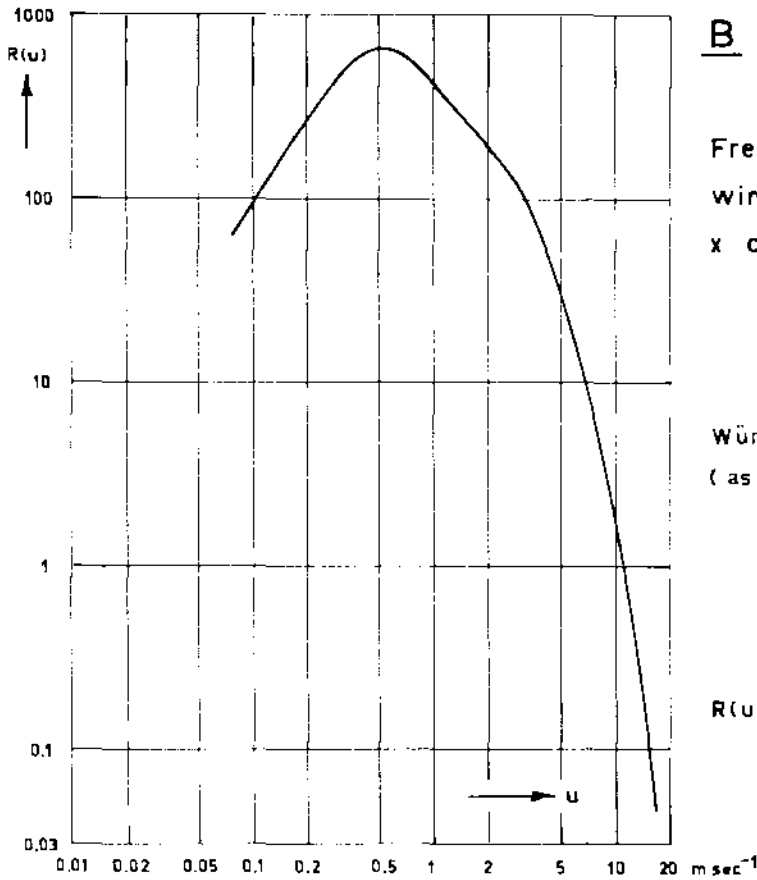
A

Frequency-distribution  
of wind velocities

Würenlingen 1965-67  
760 days  
Hourly averages (18240 values)

$h = 70 \text{ m}$

$P(u) =$  number of cases  
pro  $du = 0.1 \text{ m sec}^{-1}$



B

Frequency of a definite  
wind velocity  
 $\times$  corresponding factor  $1/u$

Würenlingen  
(as in A)

$R(u) =$  number of cases  
 $P(u) \cdot 1/u$   
pro  $du = 0.1 \text{ m sec}^{-1}$

A MODEL FOR THE EVALUATION OF THE DEEP OCEAN DISPOSAL  
OF RADIOACTIVE WASTE

G.A.M. Webb and F. Morley

National Radiological Protection Board,  
Harwell, Didcot, Berkshire, England

Abstract

A mathematical model is developed to predict the eventual surface water concentrations of radioactivity as a result of continual deep ocean disposal of radioactive wastes over hundreds or thousands of years. The behaviour of each nuclide is a function of the interaction between its half-life and the diffusion parameters. Uncertainties in the vertical diffusion coefficient are shown to have little effect on the final concentration values for nuclides with long half-lives.

The processes of release, diffusion, circulation, uptake by fish and eventual transfer to man are used to calculate doses to man. These are then compared with ICRP limits to suggest limiting rates for continuous disposal of various classes of radionuclides.

Introduction

A group of experts was set up by the European Nuclear Energy Agency of the Organisation for Economic Cooperation and Development to carry out a hazard assessment<sup>1</sup> during the planning stages of the first international operation for the ocean disposal of packaged radioactive wastes in 1967. Continuing interest in ocean disposal has made it desirable to carry out a new assessment based on the latest available data to find the limits for continuous disposal of radioactive materials to the sea bed. In particular tritium was not considered in the original calculations, and the estimates were made on the basis of a single release rather than the continuous release model described here.

Radioactivity in the Waste and the Containers used in Sea Disposal

Radioactivity in the waste. Alpha activity is largely plutonium and uranium with small and decreasing quantities of radium. General beta gamma active wastes include mixed fission products and activation products. Since tritium may form a large component of the total arisings and is highly soluble it is considered separately.

Design of the container. The methods considered for making containers utilise concrete or bitumen in metal drums. The design is such that water penetrates into the package which will then descend to the full depth of the sea without damage. It is not known what the long-term effect of sea water at high pressures may be on the structure.

Physical Transport and Dilution of Radioactivity in the Sea

A simplified model of the ocean structure<sup>2</sup> is used as the basis of the method of calculation. This comprises:-

- (a) surface waters down to 500 m which are assumed to be rapidly mixed
- (b) a barrier region from 500 m to 1000 m with a high density gradient acting as a barrier to vertical transport

(c) deep waters from 1000 m to 5000 m, where the principal vertical transport mechanism is diffusion.

All vertical transport in region (c) is taken to be by diffusion and the concentration is calculated just below the barrier region (b) as a function of time after the start of disposal operations, for a continuous rate of disposal. This concentration will tend towards a steady state when the rate of introduction is balanced by the rate of loss by outward diffusion and decay.

Having obtained these concentrations the model then assumes a sectional form<sup>3</sup> in which (a) and (c) are regarded as two compartments separated by a barrier (b). The exchange factor across the barrier is obtained by a separate calculation based on a linear diffusion model. Boundary conditions appropriate to the rapid mixing of the surface waters are imposed.

### Release of Activity

It is assumed that all the tritium is released immediately the container reaches the bottom. The other constituents will probably be retained for long periods; however it has been assumed that all the available activity is released continuously over the 10 year period after disposal. Although most of the activity is in insoluble form and sorption processes will also act to remove radionuclides from solution<sup>4</sup> it is assumed that all the activity disposed of is available for release.

### Diffusion in Region (c)

So little detail is known of vertical diffusion processes in deep ocean and the parameters involved that use of a complex diffusion model is not warranted. The form of the basic solution of the diffusion equation for a single release used is<sup>5</sup>:

$$c_t = \frac{Q}{8(\pi t)^{3/2} (D_x D_y D_z)^{1/2}} \exp - \left[ \frac{x^2}{4D_x t} + \frac{y^2}{4D_y t} + \frac{z^2}{4D_z t} \right] \dots \quad (i)$$

where t is the time after release

$c_t$  is the instantaneous concentration at point (x,y,z) at time t

Q is the quantity of radioactivity released

$D_{x,y,z}$  are the diffusion coefficients in the x,y,z directions

with boundary conditions imposed corresponding to reflection at the bottom and at the barrier (b).

Since the equation including decay cannot be integrated explicitly the concentrations as a function of time were obtained numerically from:

$$c_t = \sum_{t'=0}^t c_{t'} \exp.-(\lambda t') \dots \quad (ii)$$

where  $\lambda$  is the decay constant.

The major uncertainties in the calculation are the diffusion coefficients. Assuming transportation to the barrier region in ten years implies a vertical diffusion coefficient of 24 cm<sup>2</sup>/sec. This could be an order of magnitude too high.<sup>4</sup> Further assuming that in this time the horizontal spread is not more than 100 km implies a horizontal diffusion coefficient of 10<sup>4</sup> cm<sup>2</sup>/sec. This

could be an order of magnitude too low overall<sup>4</sup>, although it may be too high in the early stages.

In the case of the long lived  $\alpha$  emitters, with  $\lambda = 0$ , equation (iii) which is obtained by integrating equation (i) with respect to time for the point above the disposal area such that  $x=y=0$  is:

$$C_t = \frac{Q}{1.2 \times 10^8 (\pi) (D_x D_y D_z)^{1/2} (Z^2/D_z)^{1/2}} \operatorname{erfc} \left[ \frac{Z^2}{4D_z t} \right]^{1/2} \dots (iii)$$

For times such that  $4D_z t \gg Z^2$ ,  $C_t$  is not dependent on the vertical diffusion coefficient  $D_z$  and is inversely proportional to the first power of the horizontal coefficient  $D_x (= D_y)$ . This implies that for the long lived  $\alpha$  emitters, which form the most restrictive class, the actual choice of diffusion coefficients has relatively little effect on the final steady state value of concentration.

#### Transport through Region (b) and dilution in Region (a)

Using the steady state form of the sectional model<sup>3</sup> and assuming no net volume transport and no direct release in section (a) gives:

$$E_b (C_c - C_a) + E_o (C_o - C_a) - V_a C_a = 0 \dots (iv)$$

Where  $C_{a,c}$  are the concentrations of activity in regions (a), (c),  $\mu\text{Ci}/\text{cm}^3$

$C_o$  is the concentration of activity in region Open Ocean,  $\mu\text{Ci}/\text{cm}^3$

$V_a$  is the volume of region (a),  $\text{cm}^3$

$E_b$  is the exchange factor across barrier (b),  $\text{cm}^3/\text{yr}$

$E_o$  is the exchange factor between region (a) and open ocean,  $\text{cm}^3/\text{yr}$ .

It is shown later that it is valid to take  $C_o = 0$  for the open ocean concentration. The water in section (a) is assumed to be changed at least once a year so that  $E_o = V_a \text{ cm}^3/\text{yr}$ . To find a value for  $E_b$  the linear diffusion equation<sup>5</sup> is used which states that the rate of transfer (T) across a barrier is:

$$T = D_z (C_c - C_a) / l \dots (v)$$

The barrier width  $l$  is about 500 m.  $D_z$  is given for a typical ocean barrier<sup>4</sup> as  $1 \text{ cm}^2/\text{sec}$ . Substituting and calculating the concentration in region (a) in terms of  $C_c$  we obtain  $C_a/C_c \approx 0.01$ , so  $E_b = V_a/100 \text{ cm}^3/\text{yr}$ .

/Table 1



Group of Nuclides	$\lambda$ (yr <sup>-1</sup> )	Concentration below the barrier ( $\mu\text{Ci}/\text{cm}^3$ ) after continuous releases for:			Barrier transmission Eqn. (iv)
		30 years	100 years	t $\rightarrow \infty$ years	
Mn-54, Zn-65 Ru-106, Ce-144	0.7	$2.0 \times 10^{-18}$	$2.0 \times 10^{-18}$	$2.0 \times 10^{-18}$	0.006
Fe-55, Co-60	0.2	$1.2 \times 10^{-15}$	$1.3 \times 10^{-15}$	$1.3 \times 10^{-15}$	0.008
H-3	0.06	$2.9 \times 10^{-14}$	$4.7 \times 10^{-14}$	$4.7 \times 10^{-14}$	0.009
Sr-90, Cs-137	0.02	$0.5 \times 10^{-13}$	$1.7 \times 10^{-13}$	$1.9 \times 10^{-13}$	0.01
Ra-226, Pu-239	0	$0.1 \times 10^{-12}$	$0.5 \times 10^{-12}$	$1.5 \times 10^{-12}$	0.01

Table 1. Concentration below the barrier as a function of time after continuous releases at 1 Ci/year and barrier transmission.

In the remainder of the calculations the steady state (t  $\rightarrow \infty$ ) values are used although it must be emphasised that these are upper limits especially for the longer-lived nuclides.

#### Reconcentration of activity and transfer to Man

Fish will reconcentrate the radionuclides in their flesh. The "concentration factors" used, which are a measure of this increased concentration are the highest given for fish flesh<sup>4,6</sup>.

The intake of activity, I, by members of a critical group is given by:

$$I = C_a \times F \times 300 \times d \text{ } \mu\text{Ci/day}$$

where F is the concentration factor

300 is the total daily intake of protein as seafood, g/day

d is the dilution due to the distance of the fishing grounds from the disposal area. This distance is taken as 700 km for the calculations<sup>7</sup>.

#### Comparison with ICRP Recommendations

This assessed intake can then be compared with the ICRP recommendations<sup>8</sup> for adults since

$$\text{MPI} = 1/10 \text{ MPC}_w \times 2200$$

Where MPI is the maximum permissible daily intake by individual members of the public

$\text{MPC}_w$  is the ICRP recommended maximum permissible concentration in water for occupational exposure at 168 hours per week

2200 g/day is the average adult water intake

Note that in the calculation for tritium the  $\text{MPC}_w$  has been revised using a Quality Factor of 1.

Nuclide	$C_a$ $\mu\text{Ci}/\text{cm}^3$	F	Assessed Intake I $\mu\text{Ci}/\text{day}$	MPI $\mu\text{Ci}/\text{day}$	MPI/I or Limiting Environmental Capacity <sup>9</sup> Ci/yr
Mn-54	$1.2 \times 10^{-20}$	300	$2.2 \times 10^{-17}$	$2 \times 10^{-1}$	$1 \times 10^{16}$
Zn-65		4,000	$2.9 \times 10^{-16}$	$2 \times 10^{-1}$	$6 \times 10^{14}$
Ru-106		0.05	$3.6 \times 10^{-21}$	$2 \times 10^{-2}$	$5 \times 10^{18}$
Ce-144		0.3	$2.2 \times 10^{-20}$	$2 \times 10^{-2}$	$1 \times 10^{18}$
Fe-55	$1.0 \times 10^{-17}$	3,000	$1.8 \times 10^{-13}$	2	$1 \times 10^{13}$
Co-60		80	$4.8 \times 10^{-15}$	$1 \times 10^{-1}$	$2 \times 10^{13}$
H-3	$4.2 \times 10^{-16}$	1	$2.5 \times 10^{-15}$	10	$3 \times 10^{15}$
Sr-90	$1.9 \times 10^{-15}$	0.2	$2.3 \times 10^{-15}$	$8 \times 10^{-4}$	$3 \times 10^{11}$
Cs-137		40	$4.6 \times 10^{-13}$	$4 \times 10^{-2}$	$1 \times 10^{11}$
Ra-226	$1.5 \times 10^{-14}$	200	$1.8 \times 10^{-11}$	$2 \times 10^{-5}$	$1 \times 10^6$
Pu-239		10	$9.0 \times 10^{-13}$	$1 \times 10^{-2}$	$1 \times 10^{10}$

Table 2. Assessed intake for the critical group at steady state following disposal of 1 Ci/year on the sea bed compared with ICRP recommended intakes for individual members of the general public and hence the Limiting Environmental Capacity<sup>9</sup>.

Long-term limitations of the model

The major limitation on the model proposed is the cumulative contamination of the ocean. There will not be any serious breakdown of the model until the concentration at the limits of the disposal area due to general contamination exceeds 1% of the concentration at this point due to diffusion from the area itself. The model is therefore satisfactory for times such that

$$\frac{10^6 \int_{t'=0}^t e^{-\lambda t'} dt'}{V_A} < \frac{A_t}{100} \quad \dots (vi)$$

where  $V_A$  is the volume of the Atlantic Ocean

$A_t$  is the concentration at the barrier for the nuclides with decay constant  $\lambda$ .

For nuclides with long half-lives so that  $\lambda = 0$ , the condition reduces to  $t < 4,500$  years.

For nuclides with  $\lambda = 0.002$  the inequality holds for all values of  $t$  so that nuclides with these or shorter half-lives cannot build up sufficiently to invalidate the model.

### Conclusions

A mathematical model, valid for continuous disposal for times up to several thousand years, has been used to calculate the eventual surface water concentration of eleven typical radionuclides as a result of continual deep ocean disposal. By comparing the assumed intake of contaminated fish with ICRP recommended limits, values for the limiting environmental capacities are obtained. The safety factor inherent in the model is several orders of magnitude.

In practice it is convenient to divide the nuclides into three classes,  $\alpha$ -active,  $\beta/\gamma$  active and tritium. Assuming for  $\alpha$  and  $\beta/\gamma$  wastes that all the activity in a class is the most restrictive nuclide and that combinations of all three classes are permitted, the limiting rates for continuous disposal in the designated area are estimated to be:

$\alpha$ -active waste (based on $^{226}\text{Ra}$ )	$10^6$	Ci/yr
$\alpha$ -active waste (based on $^{239}\text{Pu}$ )	$10^{10}$	"
$\beta/\gamma$ active waste (excluding tritium)	$10^{11}$	"
tritium	$10^{15}$	"

These rates are derived from the ICRP Dose Limits for members of the public.

### Acknowledgments

The authors would like to thank Messrs. A. Preston, H.W. Hill and J.W. Talbot of the Fisheries Research Laboratory of the Ministry of Agriculture, Fisheries and Food, Lowestoft, for helpful discussion and suggestions.

### References

1. Radioactive Waste Disposal Operation into the Atlantic 1967, OECD/ENEA, Sept. 1968.
2. Principal Features of the physical Oceanography in the Iberian Basin. V. Romanovsky. OECD/ENEA Seminar, Lisbon 1967 (N.F.P).
3. Radioactive Waste Disposal into the Sea. IAEA Safety Series No.5, Vienna 1961.
4. Radioactivity in the Marine Environment. National Academy of Sciences, 1971.
5. Mathematics of Diffusion. Crank. Oxford Clarendon Press 1956.
6. Atlas of the living resources of the seas. Food and Agriculture Organisation of the United Nations. Rome, 1972.
7. Radiocology of Aquatic Organisms. G.G. Polikarpov. N. Holland, 1966.
8. Recommendations of the International Commission on Radiological Protection Publication 2. Pergamon Press, London (1959).
9. Atomic Energy Review, 9, 4, pp 853-868 (1971).

IMPACT OF TEN YEARS LIQUID WASTE MANAGEMENT  
PRACTICES ON ENVIRONMENTAL SAFETY AT BRADWELL POWER STATION, U.K.

M.M. Wasson

Central Electricity Generating Board, Bradwell Power Station,  
Bradwell-on-Sea, Southminster, Essex, England

N.T. Mitchell

Ministry of Agriculture, Fisheries and Food, Fisheries Radiobiological  
Laboratory, Hamilton Dock, Lowestoft, Suffolk, England.

Abstract

In the light of present concern over the environmental consequences of radioactive waste disposal from nuclear power, the paper discusses the experience gained in the control of discharge of low level liquid effluent during the first ten years of operation of one of the first CEGB nuclear power stations, at Bradwell in south-east England. Public radiation exposure from liquid waste disposal has been kept to a very low level, never being more than a small fraction of one percent of the I.C.R.P. recommended dose limit. That this has been attained is largely due to the actions taken by the Power Station to counteract the changes in effluent composition so as to minimise the significance of discharges and their consequences.

1. Introduction

Bradwell Power Station<sup>(1)</sup> is one of the first civil nuclear power stations to be operated by the Central Electricity Generating Board. Operation of its two 537 MW(th) CO<sub>2</sub>/Graphite (Magnox) reactors started in 1962 and  $2 \times 10^{10}$  KWh<sup>(2)</sup> has since been exported, representing a load factor since startup of 75.2%. It is situated at the mouth of the estuary of the River Blackwater into which the liquid effluent is discharged. Control<sup>(2)</sup> of the disposal of radioactive waste is the responsibility of central government, the CEGB working to limits imposed jointly by the Department of the Environment and the Ministry of Agriculture, Fisheries and Food. In common with procedures adopted throughout the country, waste disposal is only permitted by statutory authorization issued by these two departments; for Bradwell this specifies limits on total activity (other than tritium), with individual limits on tritium and zinc-65. Discharges are made from delay tanks in which the effluent is collected, and monitored for total activity and zinc-65, before being released to the cooling water outflow. Because of the tidal nature of the estuary and to ensure maximum dispersion to the open sea, discharges are voluntarily confined to the three hours immediately following high tide.

2. Sources of Radioactive Waste and their Management

As in all CEGB reactors, fuel changing at Bradwell is a continuous on load process, the elements being stored after discharge from the reactors for a minimum of 90 days in a water filled pond to allow for fission product decay before despatch off site for reprocessing. This pond water is chemically treated on a continuous cycle of 6% per day, to minimise corrosion of the fuel element cans. The plant consists of a non-regenerable pretreatment unit, originally for zinc removal, followed by regenerable cation and mixed bed ion-exchange units. Regenerant liquors from these units, together with wastes from the gas circuit driers account for over 90% of the activity in liquid effluent.

## 2.1 Tritium and sulphur-35

Tritium is the major isotope in terms of quantity discharged but because of its extremely low toxicity and the absence of an environmental reconcentration mechanism, it is the least hazardous. The tritium is produced in the graphite by the reaction  ${}^6\text{Li}(n,\alpha){}_3^3\text{H}$ , but is in ionic equilibrium with stable hydrogen, and hence its concentration in the gas circuit, and therefore rate of removal by the driers, is dependent on circuit moisture content. The rate of production would be expected to decrease as the lithium impurity is burned up; however it is a reduction in circuit moisture levels which has given rise to the decrease in disposals since 1966. In the early years the other major isotope was another activation product, sulphur-35. It is produced by two reactions  ${}^{34}\text{S}(n,\gamma){}^{35}\text{S}$  and  ${}^{35}\text{Cl}(n,p){}^{35}\text{S}$  and its concentration in the gas circuit is also influenced by moisture content. It is thus present in drier liquors. However in this case the major source is the regenerants from the pond water treatment plant; due to adsorption onto the fuel element can surfaces. The quantities of sulphur-35 discharged are shown in Fig. 1; a similar pattern has been found for tritium, although the total quantities are approximately 20 times greater.

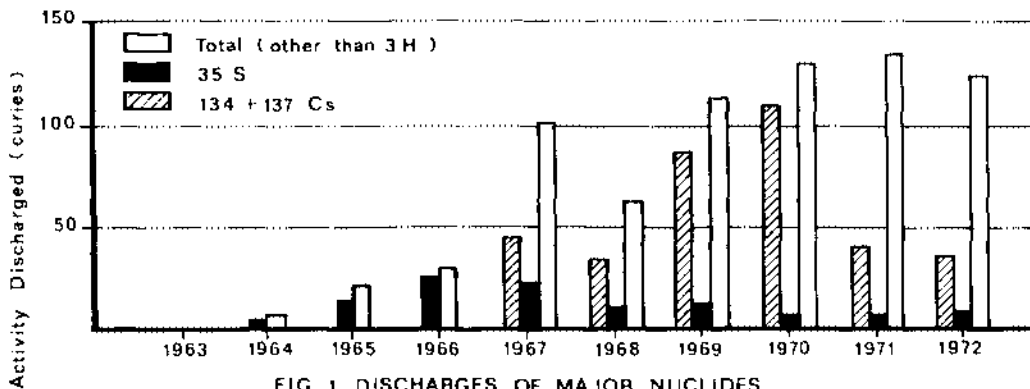


FIG. 1. DISCHARGES OF MAJOR NUCLIDES.

## 2.2 Caesium-137 and -134

Apart from a minor excursion in 1965, the cause of which was quickly traced and eliminated, caesium-137 was a minor contaminant of pond water until 1967. The existing pre-treatment unit had a small capacity for caesium (400 bed volumes) and was rapidly exhausted. Thereafter caesium was removed onto the cation unit and from thence, via its regenerant, to the effluent. The increase in 1967 coincided with the discharge of the first elements of average channel irradiation exceeding 3250MWD/Te but was not associated with any obvious increase in the corrosion of the magnox. Caesium began to accrue at such a rate that without rapid action the authorized discharge limit would soon have been exceeded.

This was a period when extensive refuelling was necessary; simultaneously there were delays in dispatch of fuel off site, so that the problem could not be eliminated without jeopardising electrical output. Although the capacity of the estuary for caesium was much higher than the authorized limit (discharges have to conform to the limit of 200 Ci set on total activity other than tritium) general policy is that discharges should be minimised so far as practicable; hence the prospect of removing and storing caesium rather than discharging it was explored. A literature search, followed by laboratory tests, showed that synthetic zeolites are efficient for removal of caesium and have a high capacity, a very important factor because of the need for shielded storage. In March 1968 the original zinc removal resin in the pre-treatment unit was replaced with a synthetic zeolite and since then, apart from a short period at the beginning of 1972, the pattern of effluent discharge (Fig 1) has been determined by the rate of ingress of caesium-137/134

to the pond water and the performance of the pre-treatment unit in removing it.

From the Bradwell experience it was anticipated that the same problem could arise at other power stations and the CEGB embarked on a programme to provide similar units elsewhere. As a result, the Bradwell unit was used to evaluate other types of zeolite in a range of chemical conditions. In consequence and because of a large backlog of fuel in the pond during this period (1969/71), the caesium concentration in pond water rose to 43  $\mu\text{Ci/litre}$  in late 1971. However, as the backlog of fuel was cleared, the caesium concentration has fallen steadily, to around 1  $\mu\text{Ci/litre}$  by December 1972, thus, by a combination of low input of caesium and steady operation of the treatment plant a very satisfactory control has since been achieved. Since caesium became important in 1967, 268 Curies of caesium-137 have been discharged to the estuary, whilst 2550 Curies have been retained on site in storage as solid waste; figures for caesium-134 are 75 and 750 Curies respectively.

By 1971 total discharges should have begun to fall; however, in December 1970 blockage of a drainpipe resulted in groundwater contaminated with chloride and sulphate entering the pond water. This anionic contamination produced extensive corrosion of freshly discharged elements resulting in the release of other fission products. The more soluble of these appeared immediately in the effluent while others, notably strontium-90, were removed on a sand filter from which they were transferred by backwashing to a sludge storage tank. During January and February 1972 cases were noticed when the delay tank liquor was acid and this was traced to the incorrect neutralising of the regenerant liquors. These liquors are passed to the delay tank via a filter which is backwashed to the same sludge tank as the pond filter and thus the resulting change in the condition of the sludge tank from alkaline to acid took significant quantities of activity into solution. The removal of the anions from the pond water and the correction of the procedure for neutralising the regenerant liquors have now removed this source.

### 2.3 Zinc-65 and silver-110m

It was anticipated that zinc-65, produced by activation of zinc impurity in magnox, would be the critical nuclide and early estimates suggested that it could be the largest single nuclide present. This was not confirmed in practice, although it did become the critical nuclide. The removal efficiency of the pre-treatment unit was lower than anticipated but sufficient for satisfactory control; however this was improved when the existing resin was replaced by synthetic zeolite in 1968, for this material, proved more efficient at zinc removal. In consequence discharges dropped sharply and have been extremely low ever since (Fig.2).

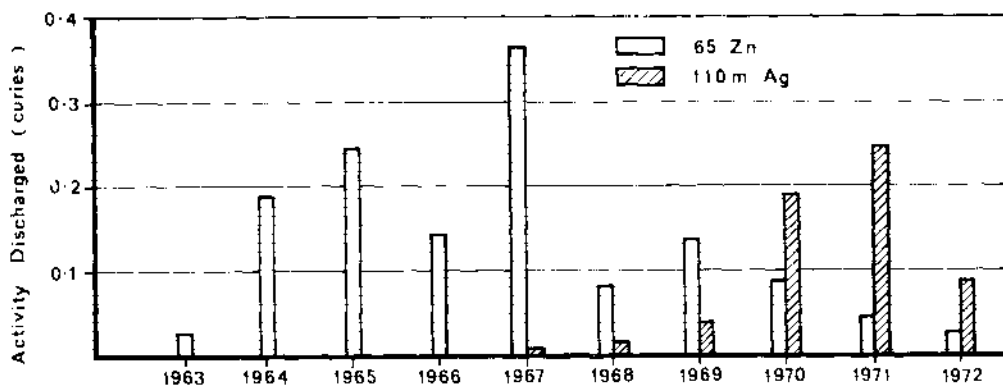


FIG. 2. DISCHARGE OF CRITICAL NUCLIDES.

At the time when discharges of zinc-65 were declining those of silver-110m, a shielded fission product but also a nuclide which can be generated by neutron activation of stable silver, began to increase. There was no evidence to suggest that its source was fission, although massive amounts are present within irradiated fuel, instead it was traced to a chromating solution used to mark high temperature fuel elements for easy identification. From fuel element records it was found that certain elements had been treated using a bath which contained silver nitrate at 0.02%W/V, incorporated to give the stain a darker colour, and the increase in silver-110m in effluent coincided with discharge of these elements from the reactors. Silver is now omitted from the chromating bath and with clearance of these elements the level has now fallen and discharges of silver-110m have declined. They will not disappear entirely because there is metallic silver associated with a spring-loaded lifting head on each element, but discharge rates should be extremely low.

### 3. Assessment of Discharge Limits

The methodology of control of radioactive waste disposal has been developed through more than 20 years of experience along the lines of a system which has come to be known as the critical path approach. For Bradwell the identity of oyster consumption as the critical pathway was established in the course of the first habits survey conducted whilst the power station was still under construction; its importance has been confirmed by two further surveys, from which it has been established that, although oysters are eaten nation-wide, the highest consumption rates are observed within the local population, especially the oyster fishermen and their families.

The capacity of the Blackwater Estuary for the safe reception of radioactive waste from Bradwell, a crucial factor in planning routine disposals, was calculated in terms of exposure via this pathway, initially on the basis of an assumed composition<sup>(3)</sup> later to be recalculated<sup>(3)</sup>, for nuclides actually found to be present in effluent. The initial assessment predicted that zinc-65 would be the critical nuclide, a reflection of the extremely high reconcentration factor, defined as concentration in oyster, concentration in seawater, of the order of  $10^5$ , exhibited by this nuclide in oysters, and this has been borne out in practice. The environmental capacity for zinc-65 was originally estimated at 0.2 Ci/day on the basis of a simple hydraulic model: refining the assessment by allowing for interaction with sediment, the estimate rose to 0.8 Ci/day, to be recalculated soon after operation began (on the basis of effluent and environmental monitoring) at 0.5 Ci/day. Meanwhile, the provisional authorization had been set at 50 Ci per year for total activity, with a specific restriction of 5 Ci/year on zinc-65. This was later relaxed to permit up to 200 Ci/year of total radioactivity, other than tritium, to be discharged while the zinc limit remained at 5 Ci/year and a separate clause for tritium was introduced at 1500 Ci/year. This authorization is still in force.

### 4. Environmental Impact of Waste Discharge

As anticipated before construction of the power station was complete the critical pathway involves internal exposure due to oyster consumption. Whilst traces of radioactivity have been found in other materials such as seaweed and sediment, these only represent minor pathways and their impact in terms of public radiation exposure has been negligible - lower even than that due to oyster consumption.

The first artificial radionuclide to be detected in oysters was zinc-65 (Fig.3) and though in this early phase of operation other radionuclides were present in effluents none reached detectable limits in oysters, so that for several years the whole of such little public radiation

exposure as was occurring, was due to zinc-65. The emphasis was on oysters and most of the analytical effort was directed to gamma spectrometry, though samples were also counted for their content of beta activity, intended as a preliminary to subsequent precise analysis of such individual beta emitters as could not be analysed by gamma spectrometry. Oysters were sampled on both sides of the estuary by CEGB and particularly extensively by MAFF. Monitoring by MAFF in these formative years was a combination of radiological control and environmental research, though much of the latter was a feedback into the control system, increasing understanding of the behaviour of zinc-65 and improving the precision with which radiological assessment could be made. This research<sup>(4)</sup> showed that concentration could be described by the equation  $C = 1.43 \times D^{-0.81}$  (where  $C = {}^{65}\text{Zn}$  pCi/g wet wt. and  $D =$  distance in statute miles from the outfall) and confirmed that the highest concentrations to be found commercially available oysters were in the beds nearest to the outfall, some 500 metres downstream.

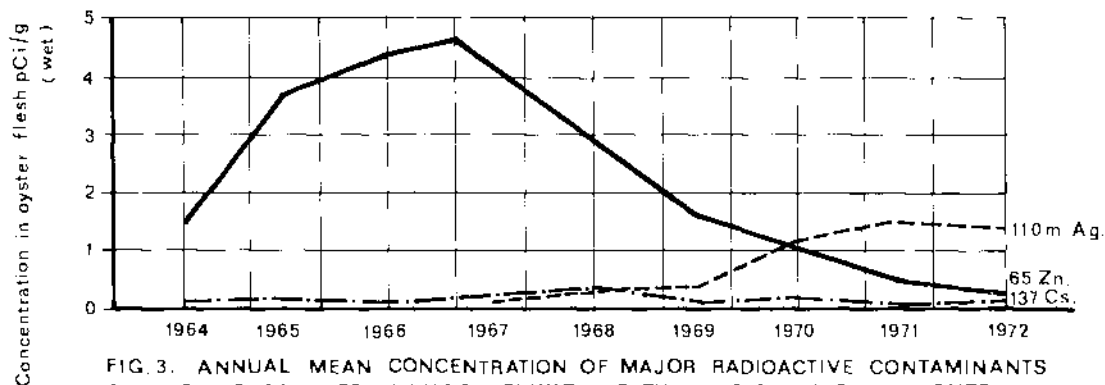


FIG. 3. ANNUAL MEAN CONCENTRATION OF MAJOR RADIOACTIVE CONTAMINANTS OF OYSTERS SAMPLED IN THE BLACKWATER ESTUARY 500m EAST OF OUTFALL.

The first notable change occurred with the appearance of neutron-activation products, notably cobalt-60 and iron-55, the fission products caesium-134 and -137, and silver-110m, another neutron-activated product. The emergence of tritium and sulphur-35 as major components of the effluents had no effect at all nor has strontium-90 been detected despite being a major component of effluent at the beginning of 1972. Low concentrations of phosphorous-32 could occasionally be found but were of negligible radiological significance.

Caesium-137 had been present at just detectable levels for some years, due to fallout. As the two caesium nuclides became the dominant components of the effluent so their concentrations in oysters increased. However, the measures adopted by the CEGB to minimise discharges to the estuary arrested this rise, and after reaching a peak in 1968 concentrations have fallen (Fig.3). Coincident with this the zinc-65 concentration in oysters has also declined and is now less than one-tenth of the peak value in 1967. As zinc-65 declined, small but increasing quantities of silver-110m started to appear<sup>(5)</sup>. Silver 110m reconcentrates in oysters to a very high degree, probably a little higher even than zinc-65, and, in a contrast sharpened by the decline of zinc-65, has now become the critical nuclide.

Cobalt-60, iron-55 and zinc-65 are largely associated with sediment and, whilst they have been detected in surface mud/silt, concentrations have never reached levels sufficient to make the ambient beta/gamma radiation dose rates in these areas significantly different from natural background.

### 5. Public Radiation Exposure

Throughout the operation of the power station public radiation exposure through oyster consumption has been kept down to a very low level. This is illustrated by Fig.4 which shows the extent of public radiation via two body organs - the GI Tract and Total Body. Maximum dose rate in any year of operation has been less than 3 mRem to the GI Tract, a very small dose



compared with natural background and less than normal fluctuations in it. These calculations are based on a consumption rate of 75 g/day of oysters harvested from the nearest commercial source to the power station. This consumption rate is the maximum found in the course of the habits surveys and that on which recent assessments of environmental capacity and subsequent control measures have been based. These estimates of exposure are therefore extreme values and as the locally-exposed population group is too small and too heterogeneous for it to be likely that a critical group could be identified within the ICRP framework, exposure has continued to be evaluated from monitoring data on this pessimistic basis.

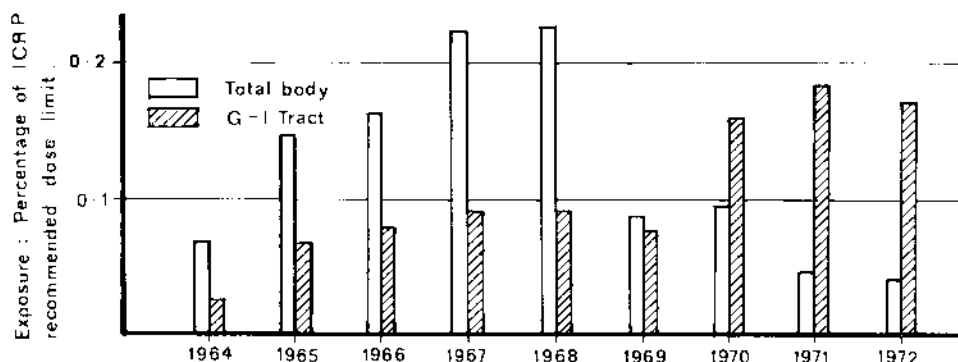


FIG. 4. RADIATION EXPOSURE OF AN INDIVIDUAL CONSUMING 75 gm / DAY OYSTER FLESH

It will be seen therefore that whilst zinc-65 was the critical nuclide total body was the critical organ. These exposure estimates include the contribution from radiocaesium, though this was relatively small until the importance of zinc-65 declined. As silver-110m took over the role of critical nuclide so that identity of the critical organ changed to the gastrointestinal tract, estimates of exposure of which, in Fig. 4, include contributions from zinc and radiocaesium, small though these are.

#### 6. Conclusions

In the course of more than ten years of operation the nature of radioactive waste and the range of management problems it has created have varied widely. Initially only one nuclide, zinc-65, determined the extent of public radiation exposure from waste disposal, though it has since been replaced in this role of critical nuclide by silver-110m, also only a minor constituent of the waste discharged. Changes such as this and just as unexpected changes in major constituents, particularly caesium-137 and -134, have generated problems calling for flexibility of approach to the management of wastes and particularly their treatment.

This policy has proved eminently successful, for despite the formidable constraints posed by the restricted dispersion of a small estuarine environment and the presence of an important oyster fishery, public radiation exposure has been kept down to extremely low levels. The mean dose rate to the most highly exposed known individual during this period, in somatic terms, has been of the order of 0.1 per cent of the ICRP recommended dose limit, whilst the genetically significant dose is counted in milli man-rems per year.

#### 7. Acknowledgments

The assistance of W. Godfrey (CEGB) and of D.F. Jefferies and L. Woolner (MAFF) in the preparation of this paper is gratefully acknowledged.

### References

1. Nuclear Power Vol. 7 pp. 78-101, April 1962.
2. ANON: The Radiactive Substances Act (1960), HMSO, London.
3. PRESTON, A., The United Kingdom approach to the application of ICRP standards to the controlled disposal of radioactive waste resulting from nuclear power programmes. In: Environmental Aspects of Nuclear Power Stations, pp. 147-157, IAEA, Vienna, 1971.
4. PRESTON, A., The control of radioactive pollution in a North Sea oyster fishery. Helgolander wissenschaftliche Meeresuntersuchungen Vol. 17, pp. 269-79.
5. PRESTON, A., DUTTON, J.W.R. and HARVEY, B.R., Detection, estimation and radiological significance of silver-110m in oysters in the Irish Sea and the Blackwater Estuary. Nature, Vol. 218, pp 689-690, 1968.

ENVIRONMENTAL RADIATION FROM ADVANCED GAS-COOLED  
REACTORS IN NORMAL OPERATION

F. H. Passant

Central Electricity Generating Board  
Cheltenham, Gloucestershire, England

Abstract

The normal operation of an AGR power station may lead to the release to atmosphere of several radioactive nuclides each of which may contribute some small radiological dose to members of the public in the immediate vicinity of the power station. The significance of the release is examined in the context of present and possible future radiological criteria.

The nuclides of significance are predicted to be  $^3\text{H}$ ,  $^{16}\text{N}$ ,  $^{35}\text{S}$ ,  $^{41}\text{Ar}$ ,  $^{88}\text{Kr}$  and  $^{131}\text{I}$  and estimates of the likely releases of each are given for a typical 1250 MW (e) AGR power station (Hinkley Point 'B'). The sources of each nuclide are explained and a brief description of the relevant design features of the AGR is given. It is shown that the releases are almost entirely due to leakage of  $\text{CO}_2$  from the primary coolant. Maximum permissible release rates for each of the nuclides are derived for the power station, which is sited in a farming area, based on the ICRP recommended annual doses to members of the public taking into account the critical route for each nuclide (e.g. inhalation, milk, etc.) and the critical group (e.g. 6 months old child).

The predicted releases are compared with the ICRP-derived permissible releases, and with permissible releases based on the current CEGB radiological design criteria and possible future criteria in the U.K. or elsewhere (e.g. the proposed U.S. criteria).

1. Introduction

Much has been published and considerable public interest has been expressed about radioactive releases from nuclear power plants, particularly in the U.S.A. Particular attention has been focussed by environmentalists and other concerned bodies on continuous operational releases leading to chronic exposure of members of the public to radiation in excess of the natural background level. Such concern in the U.S.A. has led to what is regarded by many as an irrational approach to gaseous active waste management in which cost/benefit plays little or no part and many millions of dollars are spent in attempting to reduce discharges already at extremely low levels in terms of risk to members of the public. Such an approach by designers or by authorising bodies has not been adopted in the U.K. The Central Electricity Generating Board has, however, considered it appropriate to define its own design release criteria which are significantly more restrictive than those derived directly from the Recommendations of the International Commission on Radiological Protection. They are less restrictive, however, than the requirements of the U.S.A.E.C. in Appendix 1 to 10 CFR 50.

This paper examines the predicted releases to atmosphere from a typical Advanced Gas-Cooled Reactor (AGR) in normal operation and compares these with current criteria. Liquid effluent releases at AGRs are made into tidal coastal waters and, although not dealt with here, lead to insignificant doses to members of the public.

2. The Advanced Gas-Cooled Reactor (AGR)

There are five AGR power stations under construction in the U.K. at the present time, raising to power of the first of these, Hinkley Point 'B', being expected in February 1974. Each station has two 1500 MW (thermal) reactors. The stations are situated on coastal sites three of which (Dungeness, Hinkley Point and Hunterston) are rural sites already housing nuclear power stations of the magnox type, the other two (Hartlepool and Heysham) being semi-urban

sites. The site boundary for each station is at a distance of about 100 m from the reactors, the nearest grazing land for dairy cattle being at about 400 m except in the case of Dungeness where it is at 3.5 km. None of the sites have cooling towers or any other structure more than about 70 m high neither are there any peculiar meteorological features that adversely affect effluent dispersion.

The remainder of this paper is based on Hinkley Point 'B' power station arisings, this site being the limiting one of the five in the context of gaseous effluent releases.

### 3. Gaseous Waste Management at Hinkley Point 'B'

The potential sources of radioactive releases to atmosphere from an AGR in normal operation are (i) reactor coolant leakage, (ii) discharges from the irradiated fuel route, (iii) ventilation air from contaminated areas, (iv) ventilation of the pressure vessels during man access for inspection or maintenance, and (v) reactor coolant discharges (blowdown of reactor or charge machine).

Reactor coolant leakage is collected in the contaminated ventilation systems which also collect ventilation air from all potentially contaminated areas on the station. The air is passed via 'absolute' filters to discharge points some 2 m above the reactor building roof (64 m). Reactor coolant discharges are made through the reactor and charge machine blowdown plant which incorporates charcoal iodine adsorption plant and 'absolute' filters. Release to atmosphere is again at 2 m above roof height. Cooling air from the irradiated fuel discharge route is treated in the reactor and charge machine blowdown plant before discharge with ventilation air. During man access inside the reactor concrete pressure vessels a breathable atmosphere is maintained by ventilating the vessel. The air discharged is also passed via the reactor and charge machine blowdown plant to the ventilation discharge duct.

As a result of the treatment of atmospheric discharges outlined above the only significant radioactive releases in normal operation will be the gaseous or volatile constituents in reactor coolant leakage and blowdowns. Provision has been made for the addition at a later date, if necessary, of charcoal filters in the contaminated ventilation systems which handle reactor coolant leakage.

### 4. Sources of Radionuclides Likely to be Released from AGRs

The only significant source of radioactive release to atmosphere is the reactor CO<sub>2</sub> coolant, the activity of which arises due to three mechanisms:

- (i) Direct activation of coolant in the core giving rise to nitrogen 16 by the  $^{16}\text{O} (n,p) ^{16}\text{N}$  reaction on oxygen in the CO<sub>2</sub> and to argon 41 by the  $^{40}\text{Ar} (n,\gamma) ^{41}\text{Ar}$  reaction on impurity argon in the coolant.
- (ii) Activation and subsequent release to the coolant of graphite moderator impurities. The nuclides of concern are sulphur 35 from the  $^{34}\text{S} (n,\gamma) ^{35}\text{S}$  and  $^{35}\text{Cl} (n,p) ^{35}\text{S}$  reactions and tritium from the  $^6\text{Li} (n,\alpha) ^3\text{H}$  reaction. The impurity levels of S, Cl and Li in moderator graphite are approximately 50, 2 and 0.01 ppm respectively. The total mass of graphite in each reactor is of the order of 1000 t.
- (iii) Release to the coolant of fission products from fuel. The only fission product released from intact fuel is tritium, up to 25% of tritium atoms formed by ternary fission being assumed to escape through the stainless steel cans. Gaseous and volatile fission products are released from failed fuel pins to a degree dependent on the extent of coolant/fuel interaction. The only fission products which are radiologically significant from a release to atmosphere aspect are  $^{131}\text{I}$  and  $^{88}\text{Kr}$ .

The full power coolant inventory for each nuclide is shown in Table 1, the  $^{131}\text{I}$  and  $^{88}\text{Kr}$  values being based on operation with one failed fuel pin present.

#### 5. Prediction of Discharges to Atmosphere from Hinkley Point 'B'

For the purpose of estimating the releases of the various radionuclides it has been assumed that one reactor and 250 charge machine blowdowns are carried out per annum. The coolant leakage rate is expected to be up to 1% per day per reactor. The mean daily and annual discharges for each nuclide are given in Table 1 assuming a station load factor of 75%.

The  $^{131}\text{I}$  and  $^{88}\text{Kr}$  discharge rates are estimated using the assumption that the station is operating with one failed fuel pin present in one of the two reactors continuously. This assumption is pessimistic in view of the very low expected failure rate of fuel and the fact that fuel failures can be identified and removed quickly on load.

#### 6. Derivation of Maximum Permissible Release Rates for Hinkley Point 'B'

Using I.C.R.P. data and recommendations it is possible to calculate Derived Working Limits (DWLs) appropriate to the contamination of foodstuffs and breathing air outside nuclear licensed sites during normal operation. The DWLs for each of the nuclides under consideration are given in Table 2.

The DWLs or maximum permissible doses can be converted to maximum permissible release rates of each nuclide from the station. This involves meteorological calculations and, in the case of the milk routes, knowledge of the deposition velocity of the nuclide onto grass and the transfer factor from grass to milk. The calculations have been performed manually using Reference 1 to estimate the effective stack height and the dispersion or, in the case of the noble gases, by use of the programme WEEERIE (Reference 2). Because of the effect of downdraught and mixing of effluent in the wake of the reactor buildings (caused by discharging the effluent at about roof level) the effective height of discharge is estimated to be only about 10 m. A prevailing wind at Hinkley Point gives rise to concentrations of effluent in air in the direction of the grazing land a factor of about two above those to be expected from uniform distribution of winds.

Examples of the derivation of the maximum permissible release rates of  $^{35}\text{S}$  and  $^{131}\text{I}$  are shown on Figure 1. The ICRP derived maximum permissible release rate for each individual isotope is given in Table 2. It must be pointed out that the doses from each nuclide are additive and account must be taken of this in applying the values in Table 2.

#### 7. C.E.G.B. Release Criteria for Normal Operation

For the purpose of considering the adequacy of the design of nuclear power stations the following criteria have been applied by the C.E.G.B. for releases in normal operation:

- (i) The contamination of foodstuffs and breathing air outside the licensed site must not exceed 0.05 of the appropriate derived working limits (DWLs) when averaged over one year.
- (ii) The maximum daily discharge rate must not exceed four times the daily average rate derived from (i) above.
- (iii) The external radiation dose from gaseous effluents is additive with direct radiation from the station. This total annual external radiation dose to any member of the public at the boundary of the site must not exceed 0.1 Rem assuming 100% occupancy.

In applying these criteria account must be taken of the critical exposure routes and the critical group of members of the public (normally children or pregnant women).

## 8. Comparison of Predicted and Permissible Releases

Table 3 compares the predicted and permissible release rates from Hinkley Point 'B' for each nuclide. The factors of safety in hand over the I.C.R.P. permissible releases and the C.E.G.B. design criteria are shown in the Table by B/A and C/A respectively.

## 9. Discussion

Table 3 shows that the releases to atmosphere from a typical A.G.R. power station (Hinkley Point 'B') are well below the permissible releases derived using I.C.R.P. recommendations on doses to the public. When compared with the C.E.G.B. release criteria one nuclide,  $^{35}\text{S}$ , is seen to only just meet the limit of 0.05 MWL, milk being the critical route. Discharges of other nuclides are well below the C.E.G.B. limits.

The predicted and permissible release rates of  $^{35}\text{S}$  are subject to considerable uncertainty. The release and behaviour of sulphur in the reactor coolant are not well understood, the predictions being based on experience with the AGR prototype at Windscale. Uncertainties also exist in the deposition velocity of  $^{35}\text{S}$  to grass and its transfer to milk. The predictions are thought to err on the side of pessimism but this will only be borne out by station operation. However, provision has been made for installation of charcoal filters in the ventilation systems which treat reactor coolant leakage. Such filters are efficient for both sulphur and iodine removal in the forms existing in the AGR (carbonyl sulphide and methyl iodide) and when installed would reduce the discharges of the nuclides  $^{35}\text{S}$  and  $^{131}\text{I}$  by a factor of at least 20. The total capitalised cost of the extra plant (covering installation and operation over the station life) has been estimated to be £0.2 per kW (present worth). It is not intended to instal these filters unless district survey analyses indicate them to be necessary in order to meet the C.E.G.B. criteria.

Table 4 shows the annual doses resulting from the predicted discharges from Hinkley Point 'B'. It is assumed that a child continually consumes milk from a cow grazing 440 m from the reactors in the case of  $^{35}\text{S}$  and  $^{131}\text{I}$  and in the case of external radiation from noble gases and dose from  $^{222}\text{Rn}$  that the individual maintains 100% occupancy at the site boundary.

Although the doses shown in Table 4 do not meet the U.S.A.E.C. criteria of Appendix 1 to 10 CFR 50, these criteria would be met by the incorporation of the iodine and sulphur removal filters discussed above.

## 10. Conclusions

The predicted discharges of radionuclides from a typical A.G.R. in normal operation lead to annual radiation doses to critical members of the public well below the I.C.R.P. recommended maxima.

The predicted annual doses to members of the public, although meeting the C.E.G.B. design criteria for release of radioactive material, do not meet the U.S.A.E.C. criteria of Appendix 1 to 10 CFR 20. Incorporation of charcoal filters at a cost of about £0.2 per kW would reduce discharges such that these criteria are also met. Such a step is not however anticipated for the C.E.G.B.'s AGR power stations currently under construction.

### References

1. Bryant P. M., U.K.A.E.A. Report AHSB (RP) R 42, 1964.  
Methods of estimation of the dispersion of windborne material and data to assist in their application.
2. Clarke R. H., Health Physics, Vol. 25, 1973.  
The WEBRIE programme for assessing the radiological consequences of airborne effluents from nuclear installations.

Nuclide	Reactor coolant inventory (Ci)	Station Discharge, A, (mCi/day)	Station Discharge (Ci/annum)
$^3\text{H}$	21	300	110
$^{16}\text{N}$	3050	5000	2000
$^{35}\text{S}$	0.2	3	1.1
$^{41}\text{Ar}$	522	8000	3000
$^{88}\text{Kr}$	58	580	210
$^{131}\text{I}$	0.03	0.3	0.11

Table 1. Reactor coolant inventory and station discharge rates

Nuclide	Critical Route/organ of exposure	Derived Working Limit (DWL)	Max. permissible release rate, B, per day (ICRP)
$^3\text{H}$	Inhalation/whole body	$2.10^{-7}$ Ci/m <sup>3</sup>	800 Ci
$^{16}\text{N}$	submersion/whole body	+	46 k Ci
$^{35}\text{S}$	milk/bone marrow	50 n Ci/litre	60 m Ci
$^{41}\text{Ar}$	submersion/whole body	+	860 Ci
$^{88}\text{Kr}$	submersion/whole body	-	520 Ci
$^{131}\text{I}$	milk/thyroid	0.4 n Ci/litre	37 m Ci

+ The total whole body gamma dose must not exceed 0.5 Rem per annum.

Table 2. DWLs and maximum permissible release rates

Nuclide	Max. permissible release rate, C, per day (CEGR) (Ci)	E/A	C/A	Nuclide	Organ	Annual dose m Rem
$^3\text{H}$	40	2670	133	$^3\text{H}$	Whole body	0.2
$^{16}\text{N}$	9200	9200	1840	$^{16}\text{N}$	Whole body	0.05
$^{35}\text{S}$	0.003	20	1	$^{35}\text{S}$	Bone marrow	25
$^{41}\text{Ar}$	170	107	21	$^{41}\text{Ar}$	Whole body	5
$^{88}\text{Kr}$	104	900	180	$^{88}\text{Kr}$	Whole body	0.6
$^{131}\text{I}$	0.0018	123	6	$^{131}\text{I}$	Thyroid	12

Table 3. Comparison of predicted and permissible release rates

Table 4. Annual doses from discharges at Hinkley Point 'B'

IODINE 131

SULPHUR 35

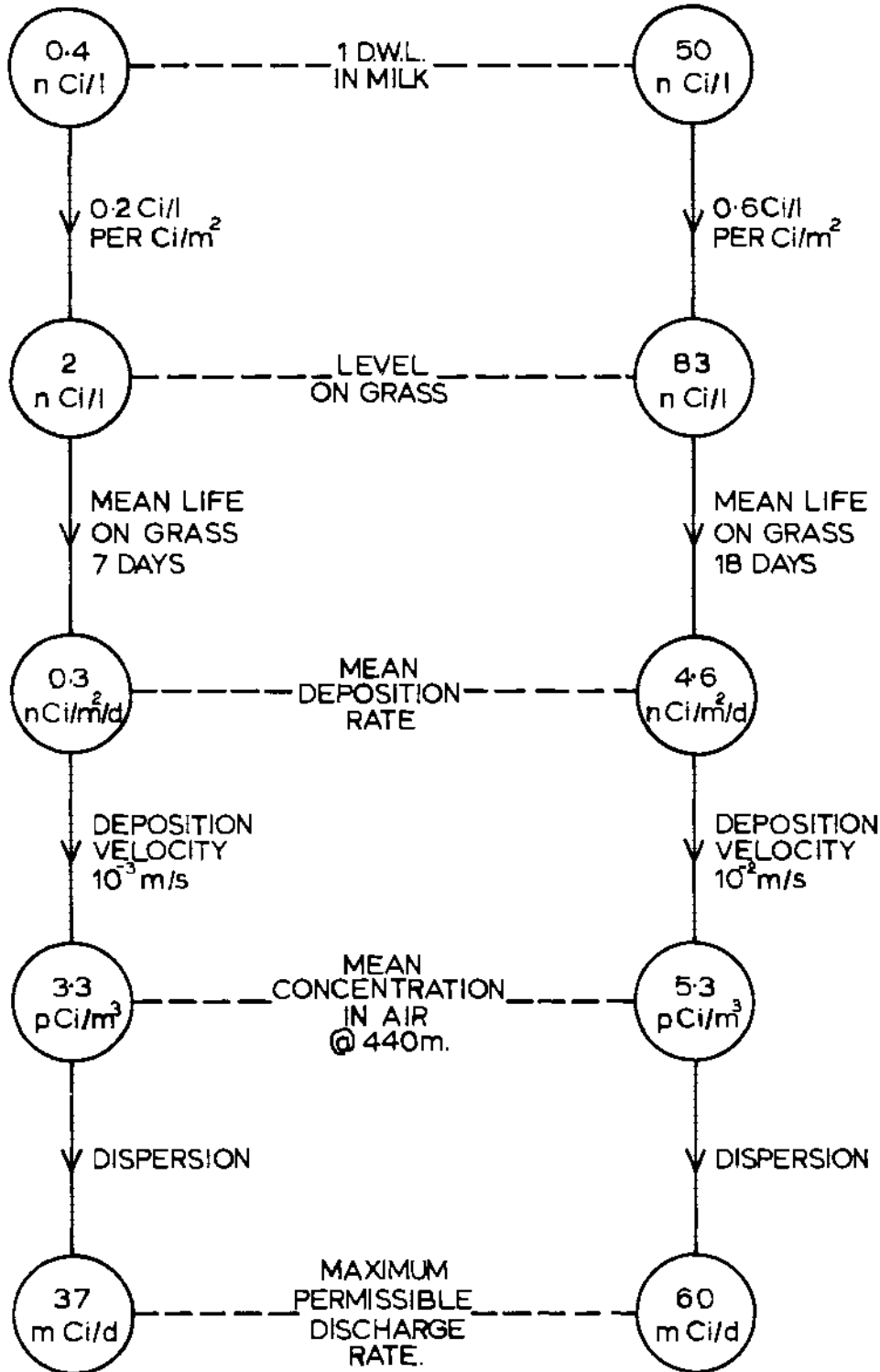


FIG.1. DERIVATION OF MAXIMUM PERMISSIBLE DISCHARGE RATES OF I 131 AND S 35 FOR HINKLEY POINT 'B'



RESEARCH TO KEEP NUCLEAR POWER SAFE:  
THE EXPERIENCE OF A MAJOR INDUSTRY

B. M. Wheatley

Central Electricity Generating Board  
Berkeley Nuclear Laboratories, Berkeley, Glos., U.K.

Abstract

The nationalised electricity industry of England and Wales is a single utility with a present installed capacity of 54 GW of which 3 GW is in nuclear stations. The Central Electricity Generating Board (CEGB) is responsible to the national Parliament for operating its system safely and economically. It necessarily maintains a large research department to assist in obtaining or interpreting the data needed for planning, specifying and operating its plant. Much of the research work is directed to environmental studies, particularly in atmospheric dispersion and hydroecology. In the nuclear field much of the work is concerned with the safety of structures and materials, and so affects radiological safety indirectly. Some of the work, the subject of this paper, is directly concerned with radiological safety and will be described under the headings of Fundamental, Applied and Operational research.

It is concluded that CEGB experience has shown it to be essential for a major utility company to maintain a vigorous health physics research programme.

1. Introduction

The nationalised electricity industry of England and Wales consists of a Central Electricity Generating Board (CEGB), and a number of Area Boards. The CEGB is responsible to the national parliament for generating and transmitting power safely and economically. It has an installed capacity of 54 GW of which 3 GW is in nuclear stations, and sells power to the Area Boards who distribute it for sale to consumers. The CEGB operations are managed on a largely autonomous regional basis with a central headquarters organisation providing major services and some necessary co-ordination. Each Region has a Scientific Services Department, whose efforts are integrated with the headquarters Research Department consisting of three national laboratories situated at Leatherhead, Berkeley and Marchwood. The Central Electricity Research Laboratories (CERL) at Leatherhead have made distinguished contributions to environmental research, particularly in atmospheric dispersion and hydroecology. Berkeley Nuclear Laboratories (BNL) is almost exclusively concerned with the safety and economics of nuclear power. In addition to research on structures, materials and instrumentation, which affect radiological safety indirectly, it also carries out a programme directly concerned with all aspects of health physics. It is the thesis of this paper that it is essential for a major utility company to maintain a vigorous health physics

programme, and an attempt will be made to demonstrate this by a series of examples presented under the headings of Fundamental, Applied and Operational research.

## 2. Fundamental Research

It is the policy of the CECB to protect its workers and the public in the light of the best available knowledge. Some of this knowledge is unpublished and, in order to become familiar with it, it is necessary to establish close links with specialists working in laboratories concerned with the basic sciences contributing to radiological protection. This can only be done by CECB staff who are familiar with the terminology and have sufficient first hand experience to engage in an effective dialogue. Discussion of specific problems often leads to the additional benefit of a deeper understanding of the contributing sciences from which future trends can be more reasonably judged. Since the period elapsing between the ordering and operation of a station is several years it is important to forecast present trends in basic standards, and to be aware of deficiencies in present derived standards, in order to avoid expensive modifications.

The method of working depends on the particular topic and the following examples are chosen to illustrate different balances between execution and sponsorship.

### 2.1 Skin Dose Estimation

In many situations involving spent fuel examination or reactor maintenance the control of skin dose is important. Before instrument readings can be interpreted in terms of skin dose it is necessary to define what is meant by 'skin dose', and to agree over what areas of skin the limits should apply, and what units should be used to express the limits. These deceptively simple requirements raise many questions such as:-

What is the depth and thickness of the layer of cells thought to be at risk? Is it peak or mean dose in that layer which is important? Does ICRP "averaging" procedure permit extrapolation to single hot spots? In view of the energy dependence of skin erythema should there be any modifying factors for dose equivalent in skin? Is it necessary to estimate skin dose in rems, or in rads in air at or near the body surface as suggested by some legislation?

Recognition that these questions have a bearing on day to day working practice<sup>1</sup> provoked discussion by the British Committee on Radiation Units and Measurements and by the Medical Research Council<sup>2</sup>. The CECB had a special interest in the depth and thickness of the layer of cells thought to be at risk because the steepness of beta depth dose curves made those values critical for interpreting skin dosimeter readings. It appeared that these values were not known, and the customary value of 70  $\mu\text{m}$  for the minimum depth of the basal layer of the epidermis certainly did not hold for all exposed body sites. As a result the staff of Berkeley Nuclear Laboratories collaborated with medical staff in London hospitals and carried out with Dr. Everall, a series of measurements whose significance in radiological protection has been described<sup>3,4</sup>. In the course of contributing to the acquisition of new knowledge the CECB staff obtained a familiarity with the radiobiology of skin which was recorded in a series of internal reports which have been useful to designers and operators. The skin work is an example of cases where a deep understanding has enabled the CECB to hold its own valid opinion on the standards used for protecting its workers.

## 2.2 Lung Dosimetry

The lung burdens or intakes of radioactive material giving 15 rem a year are normally calculated by averaging the energy of radioactive decay over a lung mass of 1 kg. When the material has a very high specific activity the permissible quantity of material may be contained in only a few particles. Thus although the mean dose to the lung is 15 rem the distribution of dose is highly non-uniform. Small volumes close to the particles may receive necrotic doses while the rest of the lung is substantially unirradiated. The obvious question is whether or not the risks of a lung burden depends on the specific activity of the material involved. ICRP publications offer conflicting advice, and yet the CEEB must have an opinion on which to base its present operational procedures and future design requirements. Having identified the problem the Research Department took the initiative in getting guidance on the interpretation of ICRP recommendations, and in discussing the research needed to obtain more definitive guidance. The hot-particle problem has been recognised for many years, but by showing its importance in practice the CEEB was able to add impetus to the biological studies. In this case the method of procedure is to operate a small contract with the Medical Research Council Radiobiology Unit at Harwell, and to participate in their work especially on physical aspects. This co-operation works well as the CEEB knowledge of the types of particles, and situations in which they could arise keeps the research work entirely relevant to real problems, and the CEEB develops an informed opinion about an aspect of radiation protection in which guidance is ambiguous.

## 2.3 Bone Dosimetry

The contract method of working is particularly useful when the CEEB wishes to become better informed about specialist studies by contributing to them. The dosimetry of bone seeking nuclides is clearly of importance to the power industry, and it is necessary to follow changes in the methods of determining bone burdens and permissible intakes. Where the utility needs the information it is very proper that it should contribute towards the research necessary to obtain it, and in this case we operated a small contract with Professor Spiers' group at the University of Leeds. Such contract work is fruitful when the contractor can take an intelligent interest, or even participate in some aspects. The sort of contract which was merely financial, and which resulted only in a report, would be of only marginal value.

## 2.4 Dose-risk Relationships

Many of the day to day decisions in health physics rely on an assumed form for the dose-risk relationship, which, following ICRP guidance is usually taken as linear. For example in a maintenance operation the linear hypothesis would see no difference in risks between 40 men receiving 1 rad each and 20 men receiving 2 rads each. Again, on the linear hypothesis the mean organ dose is the parameter of interest and the hot-particle problem disappears. It is perhaps in the environmental aspects of nuclear power that the shape of the relationship becomes extremely important since it influences decisions on the methods of disposing of effluent. The literature on dose-risk relationships is vast, mostly concerned with animal experiments with a little human data. In addition to following this, the CEEB is also concerned to follow up the speculative studies begun by Professor Mayneord some years ago<sup>5</sup>. Together with Mayneord we have developed a model which appears to fit the biological data available and which could have far reaching effects on the fundamental principles of dose control<sup>6</sup>.

## 3. Applied Research

When safety criteria have been specified for working or public environ-

ments it is necessary to determine the constraints in design or operating conditions in order to meet them. The thorough imaginative enquiry that this entails usually involves going back to first principles and following a logical sequence of calculations capable of checking where experience is available. One example of this was an assessment of airborne releases<sup>7</sup>, for which the reactor inventory of fission products<sup>8</sup> and heavy elements<sup>9</sup> had to be generated. The development of this work and examples of its application have been given<sup>10,11</sup>. By carrying out applied research within the utility company several important advantages are secured. Firstly the work is based on first hand information about specific plant and operation conditions, and secondly, the research staff are an integral part of the organisation concerned with drafting and assessing specifications for the purchasing of new plant. Within the CEGB health physics research has identified the limitations on running reactors with failed fuel, it has contributed to stack height specifications, identified shielding problems, and improved the integrity of emergency monitoring services for both workers and the public.

#### 4. Operational Research

As in any industry operational problems arise, some of which are most economically solved by reference to a central laboratory. Within the CEGB excellent relationships exist between the power stations and the laboratories resulting in a long history of research effectiveness. All types of problems arise, some of them involving little effort, others becoming projects involving several years work. Much of the operational work is concerned with ensuring the adequacy of techniques for measuring radiation and radioactivity. This frequently involves investigations at the power stations in which the environment requiring routine monitoring is properly defined by laboratory techniques. The characteristics of commercial instruments can then be determined with a view to giving guidance on the interpretation of their readings in practical situations. Occasionally it is necessary to develop a new instrument because no suitable commercial instrument is available. So far the CEGB has developed or sponsored about half a dozen instruments ranging from an exposure meter with an energy response extending to 6 MeV to a monitor measuring low concentrations of <sup>35</sup>S in gaseous emissions. A good account of one such development has been recently given<sup>12</sup>. As technology advances new measuring techniques become available and it is necessary to study them thoroughly in order to exploit them properly. For example, in recent years thermoluminescence has provided a valuable method of dose estimation whose applications in reactor health physics have been assessed<sup>13</sup>. Any assessment of a measuring technique requires calibration facilities of high integrity, and the CEGB established a facility shortly after its first reactors came up to power<sup>14</sup>. Such a calibration laboratory is concerned not only with accuracy but with determining the characteristics of instruments sufficiently well to have faith in their reliability and confidence in the interpretation of their results under field conditions<sup>15</sup>. The calibration facility is used for checking instruments from all the CEGB stations, and for disseminating sources and instruments which can be used for local checks. The standards used usually have to be developed and related to primary standards where these are available<sup>16</sup>. There is no doubt that the work of the facility has been essential to underwrite the integrity of measurements in the nuclear power industry, and that many of the problems solved would not have been identified if the industry had relied on outside contracts for obtaining opinions on its instruments and interpretations.

#### 5. Conclusions

It is certainly the experience of the CEGB that a vigorous health physics research programme is essential in order for the Board to be an informed buyer of nuclear plant, and to be a responsible operator. The operational work, involving projects providing specific services or hardware is the most easily

understood aspect of the programme, but this part cannot stand on its own. It is backed up by applied research concerned with identifying problems by objective analysis and developing general methods of solution. In turn the applied work has to be underwritten by fundamental work ensuring that the basic science of radiological protection is adequate and properly interpreted,

Within the CECB the Research Department is advisory, and this gives health physics research the advantage of freedom to examine problems without any inhibitions. The integrity of this approach has certainly contributed to the confidence shown by the public and the inspecting Government departments.

The ground rules for radiological protection are discussed by many national and international bodies, whose publications are continually extended and revised as knowledge and technology advance. A major utility relies heavily on such bodies for guidance, which can only be provided in a relevant form if the utilities problems are articulately expressed. The CECB recognises this, and the Research Department, among others, regards communication with national and international bodies as important. Such communication is not just to list new problems on which guidance would be welcome, but to make a positive contribution to the work involved.

#### References

- (1) Wheatley, B.M. and Harvey, J.R. "Skin Dosimetry Problems in Nuclear Operations" CECB Report RD/B/N1007 (1968).
- (2) Vennart, J. and Cryer, Merle A. (eds.) "Radiobiology Forum on Radiological Protection and Skin" Brit. J. Radiol. 45, 610 (1972).
- (3) Whitton, Judi T., "New Values of Epidermal Thickness and their Importance", Health Physics 24, 1 (1973).
- (4) Harvey, J.R. "Alpha Radiation, an External Hazard?", Health Physics 21, 866 (1971).
- (5) Mayneord, W.V., "Radiation Carcinogenesis", Brit. J. Radiol. 41, 241 (1968).
- (6) Mayneord, W.V. and Clarke, R.H., "Carcinogenesis and Radiation Risk: A Biomathematical Reconnaissance", Brit. J. Radiol. Suppl. (in the press).
- (7) Clarke, R.H., "The Waerie Program for Assessing the Consequences of Airborne Effluents from Nuclear Installations" Health Physics 25, (1973).
- (8) Clarke, R.H., "FISP, A Comprehensive Computer Program for Generating Fission Product Inventories" Health Physics 23, 565 (1972).
- (9) Clarke, R.H., "Radiological Implications of Uncertainties in Heavy Element Data", International Conference on "Chemical Nuclear Data", British Nuclear Energy Society, (1971).
- (10) Clarke, R.H., "Physical Aspects of the Effects of Nuclear Reactors in Working and Public Environments", CECB, Berkeley Nuclear Laboratories (1973).
- (11) Macdonald, H.F., "Assessment of Environmental Hazards following Nuclear Reactor Accidents" in "Rapid Methods for Measuring Radioactivity in the Environment" pp 43-54, IAEA, Vienna, 1971.
- (12) Harvey, J.R., Hudd, W.H.R., and Townsend, S. "A Personal Dosimeter which Measures Dose from Thermal and Intermediate Energy Neutrons and from Gamma and Beta Radiation" in Neutron Monitoring for Radiation Protection, IAEA, Vienna, (in the press).

- (13) Gayton, F.M., Harvey, J.R. and Jackson, J.H., "Thermoluminescence and its Applications in Reactor Environments", J. Brit. Nucl. Energy, 11, 125-140 (1972).
- (14) Orton, K.F., "The CECB Dosemeter Calibration Facility" in "Neutron Dosimetry", Vol. 2, pp 529-535, IAEA, Vienna, 1963.
- (15) Clarke, R.W., Lavender, A. and Thompson, I.M.G., "Experience Gained in Operating a Dosemeter Calibration Facility "Health Physics 13, pp 73-82 (1967).
- (16) Thompson, I.M.G., "Experimental Techniques and Standards used for Evaluating and Calibrating Neutron Survey Instruments" in "Neutron Monitoring", pp 639-649, IAEA, Vienna, 1967.

RADIOLOGICAL PROBLEMS ON MAGNOX REACTORS WITH INTEGRAL BOILERS,  
WITH PARTICULAR REFERENCE TO PERSONNEL ENTRY INTO THE PRESSURE VESSEL

H.C. Tresise

C.E.G.B., Oldbury-on-Severn Nuclear Power Station, Bristol BS12 1RQ, England.

Abstract

Radiological data is presented for the first 4 years of operation of Oldbury Nuclear Power Station. Radiation doses to personnel on site are very low; the average whole-body dose is about 0.2 rems annually and the highest cumulative dose in 4 years is 2.6 rems. The total annual dose-commitment for operating, refuelling and maintaining the two-reactor power station is below 100 man-rems.

During periodic entries into the pressure vessel, the main problem is heat. Protective clothing to enable men to work for extended periods at temperatures up to 60°C is described. The special medical requirements and the method of selecting personnel to work under these conditions are indicated.

Following an inspection period, sulphur 35 is released in gaseous form during the dry-out of a reactor core. A monitoring technique is described and surveys confirm that locally produced milk is not affected.

1.

Introduction

At Oldbury-on-Severn Nuclear Power Station each reactor core and the boilers are contained within a single pressure vessel constructed of prestressed concrete with a minimum thickness of 5 metres and lined with a gas-tight membrane. This "integral" design has been adopted for all the power reactors currently under construction in Great Britain. The dose-rate at Oldbury in most operational areas is about 0.03 mrad/hour including the natural background level of 0.01 mrad/hour. Where the coolant gas is extracted from the pressure vessel to be passed through ancillary equipment, the controls are located in areas where the dose-rates do not exceed 0.5 mrad/hour.

Each power station operates as a self-contained unit with the man-power and equipment available on site for normal operation, maintenance and services such as health physics. At present, there is no reserve pool of trained labour available at a central base. The annual statistics of the radiation doses received by personnel at Oldbury-on-Severn Nuclear Power Station have been analysed for the last 4 calendar years and section 2 of this paper summarises this information.

The construction of Oldbury-on-Severn Nuclear Power Station was begun in March 1962 and the first of the two units was commissioned in November 1967. With the current operating temperatures the output sent out from Oldbury is 415 MW.

Oldbury Personnel Radiation Dose Statistics

All personnel at a nuclear power station working in areas of significant radiation dose-rates or where any loose contamination may occur wear a film badge. These films are processed monthly unless the information is required more rapidly. Thermo-luminescent sachets containing lithium fluoride powder are used for extremity dosimeters. All measurements of dose are added into the personnel dose record maintained by computer for each person. There are about 350 people employed at Oldbury and a considerable number of other personnel visit the site regularly and need a film badge on arrival, so over 500 are issued each month. Table 1 shows the number of film badges and replacements issued during the period 1969 to 1972. Damage to films is due mainly to the paper wrapping absorbing water or oil.

Table 1

Annual Totals	Year			
	1969	1970	1971	1972
Film badges issued	7265	7455	6681	6652
Film badges issued to contractors	856	850	604	874
Replacement films issued	136	110	69	69
Films damaged	38	22	16	5
Films lost and not recovered	66	35	19	23
TLD sachets issued	-	-	1241	779
Man-remS (CEGB employees per year (Contractors)	95.3 25.0	78.2 15.1	79.3 8.0	75.3 10.8

Doses below 0.01 rad per month would not produce detectable darkening of the film compared with controls kept in an area where the natural radiation causes an annual dose of 0.10 remS. Anyone employed at Oldbury for a complete year would be issued with 12 film badges and credited with a minimum dose of 0.12 remS. 65 man-remS of the annual total dose to personnel at Oldbury is due to the natural background.

In Table 2, the average and the highest individual radiation dose received by personnel at Oldbury are summarised. Table 2 is for C.E.G.B. personnel employed full-time at Oldbury whereas Table 1 includes regular visitors to the site, such as apprentices and inspectors.

Table 2

Group of Personnel	Number in Group				Individual Dose in MilliremS							
					Average				Maximum			
	69	70	71	72	69	70	71	72	69	70	71	72
Engineers etc.	117	93	93	94	152	162	152	151	540	750	790	470
Admin. Dept.	41	34	34	32	123	121	121	124	160	160	170	160
Operations Dept.	127	117	97	84	207	211	218	300	570	550	660	970
Maintenance Dept.	137	127	119	113	221	151	217	194	1150	760	890	630
Health Physics Dept.	38	40	37	26	173	172	163	187	320	440	340	370
Total	460	411	362	349	187	170	198	201	1150	760	890	970

These figures show the small total dose involved in operating and maintaining a nuclear power station which has achieved load factors of 84% and 73% during 1971 and 1972. The number of personnel is high compared with the staffing at stations in other countries but almost all the work involving radiological exposure is carried out by station personnel. The average monthly dose recorded on the film badges issued to contractors was  $18\frac{1}{2}$  milliremS, including the natural background level.



Table 3 shows the number of persons in various ranges of whole-body and extremity radiation dose for the last four years. On average only 15 persons have exceeded 0.5 rems whole-body annual dose. No-one has exceeded a whole-body dose of 1.2 rems or an extremity dose of 5.5 rems in a year. The 58 doses exceeding 0.5 rems in the last 4 years were received by 51 different persons and the largest whole-body exposure integrated over the last 4 years by anyone at Oldbury is 2.6 rems, while the largest extremity total is 6.7 rems. No-one has had to be restricted from working due to an overdose of radiation, even though most of the personnel are not classed as occupationally exposed.

Table 3

Range of Dose		Number in dose bracket in year			
		1969	1970	1971	1972
Whole Body Dose	Not more than 0.5 rems	560	535	472	425
	Between 0.5 and 1.5 rems	15	6	21	16
	Above 1.5 rems	0	0	0	0
Extremity Dose	Not more than 1.5 rems	574	540	491	437
	Between 1.5 and 3 rems	1	0	2	2
	Between 3 and 7.5 rems	0	1	0	2
	Over 7.5 rems	0	0	0	0

### 3. Problems During Refuelling Operations

All C.E.G.B. reactors are designed for "on-load" refuelling and the parts which enter the neutron flux of an operating core are activated. Dose-rates up to 1000 rads/hour may be encountered during subsequent maintenance work.

After removal from the reactor, the irradiated elements are lowered into a storage area under 6 meters of water which has been treated to minimise corrosion of the Magnox fuel can. The used fuel remains in the Cooling Pond for about 100 days so that the inventory of short-lived fission products is reduced. During this period, some of the outer components of the fuel element are removed but the Magnox can around the uranium remains intact. This "desplittering" process improves the packing fraction of the fuel in the steel flask which transports the used elements for reprocessing. The displittering machine becomes contaminated and dose-rates of 100 rads/hour are measured under water when the equipment is raised for maintenance.

Problems due to the isotope caesium 137 in the treated water of the Cooling Pond have been minimised by:-

- 1) Close control of the chemical composition of the water.
- 2) Retaining irradiated fuel under water for 100 days only.
- 3) Paying particular attention to any fuel used for experimental purposes.

The average concentration of caesium 137 in the Cooling Pond water at Oldbury is about 10  $\mu$ Ci per litre and the total quantity of caesium 137 present is 25 curies. About 15 curies of tritium and 5 curies of all other isotopes are discharged as liquids annually from Oldbury. The dose-rate to personnel engaged in fuel-handling operations above the Pond water is about 5 mrem/hour. The total dose due to this work is 1.8 man-rems per year distributed among the 15 men engaged in fuel handling operations.

To maintain the equilibrium fuel cycle for the two-reactor station, the annual refuelling rates quoted in Table 4 are needed. The actual rates of fuel movement achieved over the last 4 years are shown. There is no large refuelling backlog and the last two years should be typical of future operational conditions at Oldbury.

Table 4

Fuelling Operation	Required Annual Rate	Actual Rate Achieved			
		1969	1970	1971	1972
Channels refuelled	1820	1475	1149	1249	1441
Elements desplittered	12740	4278	8696	12492	10342
Flasks dispatched	64	34	52	63	56

#### 4. Problems During Routine Inspection and Maintenance

At Oldbury one reactor is shut down each summer and the interior of the pressure circuit is entered for a statutory inspection.

The four boilers within each pressure vessel are separated from the reactor core by the boiler shield wall, consisting of 75 cm of graphite and 28 cm of steel, to attenuate the direct gamma radiation from the shut-down core and to reduce the activation of materials during power operation. The steel components contain 0.02% cobalt impurity but measurements indicate that impacted dust particles are responsible for most of the dose rate at present.

Man-access to the pressure vessel is required as soon as possible after the reactor overhaul has begun, so cold water is fed through the boiler tubes while the carbon dioxide coolant is being released. The circuit is then purged with air before man-access is permitted. Inflatable seals are guided into position and prefabricated screens are installed to isolate the particular boiler from the coolant gas flow which must be maintained to remove the reactor after-heat.

The level of loose contamination within the boiler is low but there may be residual pockets of carbon dioxide coolant gas initially and tritium may also be present. The space available does not allow men to wear self-contained breathing apparatus, so full protective suits supplied with air through a trailing hose are used for the first entries. After a period, dust respirators and "coveralls" woven from a mixture of cotton and a polyester thread are adequate.

During boiler entries the metabolic heat rate of a man may be 400 kilocalories per hour. With an average body weight of 70 kg, this would be enough to raise the temperature of the entire body by 5.7 degrees Celsius per hour unless heat can be lost by sweating. About 0.5 kg of body fluid may evaporate per hour and most people experience discomfort if the total loss of fluid from the body exceeds 1½ litres. The full protective suits restrict the evaporation of perspiration from the skin so the man is supplied with cooled air by the use of a Ranque-Hilsch "vortex" tube<sup>1</sup> which separates incoming compressed air at about 4 kg/cm<sup>2</sup> into a hot and a cold fraction. The cooled air is circulated through perforated pipes inside a foam plastic suit which acts as a thermal barrier against the surrounding temperatures. Men wearing these suits have performed arduous work under test conditions for one hour in temperatures of 80°C. Personnel have not yet entered the boiler spaces at temperatures above 60°C because of the difficulty of removing anyone who might be injured. Rescue equipment is installed at the top of each boiler by the first team to enter but fortunately it has not yet been needed.

Nuclear power station personnel do not work in hot environments and confined spaces sufficiently often to develop acclimatisation. Experienced medical observers select prospective workers and check for evidence of general physical disabilities, particularly of the locomotor system, poor vision and auditory defects, psychological disturbances such as a history of claustrophobia, and skin sensitivity to rubber or plastic materials. Specific factors which would influence selection are (a) age, (b) physique, (c) hypertension or

hypotension, (d) infective foci, (e) anaemia and (f) skin disease. There is also an "exercise tolerance test" when the resting pulse rate is measured before the subject steps up on to a platform 45 cm above the ground and down again 30 times in 1 minute. The pulse rate is then taken and checked again after 2 minutes rest, to make sure that it has returned to its normal value.

During a typical inspection programme, the gas circuit is open to air for a period of  $\frac{1}{4}$  weeks during which 250 entries by two or more men are made into the pressure vessel, each lasting for about 2 hours. In addition, about 2000 items are individually logged in and out of the area. Personnel emerging from the boilers may have 100 c.p.s. on the outside of their protective clothing, measured by a scintillation counter probe sensitive to beta and gamma radiation.

The radiation doses incurred during these overhaul periods are no greater than those received during normal station operation. The dose rates measured within the boiler annuli are now about 8 mrad/hour due to beta-gamma radiation of which about 6 mrad/hour is due to the gamma component. Smear samples taken from the gas-side surfaces of the boilers show levels of loose contamination below  $10^{-3}$   $\mu\text{Ci}/\text{cm}^2$  beta-gamma. Small piles of debris are encountered in the bottom of the boilers, usually in corners where there is little flow of gas, and dose-rates may be about 50 mrad/hour, mainly due to beta emitters. Small pieces of stainless steel foil which have become detached from the internal thermal insulation of the pressure vessel liner are occasionally found with surface dose-rates up to 100 rads/hour, mainly due to cobalt 60.

#### 5. Reactor Core Dry-Out Following Periods of Overhaul

During the period when man-access is taking place, dried air is supplied to the vessel which is closed temporarily whenever men are not inside, but in  $\frac{1}{4}$  weeks 400 kg of water vapour may enter and 300 kg of perspiration may be given off by personnel working in the gas spaces. The graphite moderator absorbs part of this moisture which then must be removed before the reactor can be operated at significant power. At the conclusion of an overhaul, boiler access equipment is removed, the reactor closed and the vessel is filled with carbon dioxide gas. The reactor core temperature is then raised gradually, ensuring that the dew point is not reached.

The moisture-laden gas is discharged to atmosphere through the installed blowdown filtration system. During this process, the majority of the radioactivity found is a gaseous form of sulphur 35, a low-energy beta emitter with a half-life of 87 days. The rate of blowdown of the gas has been limited in the last 3 years by considerations of the permissible discharge of this isotope into a milk-producing area. This topic is also discussed in the paper by F.H. Passant to this conference, reference U-0078-R-5.

Considerable work has been carried out at the C.E.G.B. Berkeley Nuclear Laboratories by I.R. Brookes and his co-workers on the mechanism of production and release of sulphur 35<sup>2</sup>. The isotope is produced by neutron irradiation both of the stable sulphur impurity in the graphite moderator and the chlorine impurity in the carbon dioxide coolant gas. During power operation in a carbon dioxide atmosphere, sulphur 35 is circulated as carbonyl sulphide gas and is then deposited on the Magnox cladding of the fuel elements as magnesium sulphide. Hydrogen sulphide is released by the action of moisture when the temperature is raised during the dry-out of the reactor core.

Brookes has also devised a method of measuring gaseous compounds of sulphur 35 rapidly which does not need to be carried out by highly-trained staff<sup>3</sup>. The gas is drawn through 250 ml of neutral potassium permanganate solution at a rate of 2 litres per minute and the sulphur is oxidised to sulphate ion. At the end of the sampling period, clean carbon dioxide gas is passed through to saturate the solution. Hydrogen peroxide is then added

to reduce the permanganate to manganous ion. Following this, the solution is electrolysed between a copper cathode and a porous paper anode coated on the under side with zinc powder. A thin layer of zinc sulphate is deposited on this lower surface behind a 1 mg per cm<sup>2</sup> "Melinox" window. This membrane is mounted directly above an end-window Geiger counter and the sulphur 35 activity can be counted without disturbing the deposited thin source. These novel anodes are prepared in a laboratory but only a few papers in each batch need be calibrated.

The work of P.M. Bryant of Harwell<sup>4</sup> was used to calculate the quantity of sulphur 35 which could be released into a milk-producing area. In 1970, 1971 and 1972, milk samples were collected from farms at distances up to 7 kilometres downwind from the reactors during and after the period of core dry-out. In 1972 air samplers were run at distances of 200 and 500 meters downwind in case the maximum deposition occurred close to the point of release due to entrainment in the lee of the building. The maximum airborne concentration found was  $8 \times 10^{-11}$   $\mu\text{Ci}$  per cm<sup>3</sup> of air, which is orders of magnitude below the M.P.C. Samples of milk and of grass herbage were analysed for sulphur 35 by radiochemical means and the situations during the last 4 years are summarised in Table 5. The last line of the table shows that the measured concentrations of sulphur 35 in milk were far below the derived working level of  $5 \times 10^{-2}$   $\mu\text{Ci}$  per litre in milk, although the rate of blowdown had been increased by 4 times the previous value in each of the last two years.

Table 5

	1969	1970	1971	1972
Total moisture evolved (kg)	640	530	340	140
Duration of moisture release (days)	9	8	6	7
Total CO <sub>2</sub> blowdown (tonnes)	250	370	390	350
Total S 35 evolved (curies)	0.5*	2.0	0.9	0.3
Duration of S 35 release (days)	18	10	20	10
Av. conc. of S 35 in CO <sub>2</sub> ( $\mu\text{Ci}/\text{tonne}$ )	2.0	5.0	2.5	1.0
Max. conc. of S 35 in milk ( $\mu\text{Ci}/\text{litre}$ )	-	$< 15 \times 10^{-6}$	$120 \times 10^{-6}$	$60 \times 10^{-6}$

\*This figure is likely to be an under-estimate because measuring techniques had not been standardised in 1969.

## 6. Conclusions

The radiological data presented, particularly for the last two years, ought to be typical of normal power operation at a two-reactor station with integral boilers inside a prestressed concrete pressure vessel containing the Magnox reactor.

The low radiation dose commitment in operating, refuelling and maintaining this type of station is often not emphasised when discussing the relative merits of different reactor systems. If the recommended annual radiation dose received by occupationally exposed workers were to be reduced by an order of magnitude, it would cause little embarrassment at a station such as Oldbury.

## References

1. U.S. Patent Number 1952281 (1934).
2. I.R. Brookes and S.F. Jones. C.E.G.B. Report RD/B/N1937 (June 1971).
3. I.R. Brookes, S.F. Jones and H.F. MacDonald. To be published.
4. P.M. Bryant. U.K.A.E.A. Memorandum AHSB (RP) M31 (1969).

LONG RANGE TRANSPORT AND DIFFUSION: AN APPROACH TO THE PROBLEM FROM  
THE HEALTH PROTECTION POINT OF VIEW

P.CAGNETTI\* and M.PAGLIARI\*\*

\* Laboratorio Radioattività Ambientale, CNEN, Roma (Italy)

\*\* Centro di Ricerche Termiche e Nucleari, ENEL, Milano (Italy)

Abstract

The problem of transport and diffusion of gaseous releases in the atmosphere after travel distances from some tens up to some hundreds of kilometers is analyzed, with the aim of deriving an expression for the cloud-dosage at ground level. Taking into account the experimental data available at present, a function is calculated which relates the coefficient of diffusion to the time of travel. With very conservative assumptions from the health protection point of view, the radially symmetrical equation of diffusion has been solved for an instantaneous release, and the integrated concentration along the travel direction calculated. The results show that, only after a travel of 100 km the values of cloud-dosage are about ten times lower than that obtained by extrapolation of Pasquill estimates, during category F of stability.

Introduction

The health-physicist is asked sometimes to assess the risk to which the population of a nearby country would be exposed in the event of a release of gaseous radioactive effluents from a source some hundreds of kilometers distant as a result of an accident at a nuclear plant sited in his own country. The present work attempts to provide an evaluation of the dilution of gaseous effluents in the atmosphere over such distances, based on the available experimental data, and taking into account the need to allow for the usual worst-event criteria but not going beyond the bounds of a realistic concept of the physical and meteorological phenomena governing such diffusion.

For the next development of this study it is necessary to recall here some results coming from the statistical theory of diffusion<sup>1</sup>: as well known this theory is concerned with following the trajectories of the various particles of air and evaluating their dispersion by means of statistical models. From the analytical point of view, this involves a Lagrangian-type mathematical approach; without going into the merits of the mathematical development of this theory, it is however worthwhile noting one result which may be helpful to us. Assuming that  $\sqrt{\overline{y^2(t)}}$  is the quadratic mean dispersion of the various trajectories after a time  $t$  has elapsed, it can be shown that the following relationship applies for long times<sup>1</sup>:

$$\overline{y^2(t)} \approx 2 K t \quad (1)$$

where  $K$  is a constant. This result will be utilized in the further development of the present study.

Physical phenomena which regulate atmospheric diffusion and their  
dependence on spatial scale

The moment at which a gaseous emission occurs in the atmosphere near the ground marks the beginning of a continuous sequence of events which results in time in the evolution of the cloud, such as we can observe externally. The increase in the size of the cloud implies the presence of two factors essential to diffusion: in the first place the part of the turbulence spectrum which becomes actively involved in the diffusion is that part whose wavelength is of the same order of magnitude as the cloud whereas, as well known, the turbulence component with a shorter wavelength tends only to mix cloud particles locally and the component with a much longer wavelength tends to move the cloud without changing its dimension; in the second place increasingly elevated atmospheric zones gradually become involved, in which both the turbulence spectrum and the thermal stability change. Within these continuously changing diffusion conditions it is nevertheless possible to distinguish an initial phase, for short periods, during which the part of the atmospheric boundary layer involved is that nearest the ground (surface boundary layer). In this case turbulence is essentially correlated to the aerodynamic characteristics of the terrain (i.e. is a function of the ground roughness parameter) and is substantially regulated by the conditions of ground surface heating and cooling. The large body of experimental data collected to date on this phase is adequate to permit reliable forecasting of dilution in air up to distances of some kilometers over terrain which is not excessively irregular: for this purpose the estimates made by Pasquill<sup>2</sup> provide a good summary of the data collected by several authors.

There is, however, a second phase of diffusion over distances of some kilometers up to some tens of kilometers during which increasingly high regions of the boundary layer gradually become involved in the diffusion. In general, diffusion in this second interval is calculated by extrapolating the data referring to the initial phase, using a technique which, though arbitrary, provides adequate safety margins especially with respect to the stable condition. These calculations are generally accepted as valid, even if no practical experimental data have been collected for such special situations.

Not seldom however, in risk evaluation relative to accidental release, the health physicist is used to assume the continuous presence of category F conditions over distances up to 100 km, and thus to base concentrations in air on the values extrapolated from Pasquill's estimates. Such an assumption seems to be not realistic; we know in effect that during the night when clear sky is present, the thermal stability near the ground increases with decreasing wind force, but the wind direction becomes more and more indetermined, so that the product of the mean wind velocity and the time of travel cannot be regarded as a measure of the distance reached, although it does measure the path of travel. At last, taking into account that the category F is present only during the night (1-2 hours before sunset to 1-2 hours after dawn) it may be said that the continuous presence of highly stable conditions will seldom be verified over distance of more than some tens of km; so, in making calculations for the purposes of public health protection, it would be absurd to make such an assumption for distances of the order of hundreds of kilometers. Let us now analyze the physical and climatological phenomena which play a part in diffusion over such long distances and consider whether it is possible to derive any benefit from the limited amount of experimental data on this scale collected so far.

The transport and diffusion of gaseous effluents at mesoscale level

In covering distances from some to some tens of kilometres the diffusing cloud tends gradually to involve the entire boundary layer up to an altitude of about 1000 m (planetary boundary layer). At these altitudes the turbulence actively involved in diffusion is chiefly that induced by thermal effects, whereas the influence of the terrain with its aerodynamic and thermal characteristics tends gradually to decrease in importance.

It can be said that concentrations are controlled by wind trajectories between the point of emission and the receptor area and by the vertical mixing characteristics along the path of travel<sup>3</sup>: any long distance diffusion model must therefore incorporate parameters which make allowance for the influence of atmospheric stability profiles on the vertical dispersion of the released material. In fact the vertical dispersion of a cloud which has been moving for several hours may be lower than that predicted by the Gaussian model as a result, for example, of the confinement of vertical mixing below a well defined stable layer over long distances<sup>3</sup>. One of the most frequent cases, and therefore a case for which we have made allowance in the proposed model, is an inversion at altitude caused by the subsidence of air masses; the effect of this inversion is similar to that shown in a suggestive picture by R.W. Davies<sup>4</sup>.

The model which we now propose in order to describe transport and diffusion at mesoscale level, i.e. up to several hundreds of km, does not claim to make any scientifically based allowance for the physical phenomena which determine dilution over such distances: actually such a study would be impossible, mainly because of the difficulty of producing experimental data which would serve this purpose. Since some data (albeit from varied sources) are available, however, it was decided to adopt the protectionist standpoint and to make allowance for them by selecting a model which would place an upper limit on all experimental concentration data. Fig.1 gives experimental horizontal dispersion data  $\sigma_y$ , obtained by 19 authors<sup>1</sup> for time of travel up to more than 100 hours, collected by J.L.Heffter<sup>5</sup>: they come from a wide variety of experiments using both instantaneous and continuous sources. The same figure also gives values for K which can be deduced from the statistical theory for long time intervals (1). The figure also shows that the straight line C places a lower limit on all the data, with the only exception of the points marked by  $\blacklozenge$ : in fact such data are inconsistent with our model because they refer to an instantaneous release of constant level balloons at an altitude of 30000 feet, together with a travel time of a few hours. The equation of the line C is a power function, i.e.:

$$\sigma_y \approx 0.2 \cdot t^{1.5} \quad (\text{km}) \quad (2)$$

from which we obtain, taking (1) into consideration:

$$K = 2 \cdot 10^{-2} \cdot t^2 \quad (\text{km}^2/\text{h}) \quad (3)$$

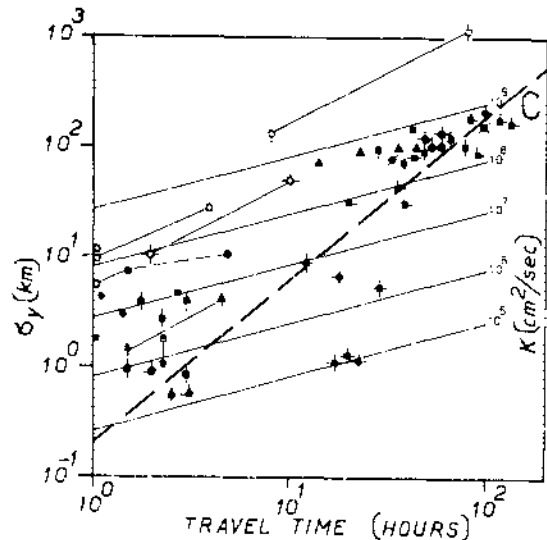


Fig. 1

If we now assume that vertical diffusion is limited by the presence of an inversion at altitude, we may suppose that vertical dispersion is homogeneous within the layer of height  $D$  above ground, where  $D$  is the height of the base of the inversion.\* This assumption appears reasonable provided that distances reach 50-100 kilometers. In this case it is enough to resolve the equation of radially symmetrical diffusion in two dimension for an instantaneous emission: the hypothesis of radial symmetry can be accepted as a good approximation since the diffusion of the cloud along the direction of travel generally differs little from the transverse diffusion.

The basic equation is therefore as follows:

$$\frac{\partial C}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( K r \frac{\partial C}{\partial r} \right) \quad (4)$$

If we assume for the diffusion coefficient  $K$  a general expression of the type  $K = a \cdot t^n$  and supposing an instantaneous, homogeneous, vertically linear emission ( $M$ : mass of pollutant per unit of height:  $Q/D$ ) the solution of this equation is given by:

$$C(r, t) = \frac{M e^{-\frac{r^2}{(\beta t)^{n+1}}}}{\pi (\beta t)^{n+1}} \quad (5)$$

where  $\beta = \left( \frac{4a}{n+1} \right)^{1/(n+1)}$ ; in our case putting  $a = 2 \cdot 10^{-2}$  and  $n = 2$  we obtain

$$\beta \approx 0,3 \text{ (km}^{2/3}/\text{h)}.$$

In health protection calculations it is useful to know the "cloud dosage". This value can easily be determined by means of formula (5), provided that a given value  $\bar{u}$  is fixed for the mean wind speed at the time at which the cloud passes over the site for which it is required to know the cloud dosage. Assuming the time of passage of the cloud over the site to be negligible in comparison with the time elapsed since emission, it can be assumed that  $t$  in (5) is constant; the cloud dosage will be given by the following expression:

$$C.D. = 2 \int_0^{\infty} C(r, t) \frac{dr}{u} = \frac{Q}{D \bar{u} \sqrt{\beta^3 t^3 \pi}} \quad (Ci \cdot h \cdot km^{-3}) \quad (6)$$

It will be apparent that the concentration is expressed as a function of time of travel  $t$ . As already stated, it is only necessary to bear in mind that the proposed model starts to become valid for travel times exceeding the period of time needed for the cloud to disperse into the vertical layer of thickness  $D$ . With excessively long times, however, diffusion on a synoptic scale becomes predominant. It is therefore reasonable to state that the proposed model is valid for an interval between a ten and one hundred hours, when low wind speeds are involved.

#### Some practical considerations for the application of the proposed model

As already noted, speed  $\bar{u}$  (km/h) in formula (6) refers to the wind near ground level, whereas time of travel  $t$  is calculated on the basis of the mean speed within the layer of height  $D$ . In general it can be said that the mean wind

\* From the analysis of vertical synoptic soundings it can be found that a minimum value for  $D$ , related to a frequency of the corresponding meteorological situations statically significant, is about 1 km.



speed value within 20-30 metres above ground is proportional to the mean wind speed within the boundary layer (within D). If we assume as a working hypothesis that the proportional relationship between the two mean values is 1/2, formula (6) can be expressed as a function of scalar path x:

$$C.D. = \frac{2Q\sqrt{\bar{U}}}{D\sqrt{\pi\beta^3x^3}} \quad (C_i \cdot h \cdot km^{-3}) \quad (7)$$

in which  $\bar{U} = 2\bar{u}$  : the mean speed within layer D.

It should also be noted that, having selected the straight line C in the graph in Fig. 1, the situation at one hour after emission corresponds to stability category F with  $u = 2\text{m/sec} = 7\text{ km/h}$ . After a maximum of 10 to 15 hours in this situation there is a change to neutral or unstable conditions, if we suppose the same order of magnitude for the wind speed. Therefore, making  $\bar{U} = 4\text{m/sec}$  in formula (7), the cloud dosage will be expressed by (with  $D = 1\text{ km}$ ):

$$C.D. \approx \frac{26.3}{x^{1.5}} \cdot Q (C_i \cdot h \cdot km^{-3}) \quad (8)$$

The graph in Fig. 2 gives two curves: curve A represents the pattern of cloud dosage according to Pasquill for a 1 Ci emission at ground level under category F conditions with a mean wind speed near ground level of 2 m/sec; above 10 km and up to 100 km this curve is drawn dashed, since in this interval the validity of Pasquill's model diminishes progressively. Curve B represents the pattern of cloud dosage according to the proposed model for an instantaneous release when the initial situation is characterized by category F with a mean wind speed near ground level of 2 m/sec; for a scalar path below 100 km the curve is dashed since, for the reasons given in the preceding paragraph, the proposed model, for such a low wind speed, is only applicable for times of travel greater than a ten of hours. Making allowance for the fact that, at low wind speeds, the vectorial path is shorter than the scalar path, the curve B will in fact be lower if the vectorial path is taken as variable x. Therefore, until a statistical study is made to evaluate the relationship between scalar and vectorial path under conditions characterized by mean wind speeds near ground level of 2 m/sec, curve B gives an overestimate of the cloud dosage values at mesoscale diffusion level if x is taken as the distance of travel

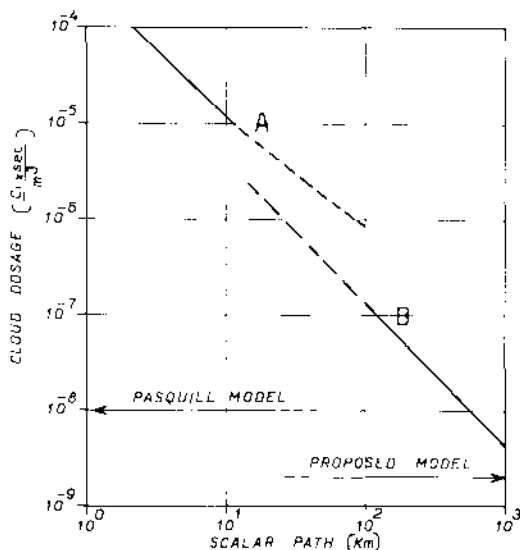


Fig. 2

### Conclusion

In conclusion, taking as a working hypothesis that the vectorial path of the wind is equal to the scalar path, even for wind speeds at ground level of 2 m/sec, curve B of the graph in Fig. 2 can be used to provide a conservative

estimate of concentrations at distance between some tens to some hundreds of km from the point of release.

The values deduced agree very closely with those proposed by Doury<sup>7</sup>. If one makes a comparison with the measurements taken at the time of the Windscale accident<sup>8</sup> in which the emission was prolonged (approximately 2 days) and not constant, one finds that the proposed curve gives values over distances between 100 and 500 km which are higher by factors ranging from 10 to 100 - this can partially be explained by the fact that the emission was prolonged and the wind field changed drastically.

It must finally be mentioned that, in addition to the pessimistic assumption that the distance reached is equal to the wind trajectory, other conservative hypothesis have been adopted, such as the absence of ground deposition during the travel-time of the cloud, the adoption of line C in Fig.1 for the relationship between  $\bar{C}_y$  and travel-time and, in calculating the cloud dosage, the hypothesis that time t remains constant during the passage over point x in question.

#### References

1. Meteorology and atomic energy, 1968, D.H. Slade Edition TID-24190
2. F.PASQUILL: "Atmospheric Diffusion", 1962, D.Van Nostrand Company Ltd., London
3. J.R.MAHONEY: "Model for the Prediction of Air Pollution", 1970, OCDE, Paris
4. "Atmospheric Diffusion and Air Pollution", 1959, Proc.of a Symposium held at Oxford, August 24-29, 1958; Academic Press, New York, London
5. J.L. HEFFTER: "The Variation of Horizontal Diffusion Parameters with Time for Travel Periods of One Hour or longer" 1965, J.Appl. Meteorol. 4 (1), 153-156
7. A.DOURY: "Une méthode de Calcul pratique et générale pour la Prévision numérique des Pollutions Véhiculées par l'Atmosphère" Rapport CEA-R-4280,1872
8. N.G.STEWART and R.N.CROOKS: "Long-Range Travel of the Radioactive Cloud from the Accident of Windscale" Nature 182, 627-628, 1958

ESTIMATION OF GASEOUS RADIOACTIVITY RELEASE  
RATES FROM AN OPERATING BOILING WATER REACTOR

Warren E. Keene, William M. Carey, Lewis Battist  
American Environmental Control Systems, Inc.  
Gaithersburg, Maryland

Abstract

Noble gas activity in BWR stack effluent is commonly estimated by applying a graphic curve-fitting technique to activity measurements on grab samples taken periodically at the steam jet air ejector. It is shown that the graphic solution may conceal significant anomalies in these data, which may be more readily discerned by changing the form of the plot. A better fit to the data is obtained by the method of least squares.

Introduction

Significant quantities of radioactive isotopes of the noble gases, krypton and xenon, are produced in fission. MEEK and RIDER<sup>1</sup> have recently published a tabulation of fission yields for several fissile materials and most of the known fission products. A portion of these products find their way into the primary reactor coolant; the remainder are trapped in the fuel elements. In a boiling water reactor (BWR) most of the gaseous fission products in the primary coolant are removed at the steam jet air ejector (SJAE). After a holdup period to permit decay of much of the short-lived activity and after filtration to remove particulate daughter products, the remaining radioactive gases are vented to the atmosphere via the stack.

Estimation of the radiation dose to the population in the vicinity of a BWR depends on a knowledge of the identity and concentration of the radionuclides present in the stack plume. Twenty-seven radioactive isotopes of krypton and xenon, together with their radioactive decay products are found in the plume. Only about thirteen, plus their decay products, are normally present in sufficient quantity to make them of more than academic interest. As it is impractical to measure the concentration of all important constituents, a combination of measured and calculated values is used to estimate stack activity release rates. Significant overestimation of activity release rates unnecessarily penalizes the public in the form of added costs of power production; underestimation results in a population dose greater than anticipated. For the protection of the public, reactor operating licenses place limits on permissible activity release rates.

### Data Acquisition

To demonstrate compliance with radioactivity release rate limits, nuclear power plants are equipped with a continuously operated stack monitoring system - usually consisting of a sodium iodide, NaI(Tl), detector together with associated electronic and recording equipment. It is not used in a spectroscopic mode, but measures gross activity release rates at the stack. In addition periodic grab samples are taken from the SJAE effluent and analyzed to quantify the isotopic activity release rates. The analysis commonly makes use of an additional NaI(Tl) detector or Ge(Li) detector and multichannel analyzer in the plant laboratory. The resulting spectrum is analyzed to estimate SJAE activity release rates for three krypton and three xenon isotopes. Elaborate spectrum stripping techniques are generally beyond the capability of most power plants. It is necessary to plot the data and use a curve fitting technique to interpolate and extrapolate the data to estimate the activity release rates for up to twenty-one additional noble gas isotopes.

### Present Data Handling Method

On theoretical grounds and experimental evidence BRUTSCHY<sup>2</sup> has postulated that the isotopic activity release rates ( $A_i$ ) for each of the noble gas isotopes can be fitted by a second order equation in the one-half power of the radioactive decay constant ( $\lambda_i$ ):

$$A_i/y_i = b_0 + b_1\lambda_i^{1/2} + b_2\lambda_i \quad (1)$$

where  $y_i$  is the fission yield (atoms/fission) of the  $i$ th isotope and the  $b_j$  are constants to be determined. An alternative form for eq. (1) is obtained on dividing by  $\lambda_i$ :

$$A_i/y_i\lambda_i = b_2 + b_1\lambda_i^{-1/2} + b_0\lambda_i^{-1} \quad (2)$$

It is further postulated that each of the terms in eq. (2) represents activity released from the fuel via a different escape mechanism. Then eq. (2) may be separated into three component equations:

$$(A/y\lambda)_{I} = b_2 \quad (3)$$

$$(A/y\lambda)_{II} = b_1\lambda^{-1/2} \quad (4)$$

$$(A/y\lambda)_{III} = b_0\lambda^{-1} \quad (5)$$

where the subscripts on the variables have been dropped for convenience. Taking logarithms of both sides of each of the last three equations it is seen that, on a logarithmic plot of  $A/y\lambda$  vs.  $\lambda$ , eq. (3) results in a straight line of slope zero; eq. (4), a straight line of slope  $-1/2$ , and eq. (5), a straight line of slope  $-1$ . The constants  $b_1$  and  $b_2$  are obtained from the intercepts of these lines with the vertical at  $\lambda = 1$ . These relationships form the basis of a simple graphic technique for estimating the individual and total activity release rates for the 27 krypton and xenon isotopes.

It is further postulated that the iodine isotope concentrations in the bulk coolant should fit an equation identical in form to eq. (2), and that the iodine data and the noble gas data share a common value of  $b_2$ , but different values of  $b_0$  and  $b_1$ . In practice, the value of  $b_2$  obtained from the iodine data is used arbitrarily and only  $b_0$  and  $b_1$  are determined from the noble gas data.

The measurements and resulting activity release rate estimates are commonly made on a monthly or weekly schedule and form the basis for calibrating the stack monitor after the SJAE activity release rates are corrected for radioactive decay during holdup prior to release at the stack.

### Recent Data Handling Experience

The authors have recently made field measurements of airborne alpha, beta and gamma activity on a continuous basis over a period of several weeks in the vicinity of an operating BWR. The monitor is sufficiently sensitive to permit on-line measurements of airborne activity at levels down to the natural background. The monitor has been described elsewhere<sup>3</sup> and some of the results of the survey are the subject of a separate paper<sup>4</sup>.

In order to be able to correlate the activity measurements in the field with stack-release data the plant records of the (weekly) measurements of the radioactivity of the SJAE effluent samples were examined, together with the stack monitor record. It was confirmed that the graphic technique had been applied on a consistent basis and that the necessary supporting computations were arithmetically correct. However inspection of the plots of the data and the curves fitted to the data by the graphic technique indicated that the fitted curves did not appear to be good fits to the data. Therefore the data were fitted by the method of least squares, using eq. (2) (the same form of the equation used for the graphic solution). A double-precision program was used on a PDP-10 computer.

With only six data points per plot, one hesitates to ascribe a great deal of significance to the results of various statistical measures of the goodness of fit. However one expects that a consistent improvement (decrease) in the residual sum of squares should be indicative of a better fit to the data. The residual sum of the squares is defined as

$$RSS = \sum_{i=1}^N (y_i - Y_i)^2 \quad (6)$$

where  $N$  is the number of data points,  $y_i$  is the measured value of the dependent variable, and  $Y_i$  is the curve-fit value of the dependent variable. (A least squares solution, by definition minimizes the value of RSS for the form of equation to which the data are fitted.) Twelve sets of weekly data were examined. Not only did the fitting to  $A/y\lambda$  fail to improve the average goodness of fit, but the coefficient ( $b_2$ ) of the second-order term was consistently negative. A negative coefficient is mathematically satisfactory but it conflicts with the theoretical postulate that each term of the equation represents activity

released via a different mechanism (how to postulate a negative escape mechanism?). The coefficient was sufficiently negative that it resulted in predicting negative radioactivity release rates for several of the shorter half-life isotopes - in conflict with both theory and experience.

When the data are replotted, on a linear scale, using eq. (1), the reason for the negative coefficient for the quadratic term becomes clear. Figure (1) is a typical example of the data. The abscissa scale is in units of  $\lambda^{1/2}$ , with  $\lambda$  in  $\text{sec}^{-1}$ . The ordinate scale is arbitrary. Only by brute force can one fit such data to a quadratic equation with a positive second order term. In practice this is accomplished by using the value of  $b_2$  obtained from the iodine measurements (curves labeled 1 & 2).

It should be noted that the noble gas data spans values of  $\lambda^{1/2}$  from 0.001 to 0.030  $\text{sec}^{-1/2}$ . The abscissa scale must be extended to 0.85  $\text{sec}^{-1/2}$  to include all of them. It is asking too much of any curve-fitting technique to provide reliable extrapolation over such a range. However extending the abscissa to 0.060  $\text{sec}^{-1/2}$  will include all isotopes which remain in significant quantity after a fifty minute holdup prior to release from the stack. (From the standpoint of a potential accidental release within the plant after only a brief holdup it would be useful to have a technique which could be used with confidence to estimate SJAЕ activity flow rates for the shorter-lived isotopes.) The critical unmeasured long-lived isotope is Kr-85. Its value of  $\lambda^{1/2}$  is  $0.45 \times 10^{-4} \text{ sec}^{-1/2}$ , essentially zero on our abscissa scale. Because of the shape of the data and the resulting poor fit of both curves #1 and #2, there is probably a factor of two uncertainty in the estimated emission rate of Kr-85 for these data.

A negative second order term which results in estimations of negative activity release rates is unacceptable, a priori. It is also inconsistent with the three-mechanism concept. So the brute force approach may be the best that can be done with data which is inconsistent with the model. We have used the method of least squares to accomplish this type of force-fitting, as well as to examine the behavior of RSS (eq. 6) when the noble gas data are fitted to equations (1) and (2) under varying assumptions. Figure (1) shows the resulting curves.

The lowest value of RSS (best fit in the sense of least squares) was consistently obtained when the data were fitted using  $(A/y)$  (curve #3) as the dependent variable (eq. 1) rather than  $(A/y\lambda)$  (curve #4) (eq. 2). It is not understood why this should be so. (As expected, the second order terms remained negative) The reduction in RSS in going from  $A/y\lambda$  to  $A/y$  was commonly by a factor of five and occasionally more than an order of magnitude.

Since the least squares method, applied directly to the noble gas data, resulted consistently in negative second order terms in both equations (1) and (2), it is clear that arbitrarily changing the value of this coefficient to some positive value must result in worsening the goodness of fit (increasing

RSS). When a least squares fit is performed first on the iodine data (curve #2) to determine the value of  $b_2$  to be used in fitting the noble gas data, eq. (1) is reduced to a first order equation in  $\lambda^{1/2}$  whose remaining constants ( $b_0$  &  $b_1$ ) are also found by the method of least squares. The resulting value of RSS lies between that for fitting to  $A/y$  and that due to the graphic solution. It is consistently closer to  $RSS(A/y)$  when the plotting variable ( $A/y$ ) is multiplied by the fission yield ( $y$ ) to give isotopic activity release rates, the discrepancy between the graphic and the least squares results is on the order of five to fifty percent for the data examined. There appears to be a consistent bias in the graphic results yielding lower release rates at longer half-life and higher release rates at shorter half life than the release rates estimated by the method of least squares (cf. curves #1 & #2). It is unclear whether this observation is due to inherent bias in the graphic technique or to the peculiar shape of the data examined. Neither the graphic nor the least squares method yields any information about the reason for the unexpected shape of the data. The study is continuing and it is expected that there will be an opportunity to examine data from other time periods for the same BWR to determine whether the "misshapen" data were peculiar to the particular time period of the study. Ad interim, it is impossible to know whether either technique results in over- or under-estimating absolute levels of radioactivity release for the period studied.

#### Conclusions

We conclude that the use of the method of least squares on the data for the iodine isotopes in the bulk coolant and on the data for the noble gas isotopes in the steam jet air ejector effluent is preferable to the graphic solution, given the availability of a programmable calculator or computer. We also note that the graphic solution itself can be reduced to a simple routine suitable for a programmable calculator. This should be done to eliminate errors of human plotting judgment in accomplishing the solution, whenever a programmable calculator or computer is available - if it is determined that there is a valid argument for not replacing the graphic method with the method of least squares.

It is suggested that BWR operators more closely examine the data they are acquiring by spectroscopic analysis of bulk coolant iodine levels and SJAE off-gas noble gas levels. The discrepancies discovered in our data are not readily apparent from the usual log-log plots of  $A/y\lambda$  vs.  $\lambda$  and an uninquisitive mechanical application of the graphic method; they are apparent by inspection of a linear plot of  $A/y$  vs.  $\lambda^{1/2}$ .

#### Bibliography

1. Meek, M.E. and Rider, B.F., "Compilation of Fission Product Yields", Vallecitos Nuclear Center-1972, NEDO-12154, Jan. 72.
2. Brutschy, F.J., "A Comparison of Fission Product Release Studies in Loops and the VBWR", AECL-1265, Jun. 61.

3. Battist, L., U.S. Patent Application No. 80572, Oct. 14, 70.
4. Carey, W.M., Battist, L. and Keene, W.E., "Measurements of the Dispersion of Gaseous Radioactive Effluents from an Operating BWR". Paper No. V-0004-R-5, this conference.

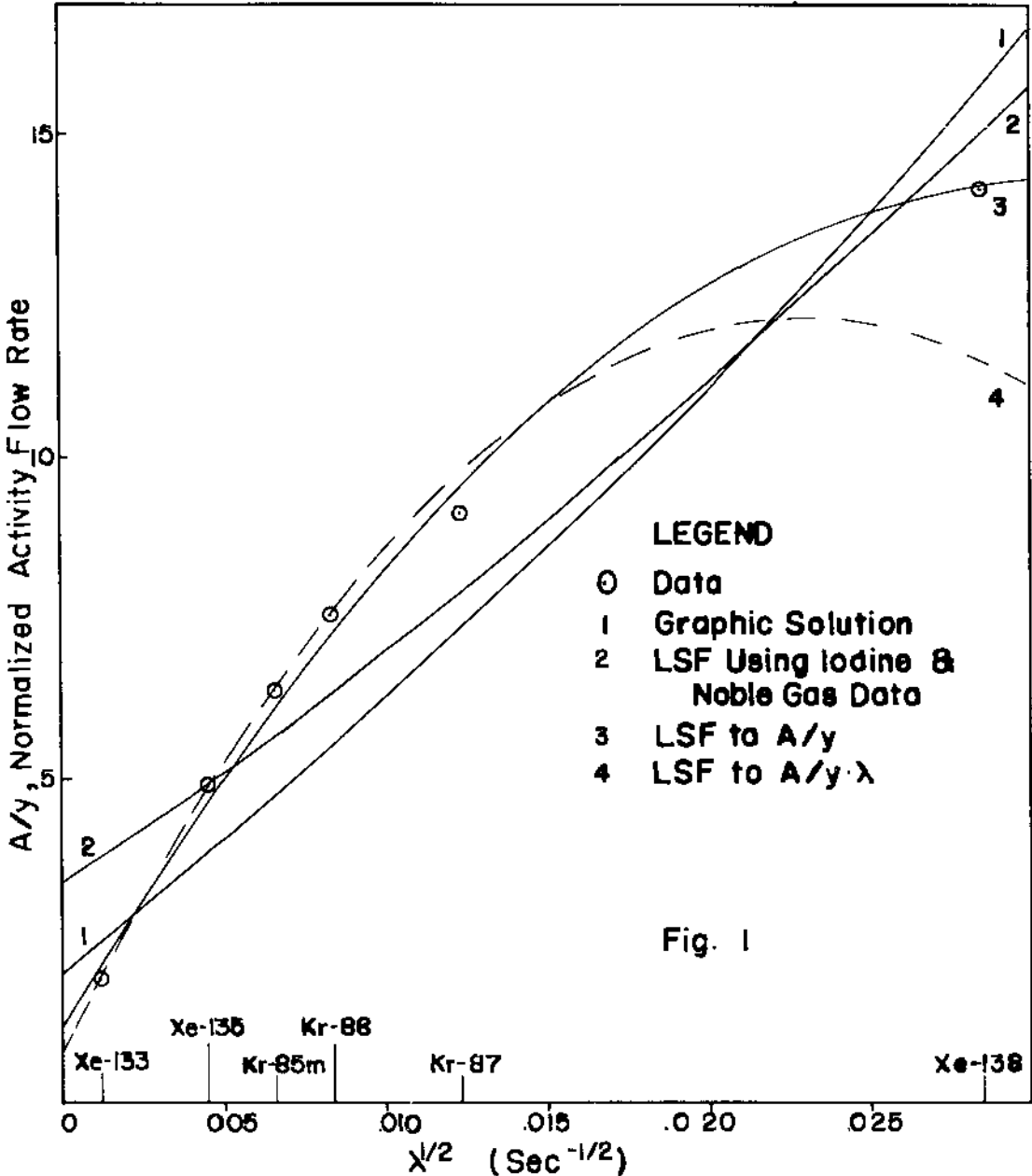


Fig. 1

Noble Gas Activity Flow Rates at the Steam Jet Air Ejector of an Operating BWR  
Results of Selected Curve Fitting Attempts



## SITING EVALUATION OF A NUCLEAR POWER PLANT IN SOUTHERN ISRAEL.\*

A. Donagi, Y. Feige, Y. Mamane and Y. Argaman

Environmental Engineering Laboratory  
Technion Israel Institute of Technology  
Haifa, Israel.

### Abstract

An investigation of the feasibility of building a nuclear power plant before 1980 along the southern Israel seashore was carried out. The various engineering, economic, ecological and public aspects of the problem were analyzed. The feasibility of a nuclear power plant depends upon economic and environmental factors as well as the need for diversification of the power production sources. Israeli power production capacity at present is relatively small. However, with the expected increase in total generating capacity and constantly rising oil prices, a nuclear power plant of about 400-500 MW capacity (or a somewhat larger one in the event of a combination of power production and desalination) could be included in the conventional network. Other factors that may influence the decision are as follows: scarcity of potential sites along the seashore, the level of various pollutants in the air, and the sensitivity of the Israeli public toward the installation of conventional and nuclear power plants. The relative importance of the various factors was weighed and analyzed. Local ecology experts are all in agreement that a nuclear power plant is environmentally preferable. It would be regrettable to miss the "break-even point" which, because of the specific Israeli conditions, may have already been reached.

### Introduction

Israeli interest in nuclear power plants goes back to the early sixties. In 1962, a special committee made the following recommendations: to reserve several sites along the Mediterranean coast for construction of nuclear power plants (n.p.p.); to study the feasibility of installing at the end of the sixties, the first n.p.p.; and to maintain a continuous follow-up of technical developments in this field.<sup>1</sup>

Due to the relatively small size of the Israeli power supply system, the largest power unit operated until now by the Israeli Electrical Corporation (IEC) is 230 MW(e). Under these circumstances, n.p.p.'s could not compete economically with conventional power plants (c.p.p.). Recently, the question of determining when to install the first n.p.p. has been discussed quite frequently at symposia and technical meetings<sup>2,3</sup>. Various bodies have become interested in the subject and participated in those meetings.

The municipality of Ashdod, one of the communities near two potential sites for the first n.p.p., commissioned the Technion, Israel Institute of Technology, to investigate the ecological and environmental aspects of the

\* Partly supported by a research grant from the city of Ashdod, Israel.

installation of such a plant in the vicinity of the city.<sup>4</sup> It should be noted that, as a result of the "ecological scare" that has recently swept Israel, local authorities often insist upon carrying out their own investigations. The present work is a result of such a study.

#### Capacity of Existing Conventional Power Plants

Electric power is produced, transmitted and distributed to Israeli customers by a single company, the IEC. Its power generating capacity grew from 3.0 MW(e) in 1930 to 570 MW(e) in 1962 and 1525 MW(e) in 1972. This rapid development compels the IEC to cope with a yearly growth rate that is one of the highest in the world. Only 6 years are required to double power production in Israel. Out of 42 countries listed in a recent survey<sup>5</sup>, Israel has the highest yearly load factor (0.68 versus 0.30-0.50 in most other countries).

Israel's power generation in 1974 will total 1920 MW(e). The power plants are located in Ashdod - 780 MW(e), Tel Aviv - 620 MW(e) and Haifa - 520 MW(e). In addition, a few gas turbines having a capacity of up to 40 MW(e) each are being installed, mainly to meet peak power demands.

#### Factors Influencing the Decision Making

The criteria for evaluating the feasibility of the n.p.p. are discussed below. The most important factors are economic and environmental criteria and the stage of technological development.

Economic Factors. Economic considerations have played a major role in preventing the government from reaching a positive decision regarding a n.p.p. The initial investment in the n.p.p. is very high compared to the c.p.p., while operating expenses are relatively low. The major factors determining the economic feasibility of a n.p.p. are the cost of conventional fuel and the initial investment, which depends on the size of the production unit and the financing conditions<sup>6</sup>.

In the last few years, the cost of conventional fuel (bunker-oil in Israel) has been rising steeply. Any forecast about the economics of a n.p.p. has to be based on various shifting factors, mostly related to the cost of conventional fuel. Nuclear fuel is considerably cheaper than fossil fuel, and its compact shape contributes to the ease of its handling and transportation, as compared with fossil fuel<sup>7</sup>.

The major obstacle to the development of a n.p.p. in Israel has been the relatively small size of the Israeli power network. However, a study by Ashner<sup>8</sup> indicates that a n.p.p. of 600 MW(e) or more may be more economical than a conventional unit, when the following conditions exist: the cost of crude oil is US\$16/tonne (US\$0.393/Btu), the load factor is 80%, and the interest rate is 10%. (Fig.1).

Surveys and forecasts, done by the IAEA, show that from both economic and environmental viewpoints a n.p.p. of 500 MW(e) or more constitutes the best and cheapest form of power generation. The share of n.p.p.'s in the world power generation is expected to grow from 8% in 1975 to 27% in 1985, and 63% in the year 2000<sup>7,9</sup>.

Public Attitude. Several years ago, in spite of widespread objection the Israeli government decided to install a 428 MW(e) c.p.p. within the city of Tel Aviv. The government was severely attacked by public organizations and a continuous dispute about the merits versus the shortcomings of the project has captured the headlines of the daily press for a long time. The decision created an unpleasant antagonism among the people and, at present, the attitude of the general public is one of suspicion towards the c.p.p. (A similar conflict arose in 1971 when plans for installing a new c.p.p. in a relatively

unpopulated area, north of Tel Aviv, were made public).

The Israeli public does not have, as yet, an antagonistic attitude towards a n.p.p. This is in contrast with other countries such as the U.S.A., where public opposition occasionally causes delays in the construction of n.p.p.'s<sup>10</sup> Israel already has two research reactors, which have been functioning without problems since the early sixties. These reactors have boosted the development of nuclear research and created a sound basis for the rapid absorption of the first n.p.p. into the Israeli power supply network.

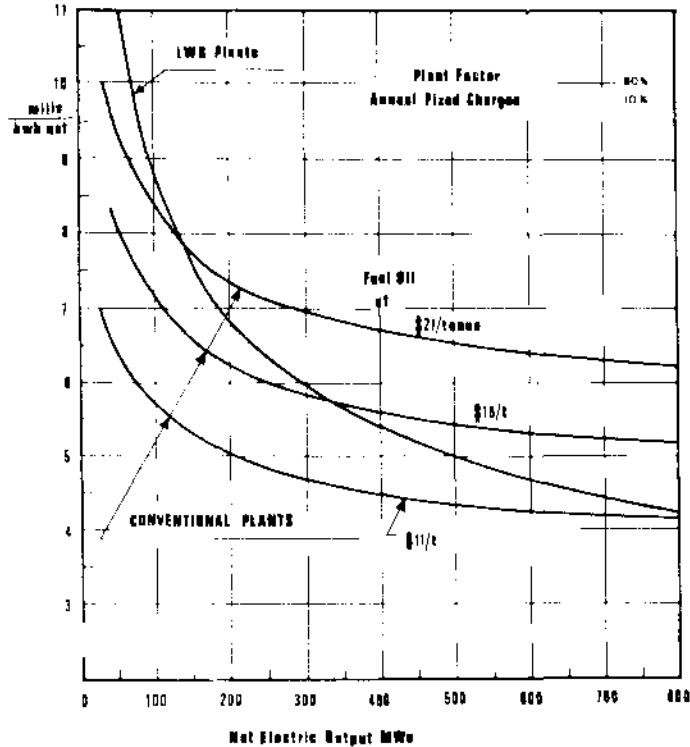


Fig.1 - Comparison of cost per kwh net in conventional and LWR power plants (after Ashner<sup>8</sup>).

Environmental Comparison of Nuclear and Conventional Power Plants.

The most important ways in which a n.p.p. may affect the environment are the following: a) releasing minute quantities of radioactive materials into the air and water, (b) releasing large quantities of heat into the cooling water, and (c) making necessary a reevaluation of plans for development and uses of the land in the vicinity of the reactor. The environmental effects of nuclear and conventional power plants are compared in Table 1.

It is not easy to make an environmental comparison of a c.p.p. and a n.p.p. While the information regarding the environmental hazards of a n.p.p. is ample, much less data exist regarding the c.p.p. It should also be noted that it is hardly possible to compare a population exposure to a few mrem/year with an exposure to a yearly average SO<sub>2</sub> concentration of 0.03 ppm. However, a few investigators<sup>11-13</sup> have tried to compare the effect of radioactive pollutants to that of conventional air pollutants, and reached the conclusion that, from the environmental standpoint, the n.p.p. is preferable.

The importance of environmental criteria is steadily increasing, as a result of the growing public awareness of the problems. There are indications that future environmental standards for c.p.p.'s will be stricter, resulting in greater expenditures for control devices. The n.p.p. has already reached a

much higher level of environmental control, which clearly gives the n.p.p. a certain advantage.

Both n.p.p. and c.p.p. affect the land-use and development of the area. However, because of the present trend to minimize land-use restrictions around nuclear reactors, it seems that the effect of a n.p.p. on its close environment becomes marginal.

Table 1. Environmental Effects of Nuclear and Conventional Power Plants

Type of pollution	Conventional Power Plant	Nuclear Power Plant
Sea and Ocean	Oil spills from storage facilities pollute the marine environment. Floating oil alters the heat and oxygen exchange balance between sea and air.	Transportation of nuclear fuel does not result in any release of pollutants to the sea. Minor quantities of radioactivity are released to the sea during the normal operation of the reactor. These have negligible environmental effects.
Soil and land pollution	Pipelines, storage facilities and the plant itself affect landscape. Accidental spills may result in soil and water pollution.	No effect due to fuel transportation or storage. No release of radioactivity to land. Effects on landscape are less than in c.p.p.
Thermal pollution	Large amounts of heat released to the marine environment may adversely affect marine ecology. On the Mediterranean coast a properly designed outfall can reduce these effects to a negligible level.	
Air pollution	Air pollutants such as SO <sub>2</sub> , NO <sub>x</sub> , CO <sub>2</sub> and particulates in large quantities are released from a c.p.p. Radioactive pollutants are also released from a c.p.p. as a result of burning coal or liquid fuel. Solutions to air pollution problems are expensive. They include tall stacks, scrubbing and filtration of stack gases, and pretreatment of fuel.	Emission of radioactivity is low and may be stored until favorable meteorological conditions prevail. The probability of major accidents is extremely low, thus almost no air pollution problems arise from a n.p.p.

Need for Diversification of Power Sources. Israel has neither solid-fuel resources nor hydroelectric options, and its power generation is completely dependent upon a single energy source - liquid fossil fuel - which must be imported and transported, mostly by tankers. By installing nuclear power plants, which use only a few tons of fissionable material that has to be partially replaced about once a year, the country could attain a certain independence. This is of importance when considering the present political conditions in the area.

Need for Desalination Plants. Israel has reached the point where it exploits practically all of its conventional water resources. Water desalination is already practiced in the country on a small scale and installation of a joint n.p.p. and water desalination plant has been investigated. However, due to the high cost of desalted water and the lack of experience, not much progress has been made in this direction.

### Future of Nuclear Power in Israel

Israel's power supply system is too small to have justified, in the past, the inclusion of a n.p.p. of a practicable size of 400-500 MW(e). Usually a single power supply unit should not exceed 10-15% of the network's total capacity. However, based on the projected demand illustrated in Fig.2 it is obvious that the operation of such a n.p.p. in the early 80's will become practicable.

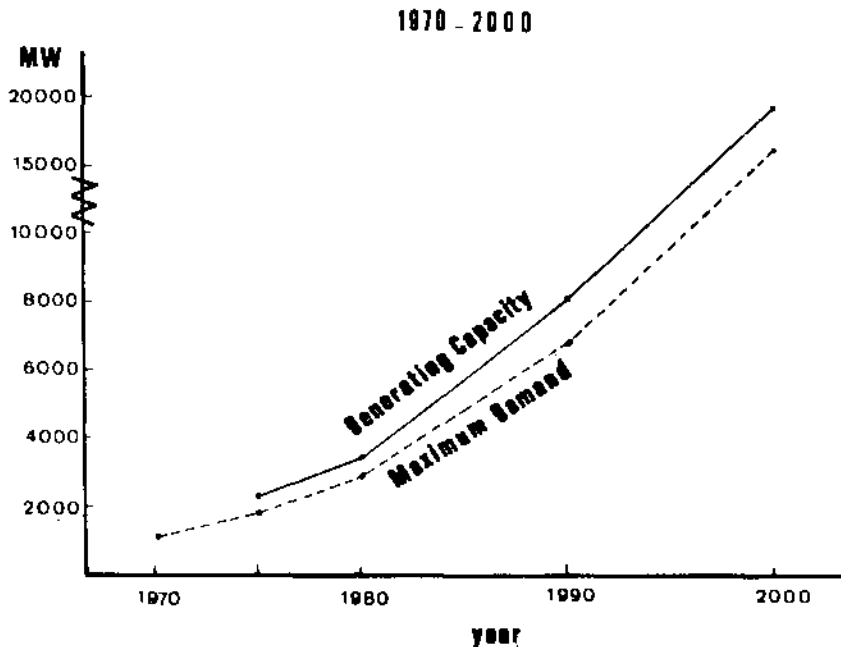


Fig.2 - Projected demand for electric power in Israel

Only reactors of proven technology will be considered as suitable for Israel in the foreseeable future.

The basic criteria for siting the n.p.p. are similar to those considered in locating a c.p.p. (e.g., proximity to cooling water, safety from floods and earthquakes, strength of the subsoil where the foundations of the station are to be laid, proximity to power consumers, existence of roads and availability of manpower to be engaged in the construction). In addition, criteria of radiation safety are also taken into consideration. The experience obtained during the last years, the improved design of the reactors and the various control measures, now permit the installation of a nuclear reactor even in the vicinity of dense population centers. A nuclear reactor should be safe everywhere its safety depending primarily upon safeguards which are an inherent part of its engineering.

As power stations should be located along the seashore, and because the population there is already very dense, only four sites could be reserved in Israel for n.p.p.'s until the end of the 20th century. This siting took into consideration forecasts of population increases in the big cities. Around each of these sites there are two regions - one which could be relatively restricted to civilian population (because it is uninhabited at present), and one (farther from the reactor) which is recommended to remain a low population density region. It is noteworthy that in some countries (Canada, Czechoslovakia and Japan) the buffer zones where construction is restricted does not exceed 500-1000 m.

### Summary and Conclusions

Many developing countries, most of them not more developed technologically than Israel, have already ordered n.p.p.'s. It is especially difficult for a developing country to justify installation of its first n.p.p. Its inclusion within a conventional power generating system poses special difficulties. The various approaches to these questions are presented in Appendix A, where a comparison between Israel and Greece (a developing country, also) is given.

This investigation, based on a study carried out for the city of Ashdod<sup>5</sup>, showed that from ecological and environmental aspects there is a distinct advantage in installing a n.p.p. rather than a c.p.p. Even from the economic viewpoint, a decision should be made now so that the n.p.p. could be operational by 1980.

According to some approaches, a delay may be desirable in the long run because it may save some money. Other investigators believe that this delay is already economically unjustified. These debates indicate that Israel will probably make a positive decision in the near future, in accordance with the general attitude of most of the Israeli workers who have investigated these questions.

### References

- 1) "Nuclear Power Plants in Israel", Israel Atomic Energy Commission, May 1962 (in Hebrew).
- 2) Proc. of a symposium on "Nuclear Power Plants in Israel", Tel Aviv, Oct. 1972 (in Hebrew).
- 3) Proc. of the Regional Conference on Radiation Protection, Jerusalem, March 1973 (Sponsored by I.R.P.A.).
- 4) Y. Argaman et al. "Examination of the Location of Power Stations in Southern Israel and its Effect on the City of Ashdod", The Technion Research and Development Foundation, Haifa, May 1973 (in Hebrew).
- 5) United Nations Statistical Yearbook, 1970.
- 6) S. Yiftah, "Nuclear Energy in Israel", Proc. of the Regional Conference on Radiation Protection, Jerusalem, March 1973.
- 7) B. I. Spinrad, "The Role of Nuclear Power in Meeting World Energy Needs" IAEA Conf. on Environmental Aspects of Nuclear Power Stations, p. 57, IAEA, Vienna, 1971.
- 8) P. S. Ashner, "Nuclear Energy in Israel", a lecture given at Technion, Haifa, 1972 (in Hebrew).
- 9) D.S. Briggs, M. A. Khan, P. H. Kruck and M. Ristic, "Small and Medium Power Reactors; Present Status and Prospects", Atomic Energy Review, IAEA 7, No.3, p.119, 1969 .
- 10) "Public Attitudes about Nuclear Power", INFO, Atomic Industrial Forum, Dec.1972.
- 11) C. M. Eisenbud and H. Petrov, "Radioactivity in the Atmospheric Effluents of Power Plants that Use Fossil Fuel", Science 144, 288, 1964.
- 12) J. G. Terril et al. "Environmental Aspects of Nuclear and Conventional Power Plants", Ind.Med. Surg. 36, 412, 1967.
- 13) A. P. Hull, "Some Compariaons of the Environmental Risks from Nuclear and Fossil-Fueled Power Planta", Nuclear Safety 12, 185, 1971.

## APPENDIX A

In this appendix an attempt is made to compare the various approaches to the question of the integration of a nuclear power plant into the power supply system of a developing country. For this purpose Greece, whose power problems are similar to those of Israel, has been selected for comparison with Israel. This comparison is based upon data reported by officials of the Greece Public Power Corporation (Skelakalos and Karangelos\*) and is presented in the following chart:

### A Comparison of Approaches towards the Question of Installation of a N.P.P. in Two Developing Countries

<u>Problem</u>	<u>Israel</u>	<u>Greece</u>
Type of body supplying the electricity	The Israel Electric Corp. Ltd. (I.E.C.) is a public utility concerned with the generation and supply of electricity.	Public Power Corporation (PPC) is responsible for the generation, transmission and distribution, transmission and distribution of electricity.
The type of energy	Mostly liquid-fuel imported into Israel; local deposits of oil and gas supply only 2-3% of yearly demand; uranium ore deposits have been found mixed with phosphate rock.	Deposits of solid fuel; indications of oil and natural gas deposits; hydroelectrical potential of 15,000 GWh/year; uranium ore deposits under exploration.
Proposed date of installation and number of units to be integrated in the national supply network	4-5 n.p.p. of 500-600 MW(e) each to be installed between 1980 and 1990 plus 4-5 fast power reactors of 800-1000 MW(e) each, to be installed between 1990 and 2000.	8 n.p.p. of about 600 MW(e) each of the LWR type will be installed between 1982-1991; 8 n.p.p. of 1000 MW(e) each will be installed between 1993-2000.
Determination of the size of the first nuclear power unit.	The appropriate size will be determined by considering two opposite trends: a) Large units are cheaper (per kWh) b) Operational requirements of the system give preference to small units.	
Type of reactor	The first reactors will probably be of the LWR type, due to the proven good experience obtained with these reactors.	
Siting of the n.p.p.	The fact that both Israel and Greece are Mediterranean countries having a long sea coast practically solves the question of thermal pollution. However, development of seaside tourism dictates the need for early n.p.p. siting. The two countries have already selected potential sites for the n.p.p.'s that will be built by the year 2000.	
Feasibility of a joint n.p.p. and water desalination plant	Because of water shortages, serious consideration is given to the possibility of installing a joint project for nuclear power production and water desalination. This integration will become a necessity in the eighties.	The idea of an integrated n.p.p. and a water desalination plant is being investigated by Greek scientists.

A Comparison of Approaches towards the Question of  
Installation of a n.p.p. in two Developing Countries  
(continued)

---

<u>Problem</u>	<u>Israel</u>	<u>Greece</u>
Forecast	Nuclear energy is probably the best answer to the rapid growth of electricity demand. This solution relieves Israel from complete dependence on one type of fuel (crude or bunker-oil), enables acquisition of experience and know-how, and partly solves the problem of conventional air pollution.	In Greece "Nuclear energy is considered as the unique solution for meeting the future energy needs, not only because the country's conventional energy resources are being depleted, but also because it is believed that they should be saved for future generations for other uses".

Both countries have power-generating systems of a relatively limited size. As yet, there is no decisive economic advantage in including a n.p.p. in the conventional power network. However, both countries are rapidly approaching the stage where such an inclusion will be economically justified. Greece, in spite of the fact that its need for diversification of its power resources is less urgent than that of Israel, has already taken a positive attitude, by deciding in principle upon the installation of the first nuclear power plants. Israel, on the other hand, has not yet made a decision. As both countries are on the verge of economic justification of such a venture the outcome may fall either way. The difference in the approach of the two countries may stem from a certain difference in the local conditions and on other factors besides economic ones which are involved.

---

\* K. Skelakalos and J. Karangelos: "Integration of Nuclear Power into the Greek System", in the Proc. of the Regional Conference on Radiation Protection, March 5-8, Jerusalem, Israel.



A REVIEW OF THE FIRST SEVEN YEARS OF OPERATIONAL  
HEALTH PHYSICS AT DUNGENESS 'A' (MAGNOX) NUCLEAR POWER STATION

E.P. Goldfinch, Station Health Physicist, Dungeness A & B  
Nuclear Power Station, Central Electricity Generating Board,  
Lydd, Kent, England.

Introduction

Dungeness 'A' Nuclear Power Station comprises two identical reactors each of the magnox type of nominal thermal power 725 MW(th), nominal electric power 205 MW(e). The first reactor achieved commercial full power in October 1965 and the second in December 1965. The reactors have been in operation with a lifetime load factor of 77.1% up to March 31st, 1973, and total electrical production  $24.5 \times 10^9$  KWH (exported). Refuelling on magnox reactors is continuous with the reactor at full power. This paper reviews the Health Physics aspects of operation over the past seven years.

Early Commissioning Radiation Surveys

On completion of reactor construction, commissioning was undertaken in three stages. Firstly general plant commissioning tests and performance studies, secondly fuel loading and reactivity experiments, and thirdly stage by stage power raising including reactivity experiments and establishment of dose rate contours, shielding integrity studies and dose rate measurements for interpolation to higher powers in areas where the dose rate would be too high for actual measurements.

The whole site was divided into 76 areas and within each area designated survey positions were marked both in situ and on drawings. All survey positions were identified by unique code number related to the reactor number, area type, area identity and survey position number. These enabled rapid comparisons to be made between results in similar geometrical positions. In all some 1500 positions were nominated.

Surveys were undertaken at nominally 2%, 20% and 100% full power, including gamma radiation, thermal neutron and fast neutron measurements. Because of limited resources, it was not expected that every planned measurement could necessarily be made at each power level. Priorities were allocated to ensure that adequate information was gained, especially for extrapolation to higher reactor powers.

The surveys showed that

- i) In general gamma radiation levels were lower than predicted.
- ii) Neutron levels were higher than predicted especially in boiler cells and on the reactor roof.
- iii) The integrity of shielding construction was confirmed.
- iv) Dose rates on shutdown boilers were higher than expected, due to a higher than expected contribution through the reactor primary shield.

Studies on fuelling machines over the first few months operation were carried out and showed that

- i) The general shielding design was satisfactory both for neutron and gamma radiation.
- ii) Access to the charge face need not be restricted during fuel discharge, provided that only the shielding slab immediately beneath the refuelling machine is removed. However, access to the interspace between the primary biological shield and the charge face, must be strictly prevented.

Site Environmental Neutron Dose Rates

Neutron dose rates were found to be higher than expected both in boiler cell areas and in some working areas. Additional shielding was installed and this reduced dose rates to acceptable levels. However, in some working areas the neutron dose rate is still approximately equal to the gamma dose rate even though both are low and in some infrequently visited access areas neutron dose rates and gamma dose rates are of the order of 1 rem/hr. It thus became necessary at a very early stage in commissioning to consider the possibility of some form of personal neutron dosimetry. Crude neutron spectral measurements were made in the hope that there may be a fixed ratio between total neutron dose rate and thermal neutron dose rate. It would then be possible to infer total neutron dose to personnel from measured thermal neutron dose on personnel film badges. The results of the experiments undertaken in 1966 showed that the ratio total neutron dose to thermal neutron dose varied between 3.5:1 and 10.5:1. A weighted value of 5:1 was accepted as an interim value taking into account the likely occupancy factors. Thus all personnel dose records included a neutron contribution equal to five times the measured

thermal neutron dose.

The minimum detectable dose on a film badge became of great importance, both for gamma dose and thermal neutron dose. The film badge used at Dungeness is that described by Heard and Jones<sup>2</sup>. Assessment of neutron dose is by comparison of film density under a cadmium filter against density under a tin filter of equal mass thickness. Statistical variation in density between areas of the same piece of film can lead to an apparent thermal neutron dose. Series of film badges were therefore exposed to low nominal thermal neutron doses of 1 m rem, 2 m rem, 3 m rem, and 4 m rem, concurrently with nominally 0 m rem, 5 m rem, 11 m rem and 80 m rem of gamma radiation. Standard deviations of the results were determined yielding minimum detectable levels (85% confidence) of 20 i.e. 1.3 m rems for thermal neutrons and 3.5 m rems for gamma doses. These values were rounded to 1.5 m rem and 5 m rems. It is not surprising that a lower value is obtained from thermal neutrons than gamma radiation because variations in density are between areas of the same piece of film as opposed to variations between separate films. The minimum detectable level of total neutrons was taken as 5 X that for thermal neutrons i.e. 8 m rems (rounded). Sophisticated neutron spectrum measurements made more recently at Dungeness by Harvey<sup>4</sup> have shown that a revised neutron factor of 4:1 for film badges should be used.

The Berkeley Nuclear Laboratories of the Central Electricity Generating Board were asked to develop a suitable personnel neutron dosimeter. This has recently been described in a paper by Harvey, Hudd and Townsend<sup>3</sup> at a Symposium organised by the IAEA in Vienna, December 1972. This dosimeter (The Albedo Dosimeter) is now used on occasions when a known neutron exposure is to be incurred but a monthly assessment of neutron dose to all station personnel is assessed from the film badges.

#### Off Site Environmental Monitoring

An environmental monitoring programme was started around Dungeness in May 1963. The first phase, a pre-operational survey, was to establish levels of radiation and radioactivity in the area around the power station before commissioning of the reactors. The later phase, the continuing operational survey, is to assess any changes in the levels that may occur after commissioning, due either to operational discharges or following an accident.

The pre operational survey included gross  $\alpha$  and gross  $\beta$  measurements on herbage, root mat and soil, gross  $\beta$  measurements on sea silt, sea water, shore silt, drinking water and  $^{131}\text{I}$  and milk, together with gamma dose rate measurements at a series of shore sites and on agricultural sites.

Gamma survey sites are selected in three rings between one half and one mile, between one and five miles and control sites at between fifteen and twenty miles. Measurement frequencies were initially set at monthly intervals for operator training and familiarity and later generally reduced to quarterly. Fig. 1 shows the mean value of herbage results resulting from the background fallout.

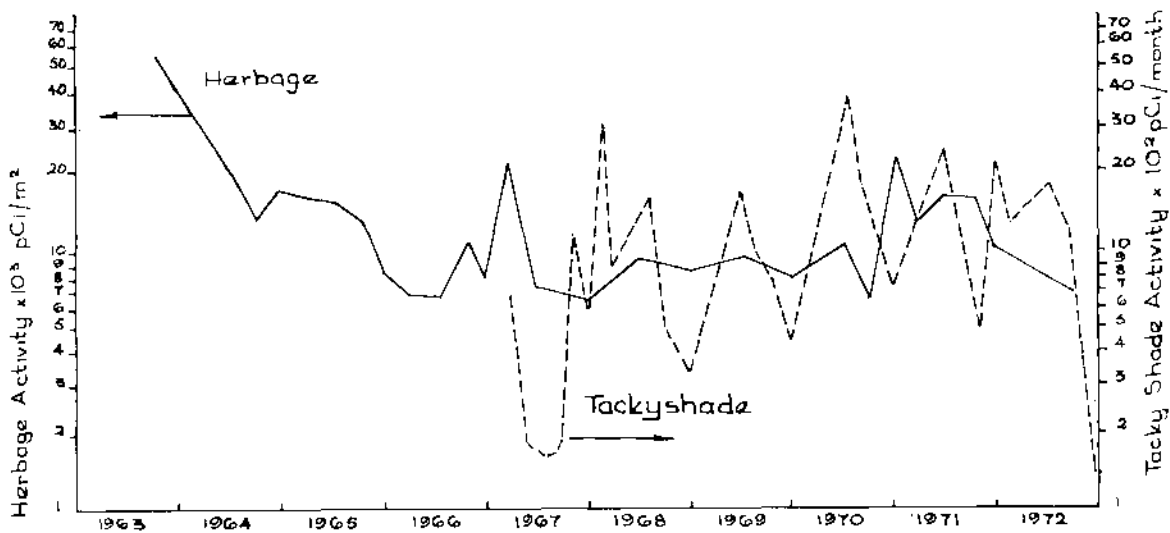


Fig 1 Herbage & Tacky Shade Results 1963-72 (mean of all sites)

In early operation of the station, the environmental monitoring programme remained unaltered but subsequently a number of alterations to the programme have been incorporated. For example sea silt measurements are now annual instead of quarterly and control sites milk samples would now only be assessed for  $^{131}\text{I}$  if  $^{131}\text{I}$  was found in local farm sites. A major change is that a more sensitive indicator than herbage has been devised to act as an indicator for station emission, namely the tacky shade<sup>5</sup>. Herbage samples are now only collected once per year from individual sites but tacky shades are exposed over two month periods at seven sites near the power station, and at five control sites. In addition shades are exposed along the station fence and within it. By measurement of the total activity or specific isotopic activity on an exposed shade, a measure of the integrated airborne activity is obtained. A deposition velocity for the collectors of about 1.5 cm/sec has been determined<sup>6</sup>. At sites with high wind runs, higher apparent integrated airborne activity levels are found, and

it has been shown that normalising factors for individual sites around a power station can be determined by analysis of site results over a long period<sup>6</sup>. These factors vary between 0.77 and 1.5 for the Dungeness sites. Experience to date shows that no measurements of radioactivity outside the station perimeter can be attributed to station emission. Mean values of tacky shade activities are included on Fig. 1.

In 1967, following a series of weapon tests, routine sampling showed a significant increase in  $^{131}\text{I}$  content of local milk production, at a time when most cattle were mainly under cover. Fig. 2 shows values of  $^{131}\text{I}$  in milk obtained at a typical site together with  $^{131}\text{I}$  activity on herbage. The occasion was used to mount a large programme of measurements to determine the practical limitation of local resources and valuable experience was gained.

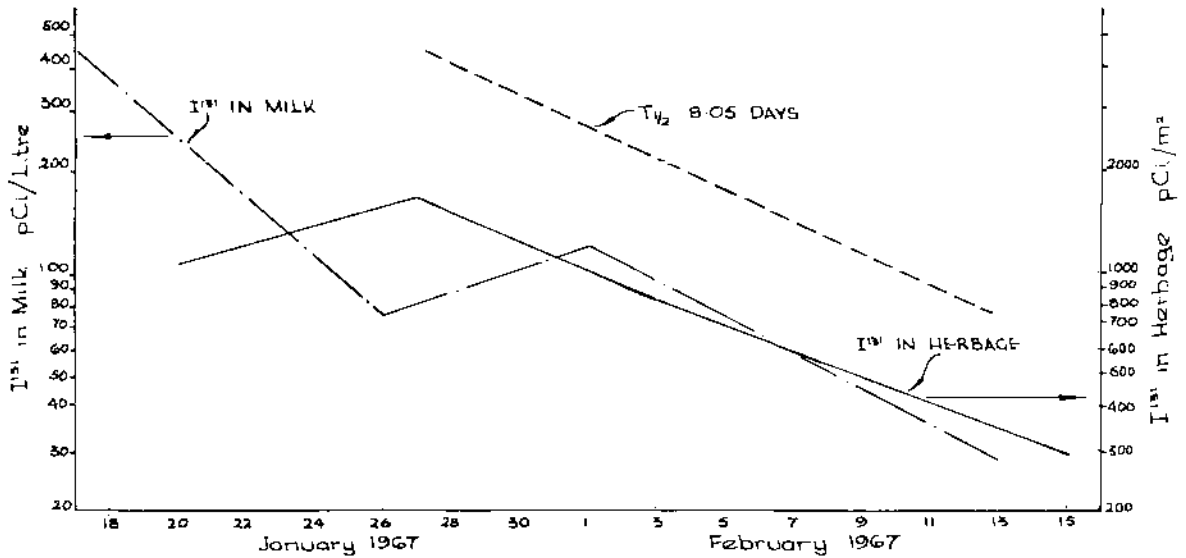


Fig 2 -  $^{131}\text{I}$  in Milk & Herbage - District Survey Exercise

Radiation Dosimetry

Whole Body Dose

Shielding design at Dungeness was on the basis of a maximum individual annual dose of 2.5 rem. The dose commitment to maintenance staff depends not only on plant conditions but on Health Physics techniques and awareness. Dose commitments to operators depend more on plant area dose rates and cannot be reduced significantly without major plant modifications.

In the very early years of operating with low integrated reactor fluxes, in pile activation levels were relatively low. Maintenance on reactor components did not therefore produce high dose commitments. Progressively dose rates due to activated components will increase while at the same time component modification and development may still be necessary. It may be expected that dose commitments would rise. A period of lower dose commitments should follow when "teething" problems have been solved.

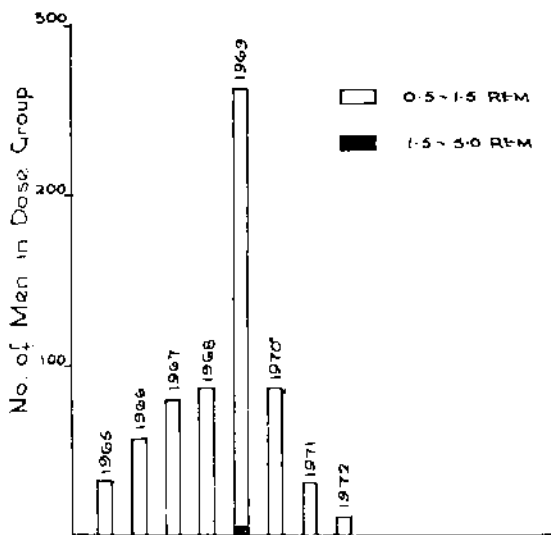


Fig 3. No. of Employees in Annual Dose Groups

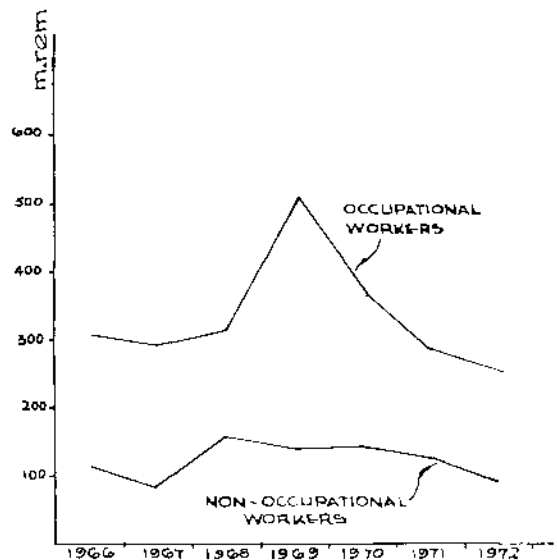


Fig.4 Mean Employee Annual Dose.

Fig. 3 shows the number of men who have received between 0.5 and 1.5 rem and between 1.5 and 5 rem whole body dose in any year between 1965 and 1972 inclusive. Fig. 4 shows the average whole body dose per year for all occupational workers at Dungeness. In 7 years there have only been 10 occasions when 1.5 rem has been exceeded in one year.

#### Gaseous Effluents

Gaseous and airborne particulate effluents arise from three sources namely carbon dioxide leakage and primary circuit purging, biological shield cooling air and active area ventilation systems.

Argon 41 is emitted at the rate of about 20 curies per hour for each reactor from the shield cooling systems, and in small additional quantities from carbon dioxide (about 10 Ci per day).

Gaseous particulate activities are measured by sampling through charcoal impregnated filter papers. Experience has shown since 1965 that particulate activities in carbon dioxide discharges include about 0.2 mCi per day total beta activity assessed against a  $Cl^{36}$  standard and alpha activities always below a minimum detectable value of about  $2(10)^{-5}$  mCi per day. A further 0.1 mCi/day total beta and negligible alpha activity are discharged with shield cooling air. The contribution from ventilation systems is negligible in comparison with shield cooling and carbon dioxide discharges. No significant trends in activity discharges have been observed since 1965 except at a point when sampling techniques changed to incorporate a charcoal impregnated filter paper with a much higher collection efficiency to  $As^{76}$  than the earlier papers used.

Carbon dioxide discharges include  $S^{35}$ ,  $As^{76}$ ,  $Ag^{110m}$ ,  $Co^{60}$ ,  $Zn^{65}$ ,  $Fe^{59}$  and  $Cr^{51}$ ,  $Fe^{55}$  with  $Ag^{110m}$  as the greatest individual contributor. Shield cooling discharges include  $As^{76}$ ,  $Br^{82}$ ,  $Cr^{51}$ ,  $Ag^{110m}$ ,  $Co^{60}$  and  $Fe^{59}$  with  $As^{76}$  as the greatest individual contributor.

Quantitative returns are made to the Department of the Environment based upon activity levels after 72 hours decay as a condition of the Authorisation to Discharge. No quantitative maximum permitted discharge has been set. The Authorisation required that discharges must be kept to the lowest practicable level.

#### Liquid Effluents

The terms of the Authorisation to discharge liquid radioactive waste, administered by the Ministry of Agriculture Fisheries and Food permit discharges up to 200 curies of total activity other than tritium per twelve consecutive months, together with 2,000 curies of tritium. Discharges are made from the active effluent treatment plant on a batch basis to the English Channel mixed with 20 million gallons per hour of cooling water. Waste passes through a sand pressure filter and sintered stainless steel filter before discharge. Samples from each 20,000 gallon batch are assessed for total beta and alpha activity and separately for tritium using liquid scintillation techniques. Principle activities are  $Cs^{137}$  and  $S^{35}$  and tritium.

Fig. 5 shows the discharges since 1965. Tritium produced as a result of the reaction  $Li^6 + n \rightarrow H^3 + \alpha$  on the lithium impurity in graphite, is removed from the reactors in the gas drying system. A peak value was expected about 2 years after start up followed by a continuous reduction due to burn up of the lithium. The figure shows this peak and shows that current discharges of tritium are about 1.5% of the authorised maximum. The figure also shows a peak in activity discharges other than tritium caused by a temporary period of release of  $Cs^{137}$  &  $Cs^{134}$  to the fuel element cooling ponds. Discharges of activity are currently about 15% of the permitted maximum.

Fig. 6 shows the variation in isotopic composition of the main contributors. In addition  $Cr^{51}$ ,  $Fe^{55}$ ,  $Nb^{95}$ ,  $C^{14}$ ,  $Sr^{90}/Y^{90}$ ,  $Sr^{89}$ ,  $Ca^{45}$ ,  $Ag^{110m}$  and  $Ru^{106}$  have appeared at concentrations greater than 1% at various times.

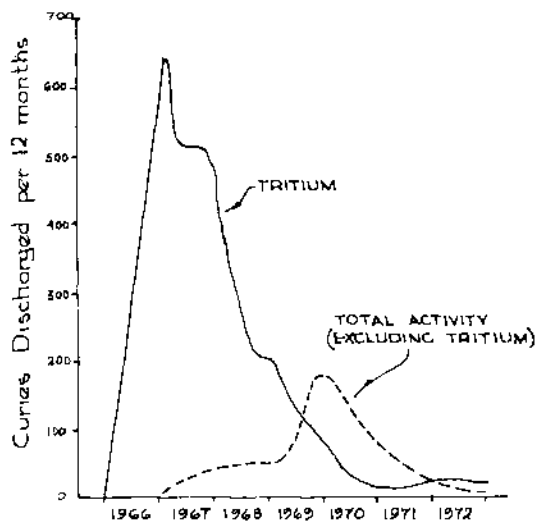


Fig. 5 - Liquid Effluent Discharges  
(Curies per Running 12 months)

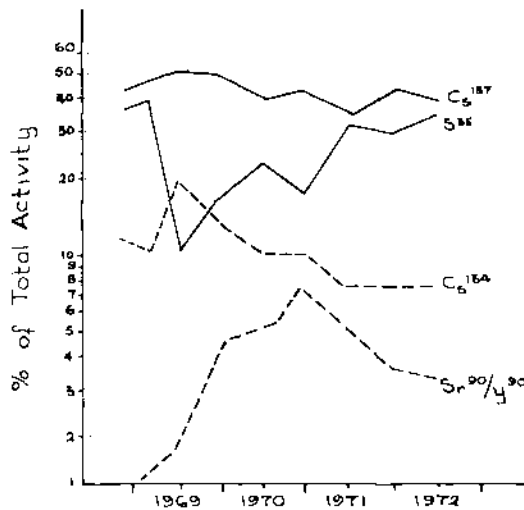


Fig. 6 - Liquid Effluent Isotope  
Composition.

## Radiological Control

The system of radiation and contamination zone control used within the CEGS<sup>7</sup> is used at Dungeness. A permit control system is used for all significant radiological work covering specific precautions and dosimetry requirements. Permits are issued by authorised senior Operation Engineers with the backing of accredited Health Physicists. Health Physics advice in the form of a written certificate is issued to the permit Engineer for work of significant potential radiation dose or for work on plant normally sealed but which may contain loose contamination. Permits or the equivalent are required even for entry into zones of highest radiological classification. Health Physics surveys may be undertaken prior to the issue of either Permits or Health Physics Certificates and Permits may require continuous Health Physics surveillance during work. These surveys would be undertaken by industrial staff Health Physics Monitors and in many instances include the measurement of airborne radioactive contamination during work progress.

Routine radiation surveys and routine surface contamination surveys are undertaken in all areas, the frequency being dependent on the likelihood of a hazard existing. Airborne radioactivity is assessed as an average over a two month period in areas where a low level hazard may exist intermittently or where no hazard should exist, by exposing indoor tacky cloth collectors in the form of a 1 square metre frame doubly covered with tack mat cloth.<sup>8</sup> Such collectors would be exposed in areas such as active laundry rooms, decontamination areas, active workshops, active effluent plant, active waste sorting rooms, fuel cooling pond buildings etc. Assessment of radioactivity by standard ashing and counting techniques has shown that averaged airborne radioactivity concentrations as low as  $10^{-6}$  pCi/cm<sup>3</sup> above background may readily be measured.

## Operational Health Physics Problems

### Radiation Accident

Early in 1966, when contractors responsibilities had not been completed, a contractor was investigating a fault on fuel pond machinery and moved what appeared to be a loose washer. The item was subsequently shown to be a stainless steel end cap from a fuel element with a contact dose rate of approximately 500 R/hr ( $\beta + \gamma$ ) and a gamma dose rate of about 1 R/hr at one foot distance. Reconstruction of the events subsequently demonstrated that a maximum hand exposure of 18.75 rem could have been received accompanied by a whole body dose of less than 100 m rem. As a result of the incident additional fixed gamma alarms were installed.

### Predominant Contaminant

In the main, loose radioactive contamination found at Dungeness is Ag<sup>110m</sup>. Its source is still unknown but whilst in general it is associated with smaller quantities of other isotopes such as Co<sup>60</sup>, Fe<sup>59</sup>, Cr<sup>51</sup> and S<sup>35</sup>, occasions have occurred when pure Ag<sup>110m</sup> has been identified. Microscopic examination<sup>9</sup> has shown that metallic silver exists sometimes associated with iron and copper. Construction records indicate that no silver, silver plating or silver bearing alloys were used in construction.

### Fuel Pond Bacterial Growth

Early in 1969 it became apparent that the walls and floor of the fuel element cooling ponds were becoming covered in a green slime even though conditions are maintained at pH 11.5. As the thickness increased, large pieces would break away leading to many problems such as blocking of filters. The slime appeared to have the characteristic that it absorbed radioactive contamination particularly insoluble Ag<sup>110m</sup>. Any pond equipment taken from the ponds became coated with slime and if allowed to dry would then cause spread of contamination. The slime turned out to result from non pathogenic bacteria. For operational reasons it was necessary to eliminate the cause. All known bacteriacides would have caused corrosion of the magnox canning material of stored irradiated fuel elements, releasing fission products to the water. It was not possible to remove all the fuel from the ponds without holding up the reactor refuelling cycle for at least three months. The course of action finally adopted was to place all fuel in one isolated bay of the pond complex and to empty and mechanically clean all the other bays. The water temperature in the bay with fuel in was then allowed to rise to 50°C by decay heating to kill the bacterial growth there. The resulting dead slime was cleared by recycling the water through a large muslin filter and finally by continuous dilution and purging. There was later an increase in the amount of Cs<sup>137</sup> and Cs<sup>134</sup> leached to the pond water by the fuel, ultimately causing the rise in liquid radioactive waste discharges shown in Fig. 5. Further formation of the bacterial growth is now prevented by keeping water temperatures below 20°C and by maintaining water movement.

### Shielding for Major Maintenance

It has been necessary on two occasions to construct major shielding assemblies for work on active components. Firstly it became necessary to modify 240 in pile steel mechanisms approximately 20 metres long with dose rates of some tens of rad/hr existed over about 5 metres. A cylindrical lead shielding assembly some 4 metres high and of diameter 2 metres with 15 cm wall thickness, lead glass viewing windows and remote tool access parts was constructed. The dose rate in the working position was about 5 mR/hr. The modification work without shielding would have taken about 15 minutes. With the shielding the work took approximately one hour for 2 men. Thus a potential radiation commitment of several hundred man rads was reduced to less than 2 man rads to complete all assemblies. Ultimately these same assemblies had to be replaced and the original ones

were cut into pieces and disposed to a waste void within the biological shield of the reactor. A second lead shielded cell was constructed over the disposal hole and the assemblies lowered in steps as pieces were cut off and allowed to drop into the void below.

#### Statutory Boiler Overhauls

It is a statutory requirement that the gas side of two boilers, and associated gas ducts are inspected every two years. This requires that the reactor is shutdown, cooled and depressurised and the sections to be inspected are isolated from the reactor CO<sub>2</sub> atmosphere and purged with air. Once an air atmosphere is established, a comprehensive Health Physics radiological survey is undertaken in all horizontal sections where small irradiated objects could ledge. The gas ducts are some 2 metres in diameter and the boilers some 20 metres high. Comprehensive surveys are therefore time consuming. They are undertaken by staff with full air line respiratory protection. On receipt of written clearance, inspection teams enter and later teams to undertake minor repairs such as to thermocouple pockets. Full telephonic contact is maintained by staff at the entry control point with teams inside the primary circuit. Surveys, inspections and minor repairs are usually completed within 1 week but other maintenance work is undertaken whilst the reactor is shut down. It has been found at Dungeness that general dose rates inside gas ducts and boilers due to loose and fixed radioactive contamination are only a few m rem/hr and reducing as years pass. Occasional small pieces of radioactive debris are found during surveys.

#### Routine Health Physics Programme

A nuclear site such as Dungeness requires a fairly comprehensive Health Physics service ranging from dosimetry, effluent control, environmental monitoring, decontamination laundries, etc. Associated with this there is a great deal of routine work either of a statutory nature or of a support nature. This work continues on a 24 hour basis. Many statutory requirements require sampling, measurement etc. and a wide range of instruments are used. These instruments require regular calibration. All breathing apparatus, air lines, suits and communications equipment need regular testing and inspection. A range of routines has been established divided into the following groups: Radiation Surveys, Surface Contamination Surveys, Gas Borne Particulate Activity Surveys (including primary coolant) District Survey, Breathing Equipment, Isotopic Analysis (including effluent analysis) Instrument Calibrations and Emergency Equipment checks. In all over 170 different routines are completed at frequencies varying from once per shift to once per year. Control of such a programme is complex and is fulfilled by dividing routines into two groups - those of frequency 1 week or less and those of frequency greater than one week. The former group are listed on either daily or weekly log sheets and the later are catalogued in a computer programme which gives a printout each 12 weeks of routine work due and date due. It is therefore relatively straight forward to satisfy inspecting authorities that statutory work is adequately completed.

#### Conclusions

Seven years experience of operating two magnox reactors at Dungeness have shown that personnel doses can be kept very low, currently at an average of 300 m rem per year for occupational workers. Off site measurements of radioactivity in the environment have shown that there is no measurable activity attributable to the operation of the station, even though the extremely sensitive tacky shade collectors are used for sampling. For the past three years liquid effluents have been below 15% of the permitted discharges. A programme of routine work controlled by computer ensures that statutory requirements are met.

#### References

1. Nuclear Power pp 76-103 April 1961.
2. M.J. Heard and B.E. Jones. A New Film Holder for Personnel Dosimetry A.E.R.E. Report M1178 1963.
3. J.R. Harvey, W.H. Eudd and S. Townsend. I.A.E.A. Symposium on Neutron Dosimetry Vienna 1972.
4. J.R. Harvey. Berkeley Nuclear Laboratories - C.E.G.B. England. Personal Communication.
5. J.K. Jones et al. Health Physics - June 1973
6. B. Cox, T.W. Evtett, E. Goldfinch, G. Lewis, M.J. Owens and B. Skelcher. Environmental Air Monitoring using Tacky Shade Collectors. Symposium Determination of Radionuclides in Environmental and Biological Materials. C.E.G.B. H.Q. London April 1973.
7. J.A. Bonnell and G.C. Dale. Health Physics 21, 637-642 (1971)
8. E.P. Goldfinch. Control of Low Level Airborne Contamination in Working Areas. 2nd European Congress on Radiation Protection. Budapest 1972.
9. D.A. Hilton. Berkeley Nuclear Laboratories - C.E.G.B. England - Personal Communication.

#### Acknowledgements

As this review covers work over the past seven years, the author wishes to acknowledge the considerable assistance given by colleagues past and present, and also wishes to gratefully acknowledge permission for the South East Region of the Central Electricity Generating Board for permission to publish the paper.

PIÉGEAGE ET DESORPTION DE L'IODE DANS LES  
REACTEURS GRAPHITE GAZ.

- J. BARBIER - Département de Radioprotection  
Electricité de France
- G. BENEZECE - DSN/SFSR  
Commissariat à l'Energie Atomique
- B. CADET - Centre de Production Thermique de Chinon  
Electricité de France
- J. KIRTBEL - DSN/SFSR  
Commissariat à l'Energie Atomique
- P. SIGLI - DPr/STEPPA  
Commissariat à l'Energie Atomique

Résumé

Les mesures d'exploitation effectuées dans les six réacteurs de type graphite-gaz exploités par Electricité de France ont permis de supposer l'existence de phénomènes de piégeage de l'iode par les structures internes.

Dans le but de confirmer cette hypothèse, une expérience fut entreprise sur le réacteur de Chinon 1. Elle consistait en l'injection d'iode radioactif dans le circuit primaire et en l'étude de l'évolution de sa concentration et de sa forme physico-chimique.

Les résultats montrent que la transformation de l'iode moléculaire en formes pénétrantes est très rapide. On observe une vitesse importante de piégeage par les structures internes. Le phénomène de désorption est lent.

Abstract

In the six gas-cooled reactors, which are operated by Electricité de France, the usual analyses have been leading us to suppose the being of iodine absorption phenomena by internal structures.

In order to corroborate this hypothesis, a test was attempted in Chinon 1 reactor. It consisted in the study of activity evolution and physico-chemical form of iodine after having introduced molecular  $^{131}\text{I}$  in the coolant.

The results show a very quick transformation of molecular iodine in penetrating forms. The trapping speed by internal structures is high, the desorption phenomenon is slow.

Introduction

Les six réacteurs de type graphite-gaz exploités par Electricité de France présentent à des niveaux différents d'un réacteur à l'autre, des contaminations en produits de fission résultant de pollution se présentant sous diverses formes.

Parmi les radionucléides produits, les iodes et plus particulièrement  $^{131}\text{I}$  font l'objet d'une surveillance soutenue.

Les mesures d'exploitation effectuées dans des conditions opératoires les plus variées montrent systématiquement une activité volumique en  $^{131}\text{I}$  beaucoup plus faible que celle que l'on pourrait logiquement attendre de la pollution en Uranium du cœur des réacteurs. C'est ainsi que le rapport entre les activités en  $^{131}\text{I}$  calculée et mesurée se situe selon le réacteur et son état de pollution entre 1000 et 10 000. Cette estimation repose sur l'hypothèse que la majeure partie des gaz nobles de fission émis reste dans le fluide caloporteur et que la répartition de l'iode et des gaz nobles entre les différents milieux constituant le réacteur (Uranium, fluide caloporteur, structures internes) se fait

dans les mêmes proportions.

L'importance de ce rapport nous a conduits à envisager un piégeage de l'iode par les structures internes du réacteur et à entreprendre des expériences destinées à mettre ce phénomène en évidence.

L'une d'entre elles, qui fait l'objet de cet exposé, consistait en une injection de quelques Curies d' $^{131}\text{I}$  dans le fluide caloporteur du réacteur de Chinon 1 et à étudier l'évolution de l'activité volumique ainsi que celle de la proportion des formes pénétrantes d'iode.

### Conditions expérimentales

Chinon 1 est le premier réacteur à Uranium Naturel, modéré au Graphite et refroidi au Gaz Carbonique, construit et exploité par Electricité de France. La première divergence a eu lieu le 16 Septembre 1962, le premier couplage au réseau est intervenu le 14 Juin 1963. Les principales caractéristiques en sont reportées sur le schéma, figure n°1, qui présente le principe du circuit primaire et permet de localiser les points d'injection de l'iode et de prélèvements.

### Dispositif d'injection

Le procédé de génération était fondé sur l'échange isotopique entre du radio-iodure de sodium sous forme solide et des vapeurs d'iode naturel.

Le dispositif d'injection se composait, en série, d'un réservoir de  $^{127}\text{I}$  moléculaire et d'un générateur contenant environ 4 Ci de radio-iodure de sodium. Le réservoir de  $^{127}\text{I}$  était pourvu d'un circuit de contournement destiné à assurer le réchauffage des circuits avant la génération proprement dite de  $^{131}\text{I}$ .

### Dispositifs de prélèvement

Le matériel de prélèvement était constitué de "Porte-charbon de Contrôle" doubles<sup>1</sup>, d'une part et de dispositifs de type "May-Pack"<sup>2</sup> d'autre part.

Les Porte-charbon de Contrôle devaient permettre de suivre l'évolution de l'activité volumique totale en  $^{131}\text{I}$  du fluide caloporteur. Chaque Porte-charbon de Contrôle était constitué de deux couches successives, de 5 cm d'épaisseur chacune, de charbon imprégné à l'iodure de potassium. Les conditions d'utilisation étaient les suivantes : débit de gaz prélevé 2 m<sup>3</sup>/h, vitesse frontale sur le charbon 20 cm/s, température du gaz analysé inférieure à 50°C.

Les dispositifs de type May-Pack étaient constitués, successivement dans le sens d'écoulement du gaz échantillonné de :

- 1 filtre en laine de verre comprimée destiné à arrêter  $^{131}\text{I}$  fixé sur les aérosols.
- 7 couches de "knit-mesh" en laine de cuivre argenté dans le but de retenir l'iode moléculaire.
- 3 couches de charbon non imprégné.
- 6 couches de charbon imprégné au triéthylènediamine (TEDA).
- 1 filtre en laine de verre comprimée pour arrêter les produits d'érosion des charbons.

Les couches de charbon avaient pour but de retenir les formes pénétrantes de l'iode. Chaque couche d'absorbant avait une épaisseur de 1 cm.

Deux dispositifs de type May Pack, prévus pour fonctionner à la pression du circuit primaire, furent placés, l'un à l'amont, l'autre à l'aval du réacteur. Leur but était de fournir des résultats intégrés sur la durée totale de l'expérience. Le débit de gaz prélevé s'élevait à 2 m<sup>3</sup>/h.

Les autres appareils, fonctionnant à la pression atmosphérique avaient pour but, de fournir des renseignements sur l'évolution dans le temps de chaque forme d'iode, moléculaire ou pénétrante. Le débit de prélèvement était réglé à 2 m<sup>3</sup>/h.



Les prélèvements effectués par les dispositifs de type May-Pack destinés à fournir des résultats intégrés sur la durée de l'expérience seront désignés par l'expression prélèvements continus. Les prélèvements effectués à l'aide des autres dispositifs de type May-Pack seront appelés discontinus.

#### Méthodes de mesure

Les activités ont été mesurées à l'aide de sélecteurs 400 et 4000 canaux à détecteur  $^7\text{Na}$  (4x4in) et Ge-Li.

#### Déroulement de l'expérience

L'ensemble de l'expérimentation s'est déroulé selon trois phases. L'activité volumique de  $^{131}\text{I}$  résultant de la pollution du réacteur avait été préalablement mesurée.

La première phase couvrait l'injection et les évolutions rapides à court terme. L'évolution escomptée de l'activité volumique de  $^{131}\text{I}$  étant rapide, il était indispensable d'effectuer des prélèvements de courte durée (5 mn) avec une périodicité de 10 mn. Il était donc nécessaire d'injecter une quantité de  $^{131}\text{I}$  suffisante pour que les activités recueillies durant cette première phase soient mesurables.

La seconde phase avait pour objet de suivre l'évolution de l'activité volumique de  $^{131}\text{I}$ , dans les conditions normales d'exploitation du réacteur. Les faibles activités volumiques du COP nécessitaient alors des prélèvements de durée accrue (15 mn, puis 30 mn, puis 60 mn).

La troisième phase devait permettre d'évaluer la rapidité de désorption des structures dans des conditions différentes c'est à dire réacteur à l'arrêt et circuits principaux vidangés.

#### Résultats expérimentaux

Une expérience préalable de génération fut effectuée en Laboratoire pour obtenir l'assurance que la production de formes pénétrantes d'iode ne devait être imputée qu'au réacteur ou au fluide caloporteur. Cette expérience consistait à générer  $^{131}\text{I}$  en atmosphère de  $\text{CO}_2$  de qualité commerciale. On a observé qu'au maximum 7% de l'iode généré était sous forme pénétrante.

On peut donc conclure de ce résultat que toute forme pénétrante constatée au cours de l'expérience principale est imputable à une transformation de l'iode moléculaire dans le réacteur ou les circuits principaux.

#### Forme physico-chimique de l'iode

Des prélèvements effectués avant l'injection sur dispositifs de type May-Pack ont montré une activité volumique en  $^{128}\text{I}$  du fluide caloporteur de  $3,5 \cdot 10^{-6} \text{ Ci/m}^3$ , uniquement sous forme pénétrante. Nous n'avons pas pu déterminer avec certitude la source de cette pollution en  $^{128}\text{I}$  qui ne peut être attribuée entièrement à l'activation de  $^{127}\text{I}$  de fission. Il y a donc lieu de supposer que  $^{128}\text{I}$  provient en majeure partie de l'activation d'iode naturel; de forme physico-chimique inconnue, introduit accidentellement dans le réacteur. Malgré ces inconnues, en raison des lois des réactions d'équilibre, on peut penser que les formes pénétrantes de l'iode sont prépondérantes en réacteur.

Les principaux résultats de l'expérience proprement dite sont les suivantes: 1°) Le tableau, figure n°2, regroupe les résultats des mesures d'activité volumique des deux formes de  $^{131}\text{I}$  obtenues à partir des dispositifs de type May-Pack.

La colonne 1 indique l'heure de début de prélèvement décomptée à partir de l'instant origine  $t_0$  correspondant à la fin de l'injection. La durée de chaque prélèvement était de 5 minutes. Les colonnes 2 et 3 indiquent l'activité volumique en  $^{131}\text{I}$  respectivement sous forme moléculaire et sous forme pénétrante.

2°) La proportion d'iode moléculaire sur les prélèvements continus est voisine de 15% sensiblement identique pour les deux échantillons. Cependant, il y a lieu de signaler que le prélèvement effectué à l'amont du réacteur a été mis en service 12 mn après son homologation, effectué à l'aval du réacteur et que, de ce fait ces prélèvements ne permettent pas, à eux seuls, de conclure quant à la vitesse de formation des formes pénétrantes.

Les principales conclusions sont les suivantes :

1°) Il y a transformation de l'iode moléculaire en forme pénétrante à l'intérieur des circuits principaux. L'examen du tableau, figure n°2, permet de le constater.

2°) La transformation de l'iode moléculaire en forme pénétrante est rapide. En effet, les constatations sont les suivantes :

- La proportion d'iode moléculaire sur le prélèvement continu effectué à l'aval du réacteur est de 15%. Ce prélèvement a débuté au temps  $T_0$ .

- L'échantillon discontinu prélevé à  $T_0 + 5mn$  contient 1% d'iode moléculaire.

Il résulte de ces constatations que l'apport d'iode moléculaire sur le prélèvement continu a eu lieu au tout début de l'expérience.

D'autre part, on constate que le second échantillon effectué à  $T_0 + 15mn$ , à l'aval du réacteur, ne contient plus d'iode moléculaire en quantité mesurable.

La disparition de l'iode moléculaire est donc beaucoup plus rapide que celle des formes pénétrantes. La différence de comportement des deux formes peut être attribuée à la transformation de l'iode moléculaire en forme pénétrante.

3°) La transformation de l'iode moléculaire en forme pénétrante s'effectue au cours du passage dans le réacteur.

En effet, le premier prélèvement discontinu, effectué à  $T_0 + 12mn$  à l'amont du réacteur, révèle une proportion d'iode moléculaire de 10%, alors que cette proportion n'est que de 1% sur le premier prélèvement discontinu effectué à l'aval du réacteur. Cette différence nous conduit à penser qu'une partie de l'iode généré est restée fixée sur les tuyauteries situées en amont du point de prélèvement amont. L'iode moléculaire retenu un temps relativement long sur les tuyauteries ne s'étant pas transformé, la conversion de l'iode moléculaire en forme pénétrante est imputable au passage dans le cœur du réacteur et s'effectue pratiquement complètement en un seul passage. Ce temps de passage est de l'ordre de 1 s.

La suite de l'exposé ne portera donc que sur l'évolution de l'iode sous sa forme pénétrante.

### Evolution de l'activité volumique totale du fluide caloporteur en $^{131}I$

Le diagramme, figure n°3, montre l'évolution de l'activité volumique en  $^{131}I$  suivant deux échelles de temps. La largeur des segments verticaux correspond au temps de prélèvement. On constate que la diminution de l'activité volumique du CO<sub>2</sub> en  $^{131}I$  est extrêmement rapide et que l'état d'équilibre est atteint au bout d'environ 80 heures. Les évolutions relevées à l'amont et à l'aval du réacteur sont identiques.

Ces résultats expérimentaux nous ont permis d'estimer que la constante de temps de piégeage de  $^{131}I$  est inférieure à 10 mn.

Le niveau d'équilibre de l'activité de  $^{131}I$  est resté constant à  $4 \cdot 10^{-10}$  Ci/m<sup>3</sup> jusqu'à l'arrêt du réacteur qui a eu lieu 20 jours après l'injection.

### Description de l'iode

L'évolution de l'activité volumique de  $^{131}I$  au cours de la période de fonctionnement aux conditions nominales a permis de fixer une limite inférieure à la constante de temps de désorption de l'iode. On a ainsi trouvé que cette constante de temps devait être plus grande que 40 jours. Il est en effet impos-

sible d'obtenir une meilleure précision en raison d'une activité volumique à l'équilibre en  $^{131}\text{I}$  après injection trop peu différente de celle mesurée avant l'expérience.

L'objectif de la troisième phase de l'expérimentation était de déterminer la constante de temps de désorption de  $^{131}\text{I}$ . Dans ce but, il fut procédé, à la pression atmosphérique, à un renouvellement aussi intense que possible de l'atmosphère des circuits, afin d'obtenir une activité volumique de  $^{131}\text{I}$  la plus faible possible. Une soufflante auxiliaire, destinée normalement à l'évacuation de la puissance résiduelle avait été maintenue en service pour assurer l'homogénéisation de l'atmosphère. Le renouvellement ayant été arrêté, l'évolution de l'activité volumique de  $^{131}\text{I}$  fut observée. Cette évolution est indiquée sur la figure n°4. On constate qu'elle présente un maximum au-delà duquel la pente de la courbe tend vers la période de  $^{131}\text{I}$ .

La constante de temps de désorption pour les conditions de température et de pression (température moyenne du graphite :  $65^{\circ}\text{C}$ , pression atmosphérique) a été estimée à environ 105 jours.

Cette expérience a permis également de constater que le phénomène d'adsorption de l'iode varie avec les conditions de fonctionnement du circuit. Ainsi, dans les conditions de cette dernière expérience, la constante de temps d'adsorption s'élevait à environ 13 heures, valeur très supérieure à celle trouvée lors du fonctionnement nominal. Le débit de la soufflante auxiliaire étant très faible vis à vis du débit nominal du fluide caloporteur, il est très vraisemblable que le débit de gaz a une influence sur la vitesse d'adsorption de l'iode.

#### Conclusion

Dans les réacteurs graphite-gaz :

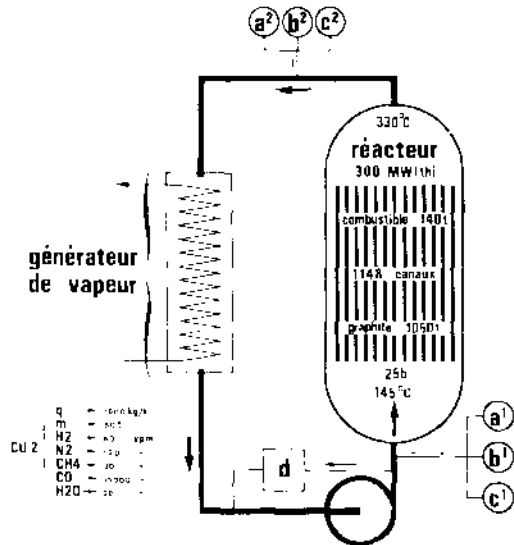
- 1°) Les formes pénétrantes de l'iode - et en particulier l'iodure de méthyle - sont des formes stables et rapidement prépondérantes en réacteur.
- 2°) Il y a piégeage rapide de l'iode par les structures internes. La constante de temps de piégeage, pour les conditions d'exploitation de Chinon 1, a été estimée inférieure à 10 minutes.
- 3°) Le phénomène de désorption est très lent.

Il faut, cependant, être très prudent dans l'utilisation de ces résultats, et en particulier ceux concernant l'adsorption, pour d'autres réacteurs et dans d'autres conditions. Il est, en effet, très probable que la vitesse de piégeage de l'iode dépend, entre autres paramètres, de la configuration des circuits et du réacteur, de la géométrie du canal et du débit de fluide caloporteur. On doit constater, en particulier, une différence de comportement de l'iode, pour un même réacteur, s'il est en marche nominale ou s'il vient d'être arrêté brusquement sur incident de combustible. Cette remarque est fondée sur des observations faites par ailleurs qui ont montré une augmentation appréciable d'activité volumique de  $^{131}\text{I}$  alors que le réacteur venait de passer de la pleine puissance à la puissance nulle. Le seul facteur important dans cette évolution croissante semblait être la diminution importante du débit de fluide caloporteur.

Ainsi, en cas d'accident survenant sur le combustible, paraîtrait-il souhaitable de maintenir le débit maximal de fluide caloporteur après la chute de barres afin de conserver le plus longtemps possible le bénéfice des phénomènes décrits dans cet exposé.

#### Bibliographie

- |                             |   |
|-----------------------------|---|
| 1 - P. BILARD - J. BRTON    | Contrôle des installations d'épuration de l'air. Essais de conformité des éléments Tests in situ. Rapport CEA R 3227-1967 |
| 2 - M.J. YEGAW and F.G. MAY | The behaviour of Iodine released in Reactor Containers - J. of Nuclear Energy - 1962                                      |



prélèvements discontinus | c - porte charbon de contrôle  
a - dispositifs de type "May Pack"

prélèvements continus | b - dispositifs de type "May Pack" à la pression du circuit  
d - générateur d'iode

Figure n°1 Schéma de principe du circuit primaire de Chinon 1 montrant l'implantation des prises d'injection et de prélèvement

Activité volumique des deux formes de <sup>131</sup> I		
Prélèvements discontinus		
temps	<sup>131</sup> I moléculaire μCi/m <sup>3</sup>	<sup>131</sup> I forme pénétrante μCi/m <sup>3</sup>
	2	3
Amont réacteur		
t <sub>0</sub> + 12 min	0,007	0,07
Aval réacteur		
t <sub>0</sub> + 5 min	0,003	0,278
t <sub>0</sub> + 15 min	non mesurable	0,05
t <sub>0</sub> + 35 min	d'	0,02
t <sub>0</sub> + 45 min	d'	0,01
t <sub>0</sub> + 55 min	d'	0,005

Figure n°2 Activité volumique des deux formes d'iode obtenue à partir des dispositifs de prélèvements discontinus

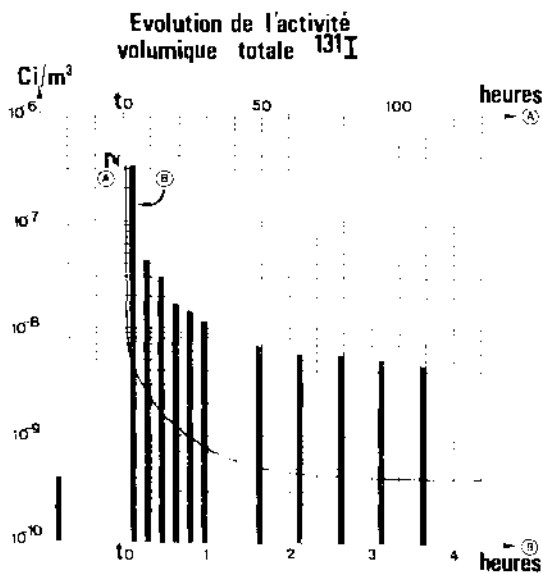


Figure n°3 Evolution de l'activité volumique totale <sup>131</sup>I en fonction du temps

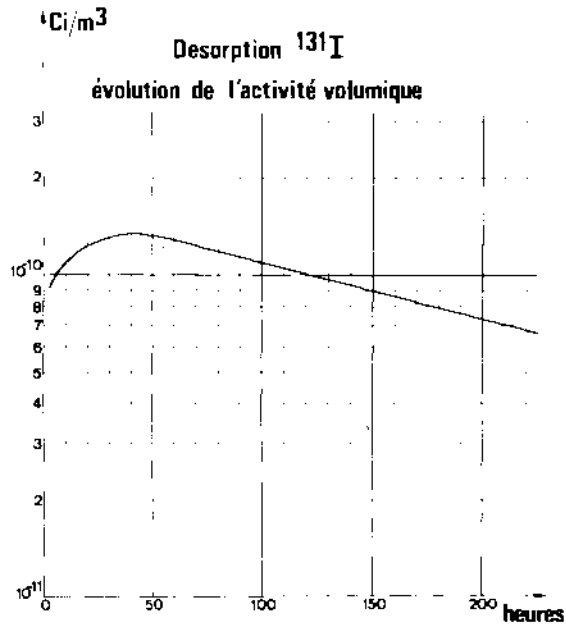


Figure n°4 Expérience de désorption de <sup>131</sup>I Evolution de l'activité volumique

## ADSORPTION OF FISSION NOBLE GASES ON CHARCOAL

Abraham S. Goldin, Dwight W. Underhill and Helena A. Trindade\*  
Harvard School of Public Health  
Boston, Massachusetts 02115

Recent research<sup>1</sup> on adsorption of krypton and xenon by charcoal from an argon carrier stream has shown that the adsorption coefficient of Kr is almost independent of pressure at  $-120^{\circ}\text{C}$ , and actually decreases with pressure at  $-140^{\circ}\text{C}$ , over the range from two to six atmospheres. This paper reports further work in correlating the adsorption of Kr and Xe from Ar streams with the adsorption properties of Ar itself. It also reports measurements of the adsorption of Kr and Xe from streams of  $\text{N}_2$ , which may be used as a cover gas in some Fast Flux Test Facility (FFTF) applications. These measurements are also correlated with the adsorption of the  $\text{N}_2$  carrier gas. Some additional data on adsorption of the two fission noble gases from air and from a 60:20:20 mixture of  $\text{N}_2$ -Ar- $\text{O}_2$  are presented.

### Experimental

The experimental system has been described.<sup>1</sup> A schematic drawing of the apparatus is shown in Figure 1. A number of adsorption beds containing from < 1 to 480 grams of Pittsburgh PCB charcoal, 6 x 16 mesh, were used. For runs below ambient temperatures, the beds were contained in a plastic foam container which was cooled by a thermostatically controlled internal spray of liquid  $\text{N}_2$ . The carrier gas was cooled to operating temperature by passage through a copper coil before it reached the bed. The bed temperature remained constant within less than  $1^{\circ}\text{C}$  during a run.

Tank carrier gas was introduced into the system at the desired pressure and allowed to pass through the bed until the desired temperature, pressure, and flow rate conditions had stabilized. A pulse of  $^{85}\text{Kr}$  or  $^{133}\text{Xe}$  was introduced through the injection port. The flow of carrier gas was then resumed, and the effluent monitored for radioactivity with a 500-ml spherical ionization chamber connected to a Cary vibrating reed electrometer. The volume passed through the bed was measured with a wet test meter. Concurrent readings of elapsed time, the electrometer, and the wet test meter were taken at intervals from the time of injection of the radioactive gas. The readings were computer analyzed to give average temperature, average flow rate, and dynamic adsorption coefficients.

Adsorption characteristics of the carrier gases were measured by passing them through the bed for a time sufficient to assure equilibrium. The input flow was then shut off and the bed allowed to warm to ambient temperature and pressure. The volume of gas desorbed from the bed, corrected for expansion of gas in the void volume and for residual adsorption at room temperature and pressure, was divided by the weight of the charcoal to give the adsorption coefficient in liters (STP) per gram. The experimental procedure

---

\*Present Address: Universidade Federal Do Rio De Janeiro  
Rio De Janeiro, Brazil

has been described in detail.<sup>2</sup>

## Results

### Adsorption of and from argon:

The adsorption of Ar on activated PCB charcoal was measured over ranges of temperatures and pressures from  $-140^{\circ}\text{C}$  to ambient temperature and from about 1.4 to 11 atmospheres (absolute). These data are graphed in Figure 2, which shows that the increase in adsorption coefficient with pressure is less at lower temperatures. These data, which had been obtained primarily for estimation of holdup of  $^{41}\text{Ar}$  in the FFTF, proved to be valuable in correlating the data for adsorption of Kr and Xe from Ar.

When these results were plotted according to the procedure of Polanyi<sup>3</sup> they fell on a single line, except for the ambient temperature points, which were high by a factor of 2.5 to 4.5. It is possible that the extrapolation of the thermodynamic functions to ambient temperature is unsatisfactory. The Polanyi plot for all our data, including some results at an Ar partial pressure of 0.34 atmospheres, is given in Figure 3. These low partial pressure data were obtained by measuring the Ar adsorption from a 3:1 mixture (v/v) of He and Ar. Similar data of Grant and Manes<sup>4</sup> agree with the low temperature portion of Figure 3.

In the Polanyi plot, the ordinate is the quantity of Ar adsorbed per gram of charcoal, expressed as the volume of liquid Ar at the temperature where the vapor pressure is equal to the adsorption pressure. The abscissa is  $(T/V_b) \log (f_s/f)$ , which is proportional to  $a$ , the work required to compress the unadsorbed gas into unit volume of adsorbed phase. In this expression,

$T$  is the absolute temperature,

$V_b$  is the molar volume of liquefied gas at the adsorption pressure,

$f_s$  is the fugacity of liquefied gas at the temperature of adsorption, and

$f$  is the fugacity of the gas.

Fugacity coefficients  $f/p$  were calculated according to Reid.<sup>5</sup> Data for vapor pressure and molal volumes were taken from Perry.<sup>6</sup> At temperatures above the critical temperature, the fugacity of the liquefied gas was calculated by applying the fugacity coefficient to the vapor pressure obtained by linear extrapolation of the vapor pressure vs  $1/T$  relationship.

### Adsorption of and from nitrogen:

Measurements of Kr and Xe adsorption coefficients for Pittsburgh PCB activated charcoal from  $\text{N}_2$  streams were made at pressures of 1.3 to 3.0 atmospheres, and at temperatures from ambient to  $-100^{\circ}\text{C}$ . Xe adsorption was measured from ambient temperature to  $-50^{\circ}\text{C}$ . These results are shown in Figure 4. As with adsorption from an Ar carrier gas, the pressure dependence is less at the lower temperatures, and the curves are concave downward, indicating the presence of saturation effects. The adsorption of the  $\text{N}_2$  carrier was also determined; these values are plotted as a Polanyi graph in Figure 5.

### Adsorption from other gases:

There is a possibility that inert gas blankets ( $\text{N}_2$  or Ar) might become contaminated with  $\text{O}_2$  from air in-leakage, particularly under accident conditions. Accordingly a program of measurement of adsorption from  $\text{N}_2$ - $\text{O}_2$  mixtures was planned. The first step in this program was measurement of adsorption from air. The measurements given in Table 1 showed that adsorption from air was essentially the same as adsorption from  $\text{N}_2$ . For this reason, no measurements were made with other  $\text{N}_2$ - $\text{O}_2$  mixtures. Table 1 also shows a few measurements from a 60:20:20 mixture (v/v/v) of  $\text{N}_2$ , Ar, and  $\text{O}_2$ . Since these measurements also showed no particular difference from  $\text{N}_2$  adsorption, no further measurements were made using this mixture.

TABLE 1

All values are adsorption coefficients in liters (STP) per gram of charcoal.

Temperature °C	Pressure atm	Krypton			Xenon		
		N <sub>2</sub>	Air	N <sub>2</sub> -Ar-O <sub>2</sub> (60:20:20)	N <sub>2</sub>	Air	N <sub>2</sub> -Ar-O <sub>2</sub> (60:20:20)
-50	1.29	0.42	0.45		17.9	17.2	7.2
-50	3.04	0.62	0.64		20.9	17.2	
-20	1.29				4.7	4.7	
	3.04	0.30*		0.31	6.8	6.8	7.2
ambient**	1.29	0.061	0.060		0.97	0.96	
ambient	2.0	0.085	0.088		1.		
ambient	3.04	0.12	0.12	0.12	1.61	1.70	
-70	3.04	1.05*		1.14			

\*Extrapolated value (from Figure 6 )

\*\*Ambient temperature = 25°C for Kr, 22°C for Xe.

#### Summary of adsorption data:

All the data on adsorption of Kr and Xe at 1.3 and 3.0 atmospheres are summarized in Figure 6. Curves for the data previously obtained<sup>1</sup> are included for comparison. Individual points for adsorption from air and from the N<sub>2</sub>-Ar-O<sub>2</sub> mixture are included. There is a difference between Ratney's single Xe points and the extrapolation of our Xe curves, and also between his Ar curves and ours. It is known that different lots of charcoal can exhibit differences of this magnitude, but another possible reason may be differences in regeneration of the charcoal. Regeneration was accomplished by heating at 95°C 30 to 60 minutes in a helium flow.

The characteristics of the curves can be briefly summarized. The curves for the adsorption of xenon are essentially exponential with 1/T, as is also the adsorption of Kr from He. The slope of the low-pressure Xe curve is very slightly less than that at higher pressures so that the pressure effect decreases with temperature. The curves for the adsorption of Kr from N<sub>2</sub> are concave downward, and the difference between the curves is again less at lower temperatures. The data points for adsorption from air and from the mixture confirm the similarity of adsorption from these mixtures and from N<sub>2</sub>.

#### Discussion

The adsorption data for Ar and N<sub>2</sub> have been interpreted, by the Polanyi theory, as a measure of the uptake by the carbon of these carrier gases as cm<sup>3</sup> of a liquid or quasi-liquid state per gram of charcoal. The pressure-normalized adsorption coefficients of krypton have been plotted against the coverage of charcoal by argon and by krypton. These graphs are shown in Figures 7 and 8. The points at zero uptake of carrier gases were obtained from adsorption data for Kr in He carrier.<sup>1</sup> These graphs are families of curves, rising sharply near zero carrier gas loading.

There are clearly two adsorption domains. One is predominant when the adsorption of the carrier gas is small. It is characterized by high adsorption at zero carrier loading and very rapid decrease with carrier loading. We may speculate that this corresponds to availability of adsorption sites when the charcoal is not completely covered. The second domain is characterized by a less sharp exponential decrease in adsorption with carrier gas loading. We may speculate that this corresponds to complete coverage of the charcoal with quasi-liquid carrier and consequent progressive unavailability of pore volume, beginning with the smallest pores and progressing to larger ones. Solubility of the trace gas in the carrier may be a sorption mechanism also.

The curves for temperatures near and below the critical temperature of  $-122^{\circ}\text{C}$  for Ar do not differ in nature from the curves at higher temperatures for both carrier gases. This similarity indicates that the anomalous pressure dependence of Kr adsorption from Ar at low temperature is not the result of a change in the state of the adsorbed Ar as had been suggested<sup>1</sup> but is caused by the progressive loading of the charcoal with carrier gas.

In Figure 8, the adsorption of Kr from  $\text{N}_2$  and from Ar at  $-100^{\circ}\text{C}$  is plotted vs quantity of liquefied carrier gas sorbed in the charcoal. The adsorption coefficients from Ar are about 40% higher than those from  $\text{N}_2$ . This difference is about the same as was observed between the previous Ar adsorption data<sup>2</sup> and the limited amount of new data reported in this work.

#### Summary

Adsorption coefficients of Kr and Xe on Pittsburgh PCB charcoal from  $\text{N}_2$  are within 40% of those from Ar carrier. Adsorption coefficients of these noble gases from air and from 50:20:20  $\text{N}_2$ -Ar- $\text{O}_2$  carrier are not significantly different from those obtained with nitrogen carrier.

Semilog plots of pressure-normalized adsorption coefficients versus volume of liquid (or quasi-liquid) carrier gas adsorbed per gram of charcoal are families of smooth curves. These curves are characterized by a very rapid decrease of trace gas adsorption at low levels of carrier uptake, followed by a less rapid exponential decrease with additional carrier uptake. Sets of adsorption data plotted in this way permit interpretation of adsorption behavior over a wide range of temperature and pressures.

#### Acknowledgement

The authors wish to express their appreciation to Mr. Floyd Hardwick for technical support, and to the U.S. Atomic Energy Commission for financial support under Contract AT (11-2) 3019.

#### References

- R.S. Retney and D.W. Underhill, "The Effect of High Pressure and Low Temperature on the Adsorption of Xenon and Krypton from Helium and Argon Streams", USAEC Report #CONF-720823, pp. 48-59, Jan., 1973.
- D.W. Underhill, R.R. Hall, A. Gold, A. Goldin, M.W. First, and D.W. Moeller, USAEC Report #COO-3019-4, pp. 2-1 to 2-15, Feb., 1973.
- D. Young, and A. Crowell, Physical Adsorption of Gases, Butterworth, London, 1962.
- R. Grant and M. Manes, "Correlation of Some Gas Adsorption Data Extending to Low Pressures and Supercritical Temperatures", Ind. Eng. Chem. Fundamentals 3, 221, 1964.
- R.C. Reid and T.K. Sherwood, The Properties of Gases and Liquids, McGraw Hill Book Co., New York, 1966.
- R.H. Perry, C.H. Chilton, and S.D. Kirkpatrick, Chemical Engineers' Handbook, 4th Edition, McGraw Hill Book Co., New York, 1963.



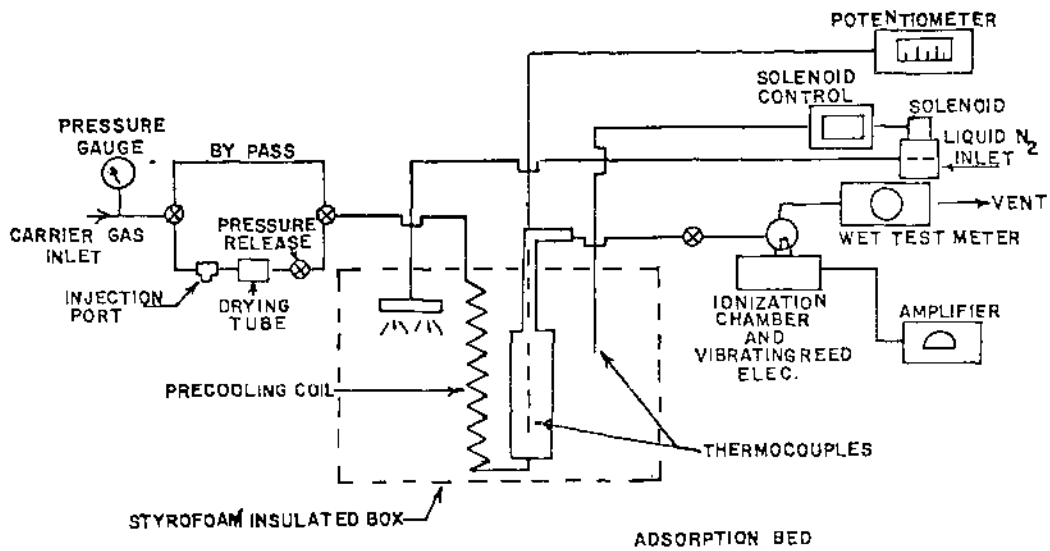


DIAGRAM OF TEST APPARATUS.

Figure 1.

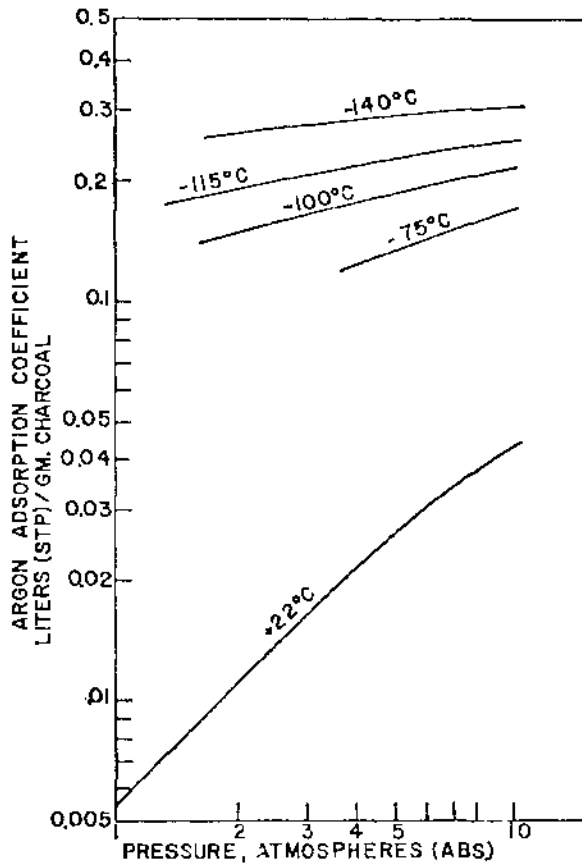


Figure 2.

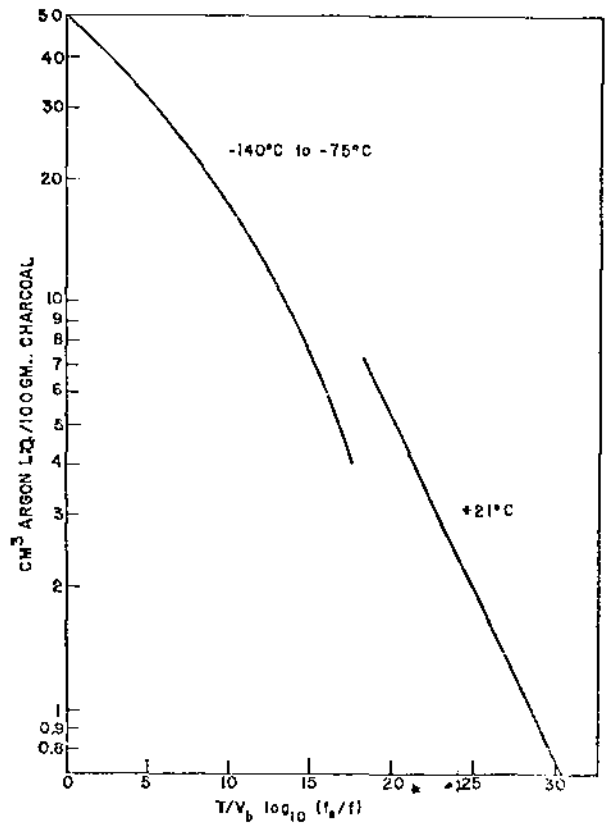


Figure 3.



NEW LOW LEVEL ENVIRONMENTAL RADIATION MONITORING  
AROUND NUCLEAR FACILITIES  
DISCRIMINATING FROM NATURAL RADIATION

Shigeru MORIUCHI, Kazuhiko IMAI  
and  
Ichiro MIYANAGA  
Japan Atomic Energy Research Institute  
Tokai-mura, Ibaraki-ken, Japan

Abstract

Recently, gaseous effluents from nuclear reactors and other facilities are controlled in such a low level, that the gamma radiation exposure from them amounts less than 10 mrem/yr at the site boundary.

Natural gamma radiation fluctuates due to the fluctuation of radioactivity in air, the sweeping effect by rain, shielding effect by snow and water content in soil, and other factors. It would be very difficult to measure gamma exposure less than a few mrem/yr from gaseous effluents discriminating from such natural background fluctuations even by using high-pressure ionization chambers.

For monitoring environmental radiations from nuclear facilities, discriminating the fluctuating natural radiation, a NaI(Tl) scintillation exposure rate meter with flat energy response is used. This equipment consists of a 2 x 2-inch NaI(Tl) scintillator and two separate electronic units, one is an exposure rate meter having a pulse height weighting circuit for flattening the energy response and other a single channel pulse height analyzer, and was tested to see the stability for a long-run operation in the field and to examine discriminating characteristics.

Natural radiation has wide energy spectrum distribution up to 2.62 MeV, and changes with time rather uniformly in wide energy range. Therefore, one can measure the environmental radiation from nuclear facilities by detecting total gamma radiation in full energy range and natural gamma radiation in the higher energy channel separately and taking the differential between them.

Results of the test was satisfactory for measuring increases in exposure less than 1 mR/yr of gamma radiation from a research reactor under natural fluctuation of background and showed a good stability in a long-run operation in the field.

Introduction

According to the prospective increase of the person-rems of the population exposure around the nuclear power stations, which may play the main role for the increasing requirement of energy, the level of environmental radiation exposure from them recently tends to be controlled as small as 10 mrem/yr compared with 100 mrem/yr of natural radiation. Under these circumstances, efforts are given to improve environmental monitoring technique to measure the small fraction of radiation from nuclear facilities separately from the fluctuation of natural background radiation, particularly by the use of a high-pressure ionization chamber.<sup>1,2</sup>

Beck et al. pointed out that the annual dose at certain point will be given

in the fractional time period of the year due to the wind-rose, so the actual dose rate will be sufficient to measure, even when annual dose rate is less than 1 mrad/yr.

The instrument of the environmental monitoring should have the characteristics such as high sensitivity, operation stability in the long range field use and simple treatment of data obtained. From the view points above, the present paper describes a new gamma radiation monitor using a NaI(Tl) scintillator for field use, and especially on the discriminating characteristics from the fluctuation of natural background radiation.

Time variation of natural radiation and its discrimination

Characteristics of the fluctuation of natural radioactivities

The cause of the fluctuation of natural radiation comes from the fluctuation of Rn and Tn daughters in the atmosphere, combined with the meteorological conditions. Because the amount of Rn is about 10 times as large as Tn at ground level, the modulation of natural gamma radiation is mainly due to Rn daughters, though there are some effects as shielding by soil moisture and snow cover.

Predominant gamma emitters of Rn daughters are RaB and RaC because the branching that occurs at RaA and at RaC can be ignored, and the fraction of gamma radiation exposures from RaC is almost 87 % in the equilibrium conditions both from air and from ground surface. In the atmosphere, the equilibrium conditions of RaB and RaC will be kept during slow fluctuations such as diurnal variation due to the stability condition in atmospheric air. The worst condition may occur in the rainfall by its sweeping effect, but the fairly good equilibrium can be expected to be kept between RaB and RaC in rain water on the ground surface. The observation of the differential spectrum from the beginning of rainfall shows small decrease of the ratio RaB/RaC with time, but the change of the ratio was negligible. Thus, the fluctuations of natural gamma radiation can be represented by the fluctuations of RaC's radiation even in the most severe unequilibrium condition during rainfall.

Discriminating method using gamma energy spectrum

It was shown that the spectrum of natural gamma radiation did not change depending on rains, and its highest energy is 2.45 MeV from RaC. The highest fraction of individual gamma exposures in the equilibrium condition comes from

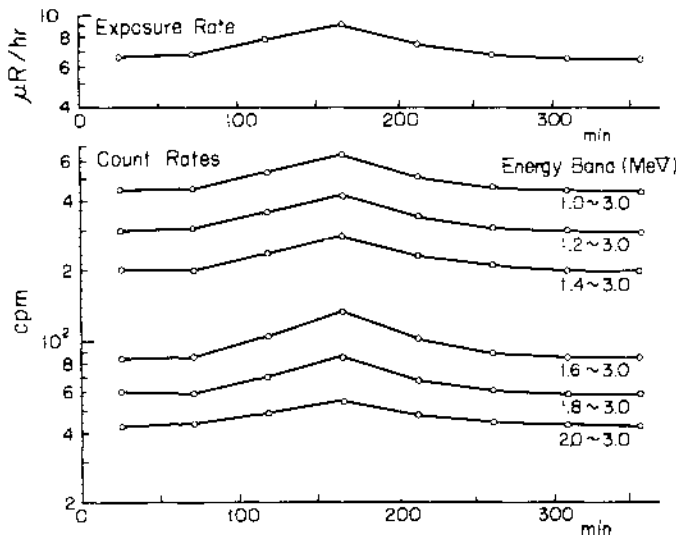


FIG.1. Time variation of background gamma radiation due to rainfall

1.76 MeV of RaC, and exposures from radiations between 1.73 MeV to 2.45 MeV of RaC occupies more than 60 % of total exposure. Gamma emitters of gaseous effluents from nuclear facilities are Ar-41 from research reactors and Kr and Xe from light water reactors. In order to decrease the release of gaseous effluents, most power reactors are equipped with some hold-up systems for them. In general, the short half-lives such as Xe-138 are negligible, and other nuclides of Xe and Kr all have the energies less than 1 MeV, excluding Kr-87,88.

With the consideration of the gamma ray spectrum above, the discrimination of the contribution of gaseous effluents from natural radiation can be performed with pulse height discrimination technique. Fig.1 shows the change with time of exposure rate and count rates above various discriminating level of natural radiation during rainfall, using a 2 x 2-inch NaI(Tl) scintillation counter which was set at 1 m above ground surface. Exposure rates were obtained from pulse height distribution by using Spectrum-Dose conversion operator.<sup>3-6</sup> The level of the background radiation after rainfall was reduced by 0.31  $\mu$ R/h compared with the level before rain, and the almost all of them may correspond to the fraction of the absorption introduced by rain water in soil. By considering a straight line connecting two points of the background level before and after rainfall, correlation between differential exposure rate and count rates from rainfall can be plotted as shown in Fig.2. The relation between both elements is found to be well proportional in any energy ranges. This shows the relation holds in any atmospheric conditions from the discussion above.

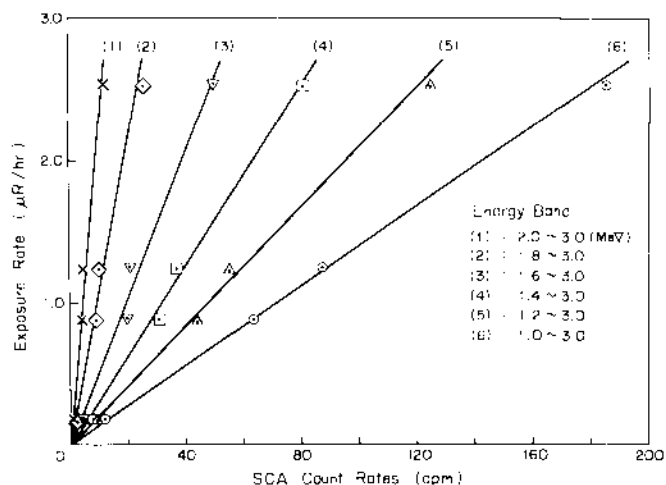


FIG.2. Relation between exposure rate and SCA count rates due to rainfall in given energy band

Features of the instrument for the discriminating measurement

NaI(Tl) scintillation exposure rate meter

We have developed a NaI(Tl) scintillation exposure rate meter, which has a flat energy response, and gave the principle and electronic circuits in several reports.<sup>6-9</sup> In the meter, energy loss spectrum produced in a NaI(Tl) crystal by gamma radiation is converted to exposure by applying Spectrum-Dose conversion function, which is carried out in an electronic circuit built in it.

Sensitivity is inversely proportional to the range of energy where the energy response is flattened, chosen simply by the electronic circuit adjustment. Counting efficiency is 110 cpm per  $\mu$ R/h in the case of a 2 x 2-inch NaI(Tl) crystal and 350 cpm per  $\mu$ R/h in a 3 x 3-inch NaI(Tl) crystal, when the flattening energy range is 60 keV to 3 MeV.

The fraction of counting due to cosmic radiation can be reduced as low as 0.2  $\mu$ R/h equivalent. The output signal is digital pulse, and consequently the measurement is simple and the accuracy is excellent. The contribution from the

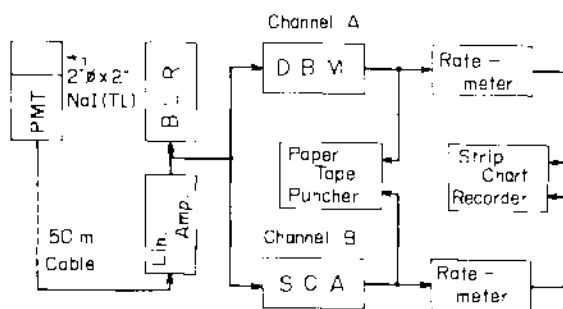


FIG.3. Block-diagram of two channel gamma radiation monitor

material contamination is gamma rays from K-40 in the glass of a photomultiplier tube and a scintillator window, and can be easily reduced to less than  $0.1 \mu\text{R/h}$  by selecting material.

Fig.3 shows the block-diagram of an instrument which can measure gamma radiations of gaseous effluents discriminating from natural radiations. In the figure, channel A is the exposure rate meter with an energy flattening circuit, namely Discriminator Bias Modulation circuit (DBM), and channel B gives count rates from natural radiation between setting level of a lower level discriminator (LLD) and 3 MeV of an upper level one. The differential between two channels gives the radiation from gaseous effluents only.

#### Long-run stability in a field test

The stability characteristics of the instrument relate mainly to temperature dependency of light emission efficiency of NaI(Tl) ( $+0.2 \sim +0.3 \%/^{\circ}\text{C}$ ) and temperature characteristics and stability of the high voltage unit. Long-run test of stability was carried out in the field over the period of five months, and the sensitivity was checked every day in first one month and every three or four days on the later four months by a checking source. The results showed  $\pm 3 \%$ /month covering the test period, in spite of the temperature change over  $10^{\circ}\text{C}$  experienced during day-time and night-time.

#### Examples of observation and the analysis

In our establishment, gamma radiations concerned are sky shine from a 200 MeV Linear Accelerator and gamma rays from Ar-41 cloud released from research reactors in JAERI and from a neighbouring gas cooled power reactor of JAPCO.

The level of LLD in channel B was set between 1.6 MeV avoiding 1.46 MeV photopeak of K-40 and 1.76 MeV photopeak from RaC. Data of the measurement were recorded on a strip-chart recorder and a paper tape puncher automatically. The former was put just to observe the counting pattern, and the latter, the punched paper tape was used for the precise evaluation of exposures by computer data-processing.

Fig.4 shows some typical examples of the pattern obtained from both channels A and B. Dotted lines in the figure show background radiation level. Fig.4(a) shows a case that small amounts of exposure rate due to the plume from JAPCO are superimposed on remarkable changes of background radiation from Rn daughters brought down by rain. As Ar-41 release rate from a 65 m stack of this plant is about  $0.8 \text{ Ci/h}$ , averaged exposure rate just under the plume is presumably less than  $0.5 \mu\text{R/h}$ . By the patterns of two channels, it is seen that the separation of the components is very clear, which a conventional one channel system can never do. The results were evaluated to be  $43.2 \mu\text{R}$  from rainfall and  $3.7 \mu\text{R}$  from the plume during the shown period. In this example the level of LLD is 1.75 MeV, and the conversion factor for rainfall from channel B

to channel A is  $0.060 \mu\text{R/h}$  per cpm. Two examples (b), (c) shown in Fig.4 were ones measured at lower discriminator setting  $1.63 \text{ MeV}$ , and in this case, the conversion factor is  $0.045 \mu\text{R/h}$  per cpm. The pattern (b) in the figure shows near 30 min-exposures due to Ar-41 cloud from a 40 m stack of a research reactor under well weather condition, with the release rate of about  $3.0 \text{ Ci/s}$ . The diurnal variations of background radiation due to changes of Rn in air, are observed along with time by channel B. The differences of minimum to maximum are about  $0.6 \mu\text{R/h}$ . In this case, the separation of two components is very easy.

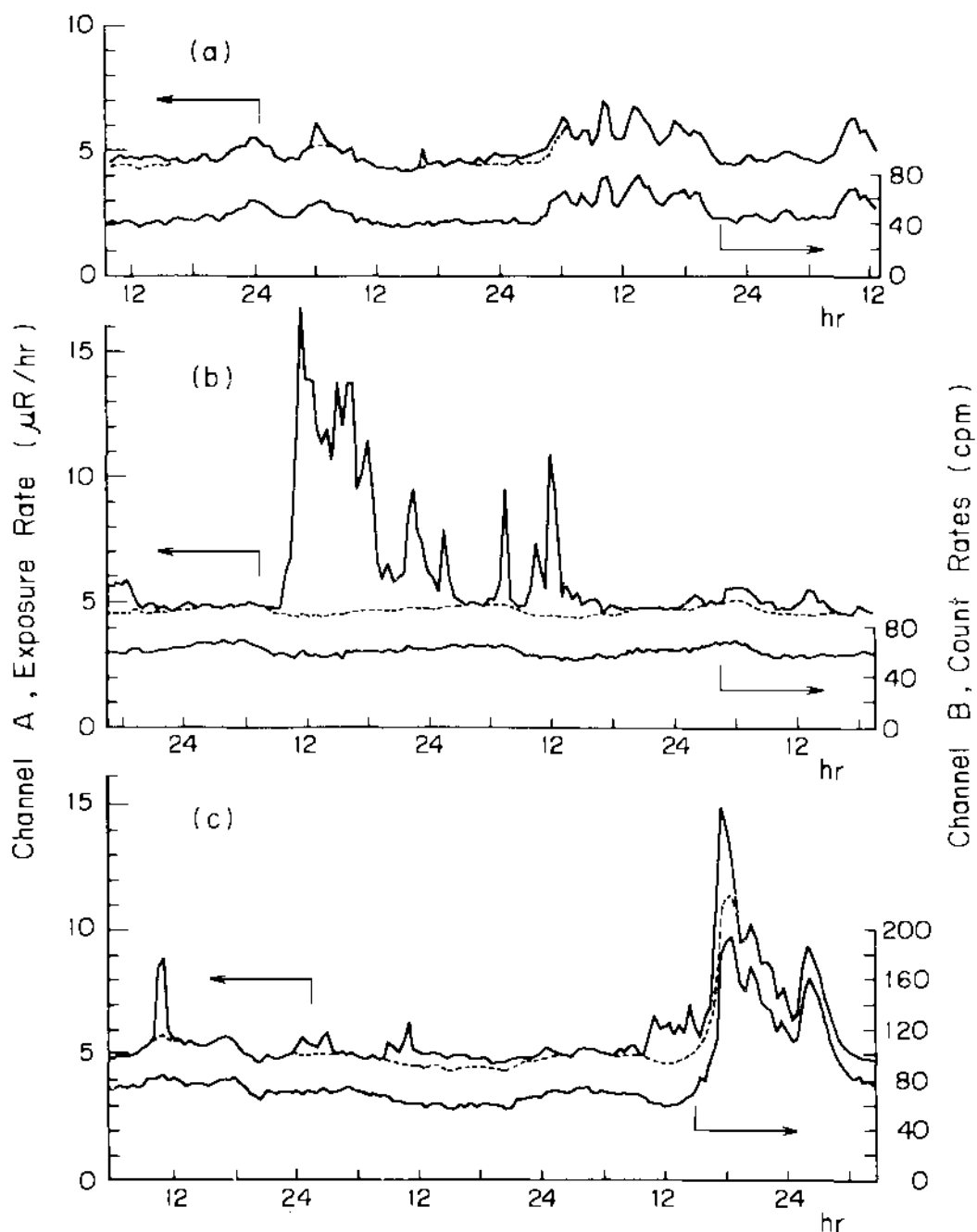


FIG.4. Records of exposure rate and SCA count rates in two channel system.

The contribution was evaluated to be  $97.7 \mu\text{R}$  from Ar-41 cloud. The pattern (c) in Fig.4 shows an example that the contributions both from JAPCO and from a Linear Accelerator are overlapped on continuing fine rains and succeeding heavy rains. In this case, the contributions from each radiation sources are  $24.4 \mu\text{R}$  due to plant operation and  $78.5 \mu\text{R}$  due to rainfall. These results show clearly that identification of radiation sources can be easily done without any other informations such as meteorological data and operating condition of plants.

#### Discussion and conclusion

By a long-run field test over a period of several months, our two channel system was proved to be very useful for discriminating the contribution from nuclear facilities even from short term fluctuations of background radiation, and was sufficiently stable in long field operation. But the two channel system will not be effective if maximum gamma ray energy from gaseous effluents and other radiation sources in interest exceeds the energy 1.76 MeV of RaC. The system is not, therefore, effective when facilities release large fractions of Xe-138, Kr-87 and Kr-88 in effluents. Recently, exposures from fallout due to nuclear weapon tests are fairly constant, since additional fallout deposition becomes very low and nuclides of rather long half-lives become predominant. Hence, the present status of fallout does not affect the operation characteristics of the system.

This system can be constructed easily by using a commercially available NaI(Tl) scintillation detector and NIM standard modules, but only a DBM module must be made individually.

The self-background pedestal of this system caused by K-40 contained in composing glass of a scintillator and a photomultiplier tube, appears only in channel A, and is less than  $0.2 \mu\text{R/h}$  equivalent. If ones of low potassium glass are selected, it can be reduced to less than  $0.05 \mu\text{R/h}$ . The contribution from cosmic ray is about  $0.6 \mu\text{R/h}$  equivalent in the case of the instrument of which the energy range flattened is up to 3 MeV, and if the pulses higher than 3 MeV is rejected by using an anti-coincidence circuit, its contribution can be reduced to  $0.2 \mu\text{R/h}$ . For example, standard error of counts in 10 minutes integral is  $\pm 0.06 \mu\text{R/h}$  in the case of a 2 x 2-inch NaI(Tl) scintillator and  $\pm 0.034 \mu\text{R/h}$  in the case of a 3 x 3-inch NaI(Tl) scintillator in radiation field of gamma radiation  $3.0 \mu\text{R/h}$  and cosmic ray  $3.4 \mu\text{R/h}$ . In addition, this instrument has another advantage that output data can be recorded and processed with a high accuracy because output signals are given as digital pulses.

Therefore, the minute rise of exposure rate as  $0.1 \mu\text{R/h}$  and integrated exposure less than 1 mR/yr due to gaseous effluents from a nuclear facility can be measured by using the two channel system of a NaI(Tl) scintillation exposure rate meter in spite of fluctuations of background radiation.

#### Acknowledgment

The authors wish to express their thanks to Mr. Hideo Kobayashi of the Environmental Survey Section for co-operation in the field work.

#### References

1. DeCampo, J.A., Beck, H.L., and Raft, P.D., USAEC Report HASL-260 (1972).
2. Beck, H.L., DeCampo, J.A., Gogolak, C.V., Lowder, W.M., McLaughlin, J.E., and Raft, P.D., Nuclear Technol., Vol.14 June, p.232 (1972).
3. Moriuchi, S., Miyanaga, I., Health Phys., 12, 541 (1966).
4. Miyanaga, I., Moriuchi, S., J. At. Energy Soc. Japan, 9, 8, 440 (1967).
5. Moriuchi, S., JAERI 1209 (1971).
6. Miyanaga, I., Moriuchi, S., NSF-Tr 137 (1968).
7. Moriuchi, S., Miyanaga, I., Health Phys., 12, 1481 (1966).
8. Moriuchi, S., Miyanaga, I., J. At. Energy Soc. Japan, 9, 9, 518 (1967).
9. Moriuchi, S., JAERI-X 5374 (1973).



## MONITORING TECHNIQUE FOR RADIOIODINE IN SODIUM AEROSOL

Nitsuo NARITOMI, Mikio MURATA and Yoshikazu YOSKIDA  
Japan Atomic Energy Research Institute,  
Tokai, Ibaraki, Japan

### Abstract

Radioiodine monitoring in a liquid-metal fast breeder reactor is different from that in the other types of reactor because radioiodine is always associated with the radioactive sodium aerosol. In this connection, experiments were made on behavior of the iodine and collection characteristics of an air sampler (JAERI type).

In the experiment, the fume of sodium containing sodium iodide was produced by vaporization of the liquid sodium, labelled with  $^{24}\text{Na}$  and  $^{131}\text{I}$ . It was then introduced through a fume transport tube into an ageing chamber with control of the temperature and relative humidity of air.

Characteristics of the airborne iodine in the sodium aerosol in the chamber were examined with May packs. About 97 % of iodine in the aerosol at room temperature is in particulate form (as sodium iodide), and percentage of gaseous iodine increases with rise of the fume temperature to about 30 % at 400°C.

More than 99 % of the iodine in the aerosol can be collected by a particulate and a charcoal filter paper in the sampler. In order to detect the radioiodine in sodium aerosol, however, the iodine should be collected separate from the sodium aerosol. For monitoring radioiodine in aerosol, a sampling technique with an arc discharge was contrived. It consists in the decomposition of aerosol, followed by the iodine sampling, the portion of sodium and of iodine thus separated are collected in the particulate and the charcoal filter paper, respectively.

### 1. Introduction

Monitoring of airborne radioiodine is important for radiation protection in a liquid metal fast breeder reactor as well as in the other types of reactor.<sup>1</sup> Air contamination due to radioiodine in a sodium cooled reactor, which is produced mainly by sodium pool fire or sodium spray fire<sup>2</sup>, is associated with the radioactive sodium aerosol because radioiodine released from the failed fuel is trapped effectively as sodium iodide in liquid sodium.<sup>3,4</sup>

There are several experiments made for the pool fire, which is caused by major sodium leakage. In this case, the ratio of I/Na in vapor and liquid is reported to be 0.3~1.0<sup>5-8</sup> and the iodine behaves as sodium iodide which can be effectively collected by a dust sampling filter paper such as cellulose asbestos filter paper. However, no experiments are made on the behavior of airborne radioiodine released by spray fire, in which sodium is introduced explosively into an oxidizing atmosphere by defect in primary system piping. It is considered that the characteristics of radioiodine in this case are different from those of a pool fire, because in spray fire, sodium is released into air in the form of fine particles of a high temperature and oxygen and moisture in

---

The work performed under the contract between Power Reactor and Nuclear Fuel Development Corporation and Japan Atomic Energy Research Institute.

the atmosphere is sufficient to oxidize the sodium. Air contamination in liquid cooled reactor may often be caused by this type of fire.

To establish the techniques of sampling the airborne radioiodine it is important to find the physical and chemical forms of airborne radioiodine when a small amount of sodium containing radioiodine is released into the air or inert atmosphere.

In the present study, assuming that radioiodine is trapped in the liquid sodium, the forms of airborne iodine were investigated by May pack, so named after its originator Fred May,<sup>9</sup> for the aerosol produced when Na-NaI was sprayed into the inert gas or air at various fume temperatures. And also collection performance of the JAERI-type air sampler for iodines was tested.

In this paper, the experimental results obtained are described and also discussed a new technique for sampling radioiodine separate from sodium aerosol.

## 2. Experimental

### 2.1 Generation of Na-NaI aerosol

A mixture of sodium (40g) and sodium iodide (40 μg of I), labelled with <sup>24</sup>Na (2 mCi) and <sup>131</sup>I (2 mCi), respectively, was prepared in a stainless steel can. The mixture in the can was melted in an electric induction furnace as shown in Fig. 1 which shows schematic diagram of the generator of sodium aerosol contaminated with iodine. In order to homogenize sodium iodide in the liquid sodium, the furnace was frequently vibrated at 350°C.

The temperature of liquid sodium was raised to 400°~550°C, with argon as the sweeping gas at 3 liter/min. Then, the temperature of sodium fume from the can was controlled to 25°~400°C by passing through the fume transport-tube with tape heater. The fume was sprayed into the air in a reaction cell with

control of the relative humidity at 25°C, and the produced aerosol was introduced into the chamber of volume about one cubic meter, at the same humidity, aged for 100 sec in it.

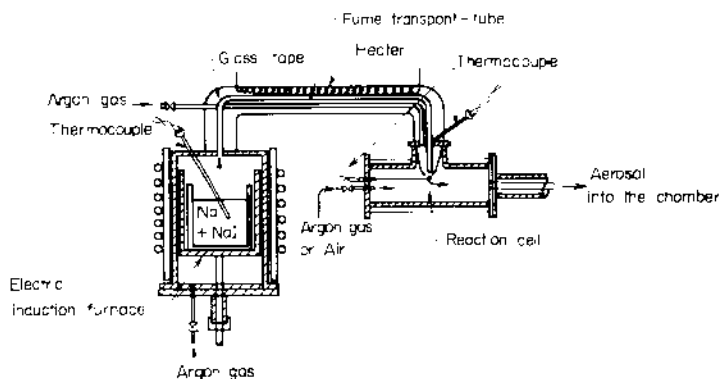


Fig.1 Aerosol generator

### 2.2 Identification of iodine form by May pack

The aerosol was sampled by two May packs placed in parallel at the chamber exit. The respective arrangements of components are shown

in Fig. 2, the components are listed below.

- 1) One Toyo HE-40 cellulose asbestos filter paper (particulate filter paper)
- 2) Four silver plated copper gauzes (80 mesh)
- 3) One Toyo CP-7 charcoal-loaded filter paper
- 4) Two 10 mm thick activated charcoal cartridges

Sampling was made for 30 min at the flow of 15 liter/min (face velocity 30 cm/sec).

The forms of iodine in sodium aerosol were estimated from the distribution of sodium and iodine collected in the components of May pack. The distribution was determined by measuring <sup>131</sup>I and <sup>24</sup>Na as the tracers. Radio-activities were measured by a γ-ray spectrometer with a 1-3/4"Øx2" NaI(Tl) well type detector connected to a 200 channel analyzer.

Identification of the iodine forms was made on the basis of collection performance of the May pack.<sup>10,11</sup> Iodine in particulate filter paper is particulate; in silver gauzes, elemental iodine; in charcoal filter paper,

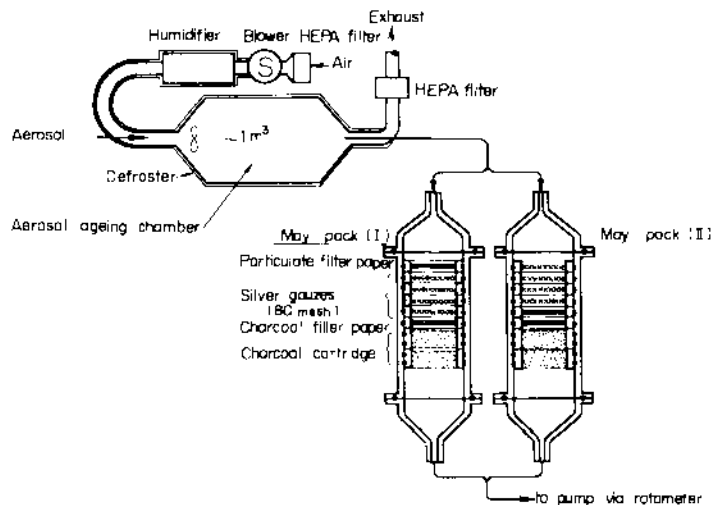


Fig.2 Apparatus for identifying iodine forms in sodium aerosol.

iodine compounds; and organic iodide such as methyl iodide is in the charcoal cartridge. In this method, it is necessary to determine the amount of gaseous iodine collected by sodium aerosol in the particulate filter paper. This was made by comparing the fractional distribution of iodine in the May pack when the gauzes were placed before the particulate filter paper and when placed after it.

### 3. Properties of iodine in sodium aerosol

#### 3.1 Iodine form in an inert gas

Sample was taken at the fume generator exit as shown in Fig.1, in the absence of air. Fig.3 shows the fractional distributions of sodium and iodine in components of May packs for the sodium fume in an inert atmosphere.

As seen in the figure, the iodine penetrating through the particulate filter paper is less than 0.1%, and the ratio of iodine to sodium fractions in the filter paper is the same for both the May packs and the iodine and sodium fractions in the four silver gauzes are the same in May pack (I). Therefore, the iodine form in sodium fume under an inert atmosphere is considered to be particulate such as Na-NaI particles. Fractional distributions of iodine and sodium at various temperatures of sodium fume were the same as shown in Fig.3. Most of the iodine (99.9%) is presumed to be particulate, independent of the temperature of sodium fume.

#### 3.2 Forms of iodine in the air

Fig.4 shows the fractional distributions of sodium and iodine in the components of May packs for the aerosol produced from the fume of 300°C. The fraction of iodine trapped in the particulate filter paper is the same in both the May packs, and also the fractional distributions of iodine in the four silver gauzes and in the charcoal filter paper were similarly the same. This shows that gaseous iodine, including elemental iodine, is not trapped in the filter paper loaded with a large quantity of sodium aerosol. Consequently, the forms of iodine and the respective percentages in the aerosol were estimated by the May pack (I).

Fig.5 shows the abundances of gaseous iodine in the sodium aerosol at the sodium fume temperatures of 25° to 400°C. The gaseous iodine increases with fume temperature except near 100°C, 37% at 400°C and 1.5~3% at 25°C.

The iodine trapped in the silver gauzes, charcoal filter paper and charcoal cartridges are approximately 30, 70 and 2% of the gaseous iodine, respectively, at all temperatures, except 100°C. The percentage of gaseous iodine at the fume temperature of 100°C was about 10 times as large as that on the "general" curves of abundance and temperature. However, the elemental iodine trapped in the silver gauzes was about only 3 times as large. It is thus seen that the abundance of less reactive iodine increases remarkably near 100°C.

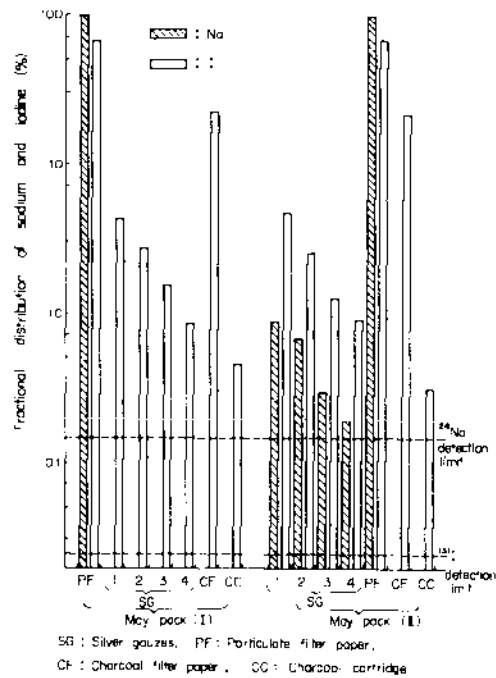
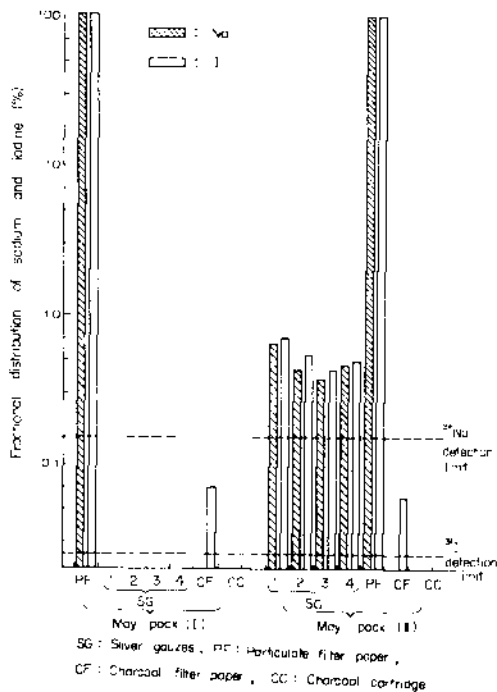


Fig.3 Fractional distribution of sodium and iodine (in inert atmosphere at 450°C)

Fig.4 Fractional distribution of sodium and iodine (in air atmosphere at 300°C)

The percentages of particulate and gaseous iodines under various conditions are shown in Table 1. It is seen from the table that abundance of gaseous iodine decreases with increase of the humidity of the air.

#### 4. Collection performance

The air sampler consists of a particulate filter paper and a charcoal filter paper in this order. The collection efficiencies of these two components were estimated for radioiodine in the sodium aerosol described in Sec.3. The results are shown in Table 2 together with the overall efficiency of the sampler.

In the sodium fume containing sodium iodide, sprayed into the inert atmosphere of 25°C, more than 99% of the total iodine is collected in the particulate filter paper together with the sodium, because radioiodine exists as Na-NaI particles.

When the sodium fume of 25°C is sprayed into the air, on the other hand, 3% and 1% of the iodine are trapped alone (i.e., with no sodium) in the charcoal filter paper at the air humidity of 40% and 80%, respectively. At the fume temperature of 300°C, the fraction of iodine collected in the charcoal filter paper increases to 29% and 5% at the humidity of 40% and 80%, respectively. The overall collection efficiency for iodine is more than 99% for both the inert and air atmospheres, but the amount of radioiodine is difficult to measure because of the large amount of radioactive sodium. The amount of gaseous iodine in the sodium aerosol can be estimated by measuring the activity in the charcoal filter paper, separate from that in the particulate filter paper. However, the total iodine in the aerosol is difficult to measure from the activity in the charcoal filter paper, because the fraction of gaseous iodine varies largely with temperature of the sodium fume and the air humidity.

In order to improve the detection sensitivity for the radioiodine in the radioactive sodium aerosol, the iodine should be collected separate from the sodium aerosol. This can be made by decomposition of the sodium iodide using an arc discharge<sup>12</sup> tube upstream the sampler. By the arc discharge technique,

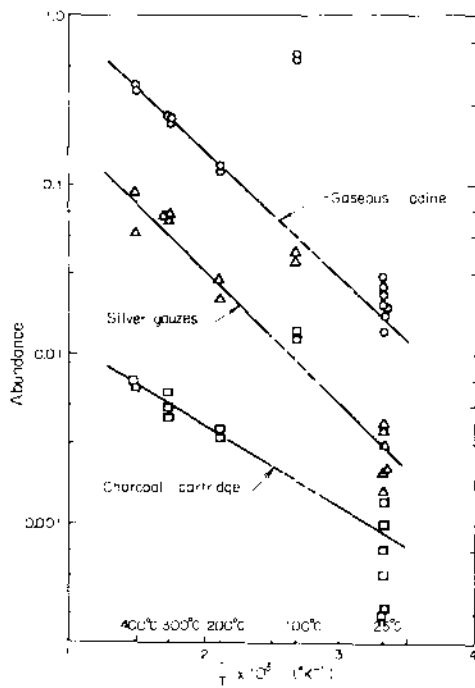


Fig.5 Effect of temperature of released sodium on abundance of gaseous iodines

Table 1 Percentage of particulate and gaseous iodines in the sodium aerosol

Atmosphere	Temperature of sodium fume (°C)	Relative humidity (%)	Run number	Percentage of particulate	Percentage of gaseous
Argon	25	dry	8	99.9	0.1
	300	dry	4	99.9	0.1
Air	25	40~95	2	98	2
	100	40	2	42	58
	200	40	2	88	12
		40~60	10	74	26
	300	80	4	95	5
		95	2	98	2
	400	40	2	63	37

80~90% of the iodine in the aerosol was decomposed into gaseous iodine, which can be collected efficiently in the charcoal filter paper. The collection efficiencies of the sampler using this technique are shown on the right side of Table 2. As seen in Table 2, 80~90% of the iodine is selectively collected in the charcoal filter paper from the sodium fume temperature and the air humidity. Thus, the concentration of total radiiodine in the radioactive sodium aerosol can be measured sensitively and accurately from the iodine activity collected in the charcoal filter paper.

Sodium iodide in the inert atmosphere, however, was not decomposed by arc discharge technique, as seen in Table 2. To use this technique, therefore, arc discharge must be made in air.

Table 2 Collection efficiency of the sampler for the radiiodine in sodium aerosol

Temperature of sodium fume (°C)	Atmosphere	Relative humidity (%)	Collection efficiency (%)					
			Without arc discharge Overall	With arc discharge charcoal filter paper	With arc discharge particulate filter paper	With arc discharge charcoal filter paper		
25~400	argon	dry	99	99	<1	99	99	<1
25	air	40	99	98	3	99	9	80
		80	99	98	1	99	11	88
300	air	40	99	70	29	99	22	77
		80	99	94	5	—	—	—

\* Sum of collector efficiencies for two filter papers

## 5. Conclusion

The forms of airborne iodine, when the sodium fume containing 1 ppm of sodium iodide is sprayed in the inert and air atmospheres, were examined experimentally. The results obtained are summarized as follows.

- 1) When the Na-NaI fume is sprayed into the inert atmosphere (Ar), no gaseous iodine is produced; the airborne iodine is considered as sodium iodide.
- 2) When sprayed into the air, the percentage of the gaseous iodine increases with increase of the fume temperature, about 30% at 400°C and 3% at 25°C at the air humidity of 40% and the percentage also depends on the humidity. The fraction of elemental iodine and inorganic iodide in the gaseous iodine are about 30% and 70%, respectively. At the fume temperature of near 100°C, however, the percentage of gaseous iodine increases to 60%, the forms are presumed to be different from those at other temperatures.
- 3) In monitoring radioactive iodine in the sodium aerosol such as in sodium coolant release from LMFBR, the sodium iodide with sodium aerosol is collected in the particulate filter paper and the gaseous iodine in the charcoal filter

paper. The percentage of iodine collected in the charcoal filter paper depends largely on the release conditions and atmospheric conditions. More than 99% of the iodine can be collected in the sampler consisting of the particulate and charcoal filter papers.

4) An approximately 90% of the iodine can be changed to the gaseous state by arc discharge technique, independent of the atmospheric conditions. The gaseous iodine is selectively collected in the charcoal filter paper from the sodium aerosol. Therefore, it is possible to measure airborne radiiodine accurately in the highly radioactive sodium aerosol, using the air sampler with the arc discharge tube.

#### Acknowledgement

The authors wish to gratefully acknowledge the collaboration of Mr. Hiroshi MATSUI, Mr. Yoshio IKEZAWA and Mr. Tsutomu OHATA of Health Physics and Safety Division of Japan Atomic Energy Research Institute. They are also indebted to Dr. Ichiro MIYANAGA, Chief of Division, for his useful suggestions.

#### References

1. W.F. Heins and R.S. Hart, Health Phys. 16, 449 (1971)
2. G.W. Keilholtz and G.C. Battle, ORNL-NSIC-37 (1969)
3. A.W. Castleman, ANL-7120, 848 (1965)
4. W.S. Clough, J. Nucl. Energy, 21, 225 (1967)
5. B.D. Pollock, M. Silberberg, and R.L. Koontz, ANL-7520, (1968)
6. A.W. Castleman, Nuclear Safety 11 379 (1970)
7. W.S. Clough, AERE-R-5848 (1970)
8. M. Murata, M. Naritomi and Y. Yoshida, (to be published)
9. W.J. Megaw and F.G. May, Jr. J. Nucl. Energy Part A and B (1963)
10. D.A. Collins, R. Taylor and W.D. Yuille, CONF-660904, 1096 (1967)
11. J.D. McCormack, BNWL-145 (1969)
12. M. Naritomi, M. Murata and Y. Yoshida, (to be published)

## LONG-TERM USE OF VARIOUS SOLID-STATE DOSIMETERS FOR ENVIRONMENTAL MONITORING OF NUCLEAR PLANTS - EXPERIENCE AND RESULTS

B. Burgkhardt, E. Piesch and M. Winter  
Health Physics Division  
Karlsruhe Nuclear Research Center, Fed. Rep. Germany

### Abstract

Solid-state dosimeters placed in the open air in the vicinity of the Karlsruhe Nuclear Research Center have been used to assess the natural radiation background and influences from radiation sources. The exposure periods of Phosphate glass, LiF, and CaF<sub>2</sub> dosimeters ranged from six months down to four days, depending on different monitoring problems.

### 1. Environmental Monitoring with Solid-state Dosimeters

At the Karlsruhe Nuclear Research Center accumulating solid-state dosimeters have been used since 1966 to determine the local distribution of the radiation dose in buildings and in the environment<sup>1,2</sup>. This kind of environmental monitoring should assess short-term and long-term variations of the radiation level encountered in the immediate vicinity of nuclear plants as a result of artificial radiation sources and emitters of radioactive materials. Moreover, such a network of measuring points allows the rapid evaluation of the dose received by the population after an accidental release of radioactive materials.

The network of measuring points equipped with dosimeters consists of five concentric circles with radiuses of 100 m, 250 m, 1 km, 2 km and 3 km, surrounding the stack of the FR 2 reactor, whose <sup>41</sup>Ar release entail a noticeable increase of the radiation background in the immediate neighborhood of the stack. In addition, dosimeters have been spaced at equal distances of 50 m along the site fence. Since 1967 the network of measuring points has consisted of about 250 measuring points in total, each of which equipped with two phosphate glass dosimeters; since 1969, 189 points have been additionally provided with LiF dosimeters. Further dosimeters have been installed at ten monitoring stations, which monitor continuously the radiation level in the surrounding villages up to 10 km distance from the center. These dosimeters are evaluated every six months. Moreover, CaF<sub>2</sub>:(nat) and CaF<sub>2</sub>:Dy dosimeters have been used since 1971 for short-term surveillance of the radiation level inside the Nuclear Research Center<sup>3</sup>. Moreover more than 3000 persons are monitored by phosphate glass dosimeters at the Nuclear Research Center so that even in the interior of the buildings comprehensive surveillance and balancing of the accidental radiation burden would be guaranteed.

### 2. Properties and Application of Dosimeters

- Depending on the application different types of dosimeters are used:
- Phosphate glass dosimeters in spherical capsules for long-term accumulation of the natural radiation background during a period of six months; within  $\pm 8\%$  energy-independent measurement of the  $\gamma$ -exposure for energies  $> 40$  keV
  - LiF dosimeters for long-term accumulation of the natural radiation background during a period of 3 months; within  $\pm 40\%$  energy-independent measurement of

$\gamma$ -exposures for energies > 15 keV and of  $\beta$ -radiation.

- CaF<sub>2</sub>:Dy dosimeters in spherical capsules for short-term accumulation of the natural or artificial radiation level during a period of 6 days; within  $\pm 25\%$  energy-independent measurement of  $\gamma$ -exposures for energies > 30 keV.

The dosimeters were sealed into plastic bags to protect them from weather influences. They have been installed on trees or aluminum tubes at about 3 m above ground level. A light proof packing for LiF dosimeters is obtained by sealing them in a black coloured 7 mg/cm<sup>2</sup> plastic foil, while the other dosimeters are protected from light by encapsulation. The fading of LiF and CaF<sub>2</sub>:Dy dosimeters is reduced by a thermal treatment (15 min at 100°C). In this case a correction compensating for fading after short-term exposure is not required.

TL-dosimeters must be regenerated before exposure (1 hour at 400°C, then 2 hours at 100°C). Besides regeneration the plotting of the glow curve is of essential importance in the

measurement of low doses, which allows to discover erroneous measurements. For the measurement of low doses with TL-dosimeters the accuracy is influenced mainly by the variation of the dark current of the multiplier and in the case of phosphate glass evaluation by dirtiness of the glass. A <sup>137</sup>Cs source was used for calibration of the dosimeters and the readers.

Tab.1: Average annual dose values measured outside the Karlsruhe Nuclear Research Centre

Exposure Period <sup>+) </sup>	AVERAGE ANNUAL DOSE IN mR			
	1 km-Circle Glass	2 km-Circle Glass	3 km-Circle Glass	LiF
1966/67	61	67	52	-
1967/68	93	93	86	-
1968/69	38	38	42	-
1969/70	34	36	47	-
1970/71	48	52	55	72
1971/72	64	56	57	70
1972/73	57	62	67	72
mean value	56	58	58	71

<sup>+)</sup>  from May to May

### 3. Monitoring of the Natural Radiation Background

#### 3.1 Long-term Exposure

Table 1 shows the average values of the annual dose determined with glass and LiF dosimeters for the concentric circles surrounding the FR 2 stack at distances of 1, 2 and 3 km. The annual dose of the natural radiation background averaged over several years is about 58 mR/a for glass dosimeters and about 70 mR/a for thermoluminescence dosimeters. By using phosphate glasses the natural radiation background - mainly due to <sup>226</sup>Ra and cosmic rays - will be underestimated in the order of 10 mR compared to the LiF results.

#### 3.2 Short-term Exposure

When CaF<sub>2</sub> dosimeters are used a 7 days exposure period is adequate to measure the natural radiation background. To assess the accuracy of the measurement dosimeters were evaluated during a two months period both after one week and two months of exposure. The width of frequency distribution of the weekly dose readings shown in Fig.1 depends on the local and time variation of the natural radiation background including measuring errors and fading. The radiation level measured with CaF<sub>2</sub> dosimeters and a scintillation counter corresponds to an exposure rate of 9.1  $\mu$ R/h and 8.5  $\mu$ R/h, respectively. The internal natural radiation of the CaF<sub>2</sub> material might result in an increase by about 0.5  $\mu$ R/h in the dosimeter reading.

To determine the influence of fading on the reading of CaF<sub>2</sub> dosimeters a comparison is made between the results of two months dose and the weekly dose totaled up over the same period for an exposure within the sky shine radiation field of the waste storage (cf. Fig.2). The fading of CaF<sub>2</sub> dosimeters of about 3% after two months is comparable with the evaluation error of one dosimeter only. Compared to LiF, the dose reading of CaF<sub>2</sub> dosimeters was found to be too



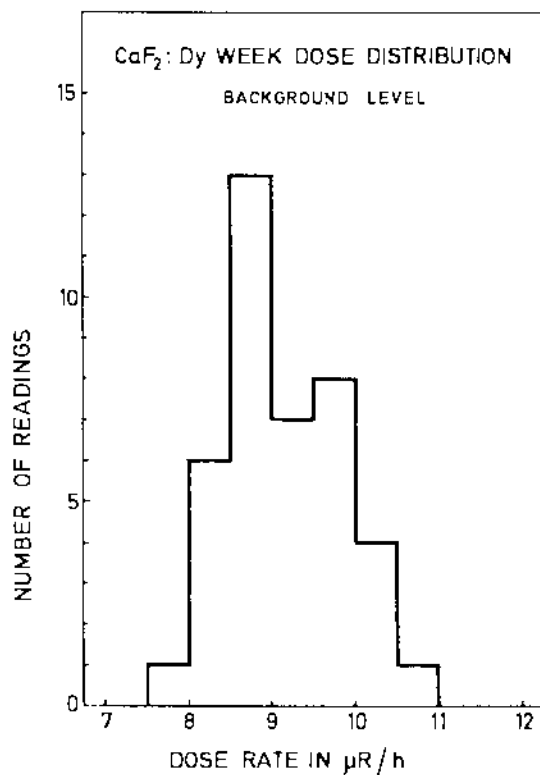


Fig.1: Frequency distribution of the week dose

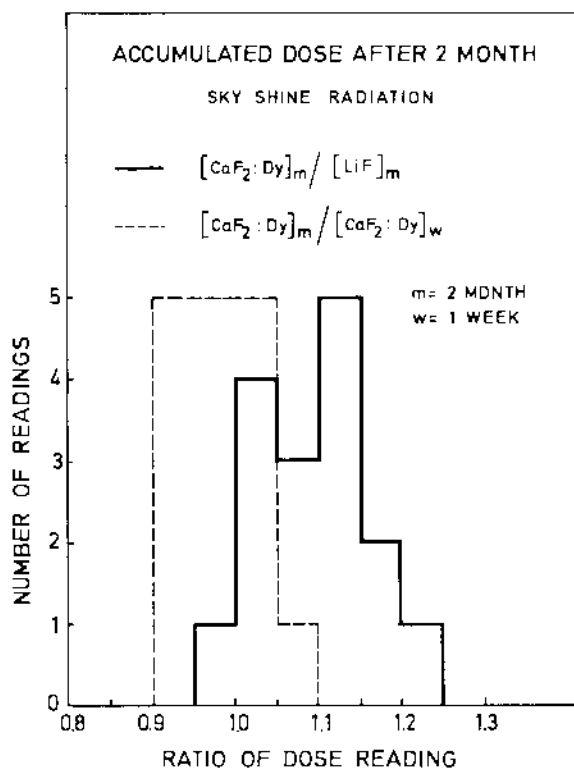


Fig.2: Frequency distribution of the dose reading ratio

high by 15 %; this had to be anticipated on account of the well known energy dependence in the energy range of 100 keV.

### 3.3 Comparison of Dose Readings for Different Exposure Periods

The annual doses represented in Table 2 were measured or extrapolated over exposure periods of 12 months, 6 months, 2 months and 1 week, respectively, using different solid-state dosimeters at six locations. The differences of the dosimeter readings are mainly due to differences in the measuring accuracy and in the fading of the different types of dosimeters. For the glass dosimeter the

Tab.2: Measurement of the Natural Background Radiation with Solid-State Dosimeters

RELATIVE DOSE READING IN I							
DOSIMETER	PERIOD OF EXPOSURE	1	2*)	3	4	5*)	6
GLASS	annual dose over 2 years (=100%)	68 mR	70 mR	68 mR	71 mR	64 mR	74 mR
	1 year	140	136	125	120	145	132
	6 months	162	114	132	127	164	162
LiF	2 years	109	119	104	99	108	95
	6 months	109	123	111	115	125	111
	2 months	112	121	90	99	127	103
CaF <sub>2</sub> :Dy	2 months	88	119	85	85	100	77
	1 week	122	157	112	118	125	107
	(average value for 2 months)	±11.5	±15.5	± 13	±14.8	±14.5	± 7

\*) radiation level due to the influence of nuclear plants (β-radiation)

influence of fading is less than 10 % over ten years. On the other hand, CaF<sub>2</sub> shows a temperature-dependent fading which even for a long-term exposure can be corrected only to a limited extent by simultaneous exposure of additional calibration dosimeters. Thus, for instance, an increase in temperature within the first or last week of a long-term exposure will cause an extremely different influence of fading. A difference of 20 % was found for CaF<sub>2</sub> after an accumulation of the natural radiation background over a period of two months and one week, respectively. Further investigations will show whether this difference in the dose reading must be attributed to an increased fading for low radiation levels.

Based on an extensive calibration technique the natural radiation background of about 10 µR/h can be measured with a CaF<sub>2</sub>:Dy dosimeter with an accuracy of ± 10 % already at the end of six days of exposure period. With an LiF dosimeter an adequate accuracy can be achieved after a period of three months and with a glass dosimeter after a period of 12 months. Contrary to LiF, glass dosimeters are not sensitive to β-rays. However the handling, reading and calibration is simpler and no regeneration is required.

#### 4. Monitoring of Increased Radiation Levels in the Immediate Neighborhood of Nuclear Plants

##### 4.1 <sup>41</sup>Ar Emission by the FR 2 Reactor

The increase in the radiation level at distances of 100 and 250 m from the FR 2 exhaust stack is indicated in Table 3. The influence of <sup>41</sup>Ar emission takes an average dose reading of about 60 mR/a at a distance of 100 m. Glass dosimeters are also exposed directly in the exhaust stack to monitor the <sup>41</sup>Ar emission.

Tab.3: Average annual dose values measured within the Karlsruhe Nuclear Research Centre at different distances from the FR 2 Stack

Exposure Period *)	AVERAGE ANNUAL DOSE IN mR							
	30 m		100 m		250 m		along the site fence 0.4-1 km	
	Glass	LiF	Glass	LiF	Glass	LiF	Glass	LiF
1966/67	-	-	-	-	-	-	74	-
1967/68	110	-	94	-	89	-	102	-
1968/69	126	-	118	-	116	-	51	-
1969/70	87	123	98	131	95	142	58	76
1970/71	112	146	118	161	120	176	67	81
1971/72	193	164	140	142	105	103	74	69
1972/73**	339	380	182	223	104	130	66	87

\*) from May to May

\*\*), Special influences from the Decontamination Plant in this period

##### 4.2 Local Dose Distribution at the Radioactive Waste Storage

In the immediate neighborhood of the radioactive waste storage LiF and CaF<sub>2</sub> dosimeters were exposed over two months at 8 locations spaced over a length of 750 m. The dose reading profile is indicated in Fig.3. The increase in the radiation level due to the waste storage could be determined here with an accuracy of 10 % after an exposure period of one day already, using CaF<sub>2</sub> dosimeters.

##### 4.3 Variations in Time of the Radiation Level in the Region of the Decontamination Facilities

Outside of the buildings housing the decontamination facilities for solid and liquid waste the variation in time of the radiation level is monitored by CaF<sub>2</sub> dosimeters at 16 measuring points after an exposure period of one week each. The examples in Fig. 4 demonstrate the different increase of the weekly dose readings over two months compared with a continuous increase assuming a

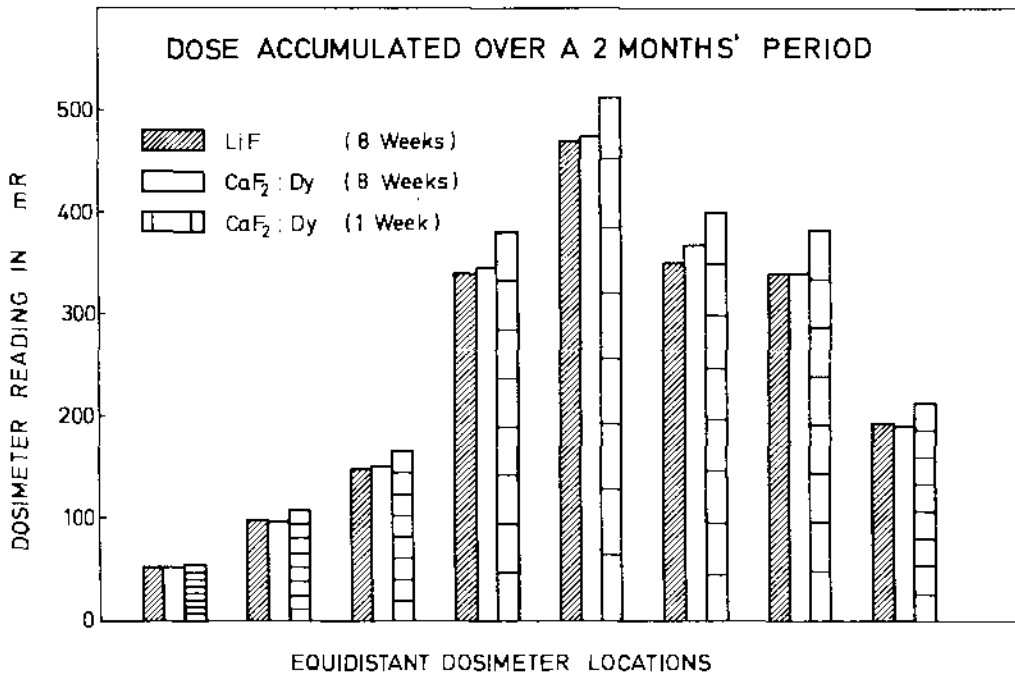


Fig.3: Dose profile in the closed vicinity of the waste storage

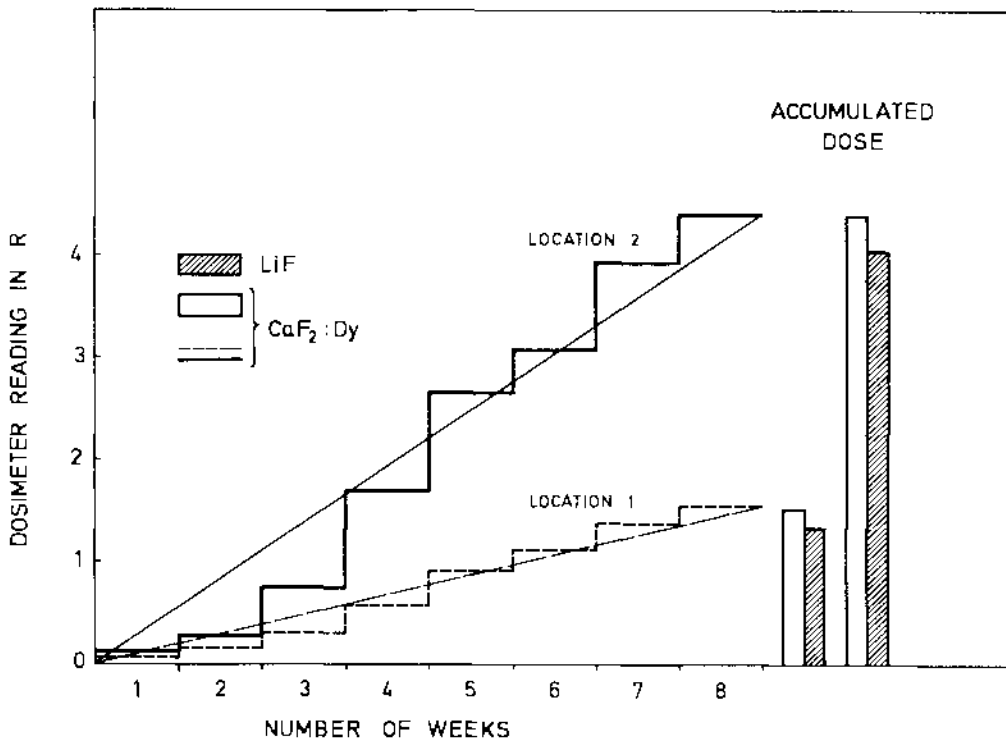


Fig.4: Dosimeter readings evaluated weekly compared with the total dose reading after 8 weeks

constant radiation level. The readings of additional dosimeters after an exposure period of 8 weeks are also included in Fig. 4. The importance of short-term monitoring lies in the detection of increased dose rate levels outside of the facilities caused by operational reason, which allows to initiate in time the radiation protection measures required.

### 5. Comparison with Dose Rate Meters

For direct and continuous monitoring of the  $\gamma$ -dose rate level GM-counters with energy compensation filters have been installed at 30 locations of the Karlsruhe Nuclear Research Center. The count rates, transferred by cable, are indicated in a central station (within  $\pm 30\%$  energy-independent dose rate indication for energies  $> 25 \text{ keV}^5$ ). Graphic integration of the dose rate plotted on recorders was performed for two locations over a two months period. The graphically determined results were compared with the results measured by  $\text{CaF}_2$  and  $\text{LiF}$  dosimeters, respectively, exposed at the same locations (cf. Table 4). The table includes the reading of a sensitive scintillation counter whose dose rate indication for energies  $> 25 \text{ keV}$  is energy-independent within  $\pm 10\%$ . For both locations and radiation levels an agreement was found between the results of solid-state dosimeters and dose rate meters which is more than sufficient for monitoring purposes.

Tab.4: Comparison of results measured with dose rate meters and dosimeters

ACCUMULATED DOSE IN mR AFTER 2 MONTHS <sup>1)</sup>		
Detector	Location 1 (200 $\mu\text{R/h}$ )	Location 2 (-8 $\mu\text{R/h}$ )
BZ 120 counter	401 (108%)	16.9 <sup>++)</sup> (115%)
Scintillation counter <sup>3)</sup>	-	11.5 (79%)
$\text{LiF}$	370 (100%)	14.6 (100%)
$\text{CaF}_2:\text{Dy}$ <sup>4)</sup>	409 (111%)	15.2 (104%)
$\text{CaF}_2:\text{Dy}$	381 (103%)	10.9 (75%)

+) short-time dose rate measurement  
 ++) sum of weekly accumulated doses  
 +++) GM counter without energy compensation filter 330 c/min = 10  $\mu\text{R/h}$

### References

- <sup>1</sup> Maushart, R., Piesch, E., Winter, M.: Report KFK-551, 1967
- <sup>2</sup> Winter, M.: Proc. of IAEA-Symp. on Rapid Meth. Environment, p. 525, 1971
- <sup>3</sup> Burgkhardt, B., Piesch, E.: Kerntechnik 14, 128, 1972
- <sup>4</sup> Günther, G., König, L.A.: Atompraxis 14, Heft 8, 1968
- <sup>5</sup> Piesch, E.: Kerntechnik 9, 198, 1967
- <sup>6</sup> Kolb, W., Lauterbach, U.: Proc. of IAEA-Symp. on Rapid Meth. Environment, p. 565, 1971

INVESTIGATION OF THE TRITIUM LEVEL IN THE ENVIRONMENT OF  
THE KARLSRUHE NUCLEAR RESEARCH CENTER

L.A. König and M. Winter  
Health Physics Division  
Karlsruhe Nuclear Research Center, Fed. Rep. Germany

Abstract

In the immediate and more distant neighborhood of the Karlsruhe Nuclear Research Center a comprehensive tritium monitoring program has been carried out. The results obtained in the measurement of tritium concentration of precipitations are characterized by strong seasonal variation. By contrast, the measured values for drinking water only show statistical scattering. The values of tritium concentration in surface water are on the average slightly higher than the values for drinking water. The tritium radiation burden to the population is estimated to 0.1 mrem/a on the basis of the measured values available.

1. Introduction

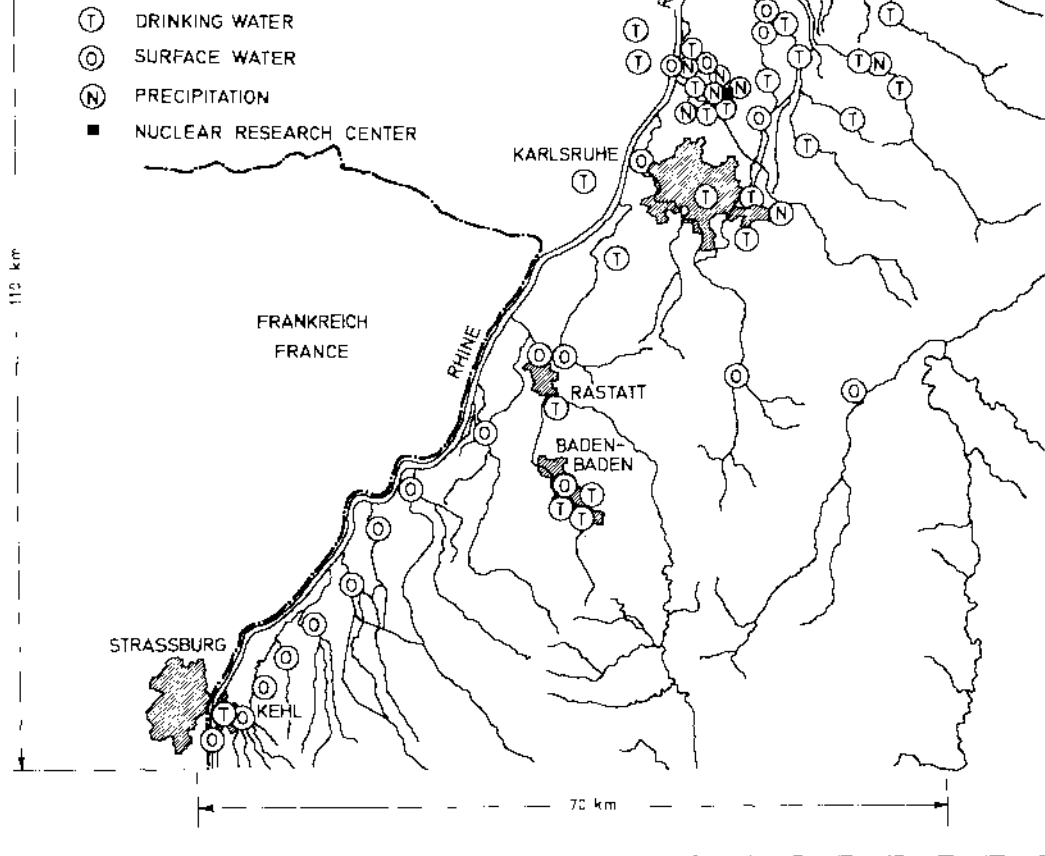
In the immediate and more distant neighborhood of the Karlsruhe Nuclear Research Center a comprehensive tritium monitoring program<sup>†</sup> has been carried out to determine the tritium content in the ground, drinking and surface water as well as in precipitations. These measurements have been initiated by tritium released from the two heavy water reactors FR 2 (44 MW<sub>th</sub>) and MZFR (200 MW<sub>th</sub>) and from a reprocessing plant. The amounts released every year from the two reactors amount to 0.5% from the inventory [1]. Since the tritium inventory will increase by about 1 Ci/l per year a considerable rise of the amounts so released must be anticipated. The same is true for the amounts discharged by the reprocessing plant which in future will have to process both greater amounts of fuel and fuel with higher burnups. The overall tritium release via the effluent air and water presently amounts to about 4000 Ci/a [2]. The portion of tritium discharged with waste water is steadily increasing and is now already more than half of the total discharges, while in 1971 it was still at nearly 20%. On account of the continuously rising tritium concentration in waste water, it is intended to feed waste water with heavy tritium contents into depleted oil wells for final disposal, which are located close to the Nuclear Research Center and from which approximately 20 000 m<sup>3</sup> of an oil-water mixture have been pumped off so far at a depth of roughly 1000 m. The implementation of this project also calls for a very careful environmental monitoring for tritium to be able to demonstrate the reliability of this type of tritium disposal. Finally, a third reason for the performance of a tritium measuring program was the interest in ecological questions. Since tritium can be detected at relatively little expenditure, in most of the water samples of the environment, systematic tritium measurements extended over a longer period of time open up the possibility of investigating questions related to the water cycle.

---

<sup>†</sup> Part of a co-ordinated program of research under the sponsorship of the IAEA.

Fig.1. MAP OF SAMPLING LOCATIONS

TYPE OF SAMPLE	NUMBER OF SAMPLING LOCATIONS	FREQUENCY OF SAMPLING	NUMBER OF SAMPLING LOCATIONS WITH AN AVERAGE ANNUAL TRITIUM CONCENTRATION IN pCi/ml OF						
			< 0.3	0.3-0.5	0.5-0.7	0.7-1.0	1.0-1.5	> 1.5	
DRINKING WATER	28	EVERY FORTNIGHT	16	7	5	0	0	0	
SURFACE WATER	31	WEEKLY AND BI-WEEKLY RESP	8	15	6	1	1	0	
PRECIPITATION	8	WEEKLY AND BI-WEEKLY RESP	0	0	2	3	2	1	



## 2. Measuring Program and Measuring Procedure

The current sampling program covers the Upper Rhine Valley region between Mannheim and Straßburg. Fig. 1 is a site plan of the sampling locations. As of late 1972 11 samples of precipitation, 28 samples of drinking water and 46 samples of surface water were measured within a fortnight each. Within the framework of this program both the river Rhine and its most significant affluents on the right side are being monitored. The drinking water samples are taken by the staff of the department at their places of residence and brought for measurement. Sampling of precipitations has been restricted to 10 km round the Nuclear Research Center, since the management required of the collecting vessels does not allow distribution over larger distances.



In 1972 about 1600 water samples were investigated in total within the framework of this program.

The tritium concentration of the water samples is determined with the help of a liquid scintillation spectrometer Mod. 3375 of Messrs. Packard Instr. The quench correction used in this equipment is based on a combination of the so-called channel ratio method and the method of the external standardisation. The three-component scintillator 'Insta-Gel' used is available as a ready-to-use mixture. All water samples are distilled before the measurement. 10 ml water of each sample are added to 12 ml 'Insta-Gel'. This work is done in a dark chamber to avoid photoluminescence. Also subsequent reheating of the samples over 2 hours at about 70 °C helps to avoid thermoluminescence and photoluminescence effects. When the samples have cooled to the measuring temperature in the liquid scintillation spectrometer at 11 °C the samples are measured for 50 minutes.

With setting of the measuring channel which is optimum for mere tritium measurements, a background of 4 to 5 cpm and counting efficiencies of 14 to 18%, depending on the degree of quenching, detection limits between 0.2 to 0.3 pCi/ml can be obtained for the tritium concentration  $\pm 3\%$ .

The following relation holds for the relative  $2\sigma$ -error of the tritium concentrations measured:

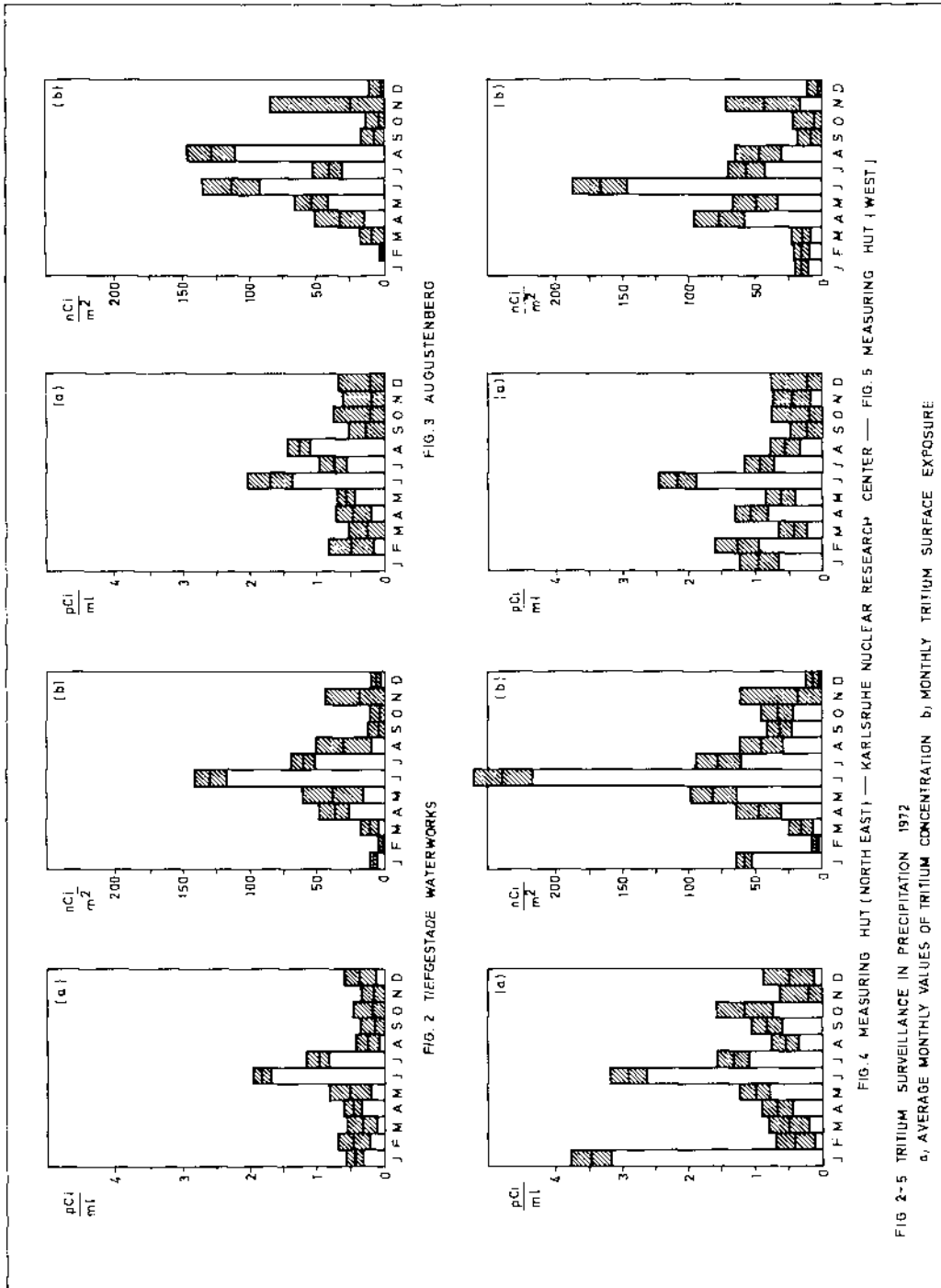
$$\frac{\Delta a}{a} = \pm \sqrt{\frac{4(\lambda+1)}{R_0 T_M (\lambda-1)^2} + 0.0007} \quad \text{with } \lambda = \frac{R_M}{R_0},$$

where  $R_M$  and  $R_0$  are the measuring and background rate, respectively, and  $T_M$  is the time of measurement. The constant under the root is composed of the errors of the calibration standard and the amount of the material to be measured. The shaded zones in the graphic representations of the monthly average values represent the measuring errors and in addition the uncertainty obtained in the averaging process from the treatment of measuring values below the detection limit. For measuring values below the detection limit 0 or the values of the detection limit itself can be alternatively used in the averaging process. The uncertainty of the statement resulting from this in addition to the measuring errors is included in the shaded zones.

### 3. Measuring Results 1972

The results of precipitation measurements are represented in Figs. 2 to 5. The determination included both the monthly amounts of precipitation weighted with the concentration averages and the tritium activity introduced every month into the surface unit of the soil. The differences of monthly concentration mean values between the collecting points are clearly visible. While the more distant collecting points Augustenberg and Tiefgestade waterworks show a marked concentration maximum in June, the measuring point located in the north-east of the Nuclear Research Center is characterized also by a secondary maximum in January and another in October. This obviously involves tritium immissions caused by discharges. Differences in the area burdens are conditioned by differences in concentration and above all by different amounts of precipitation.

Examples of measuring results from surface waters are shown in Figs. 6 to 11. It appears immediately that the tritium level is higher in the river Rhine compared to the smaller surface waters monitored. A definite explanation has not yet been found for this phenomenon. A major cause of this effect might be that in the small waters the portion of waste water from residential areas (mostly ground water poor in tritium) is greater than in the Rhine itself. Also in the Rhine the tritium concentration in the middle of the year is lower than



in the precipitation. The peak values of tritium concentration recognizable in Figs. 10 and 11 are obviously due to discharges from the Nuclear Research Center into these two waters. The relationship is especially evident in the case of the ancient Rhine (Fig. 11) which takes up all waste waters from the Nuclear Research Center, while the diagram shown in Fig. 10 reminds of the tritium concentration detected in the precipitation at the measuring hut north-east of the Center.

The variation in time of the tritium concentration in surface water is remarkable; it corresponds to the variation of tritium concentration of the



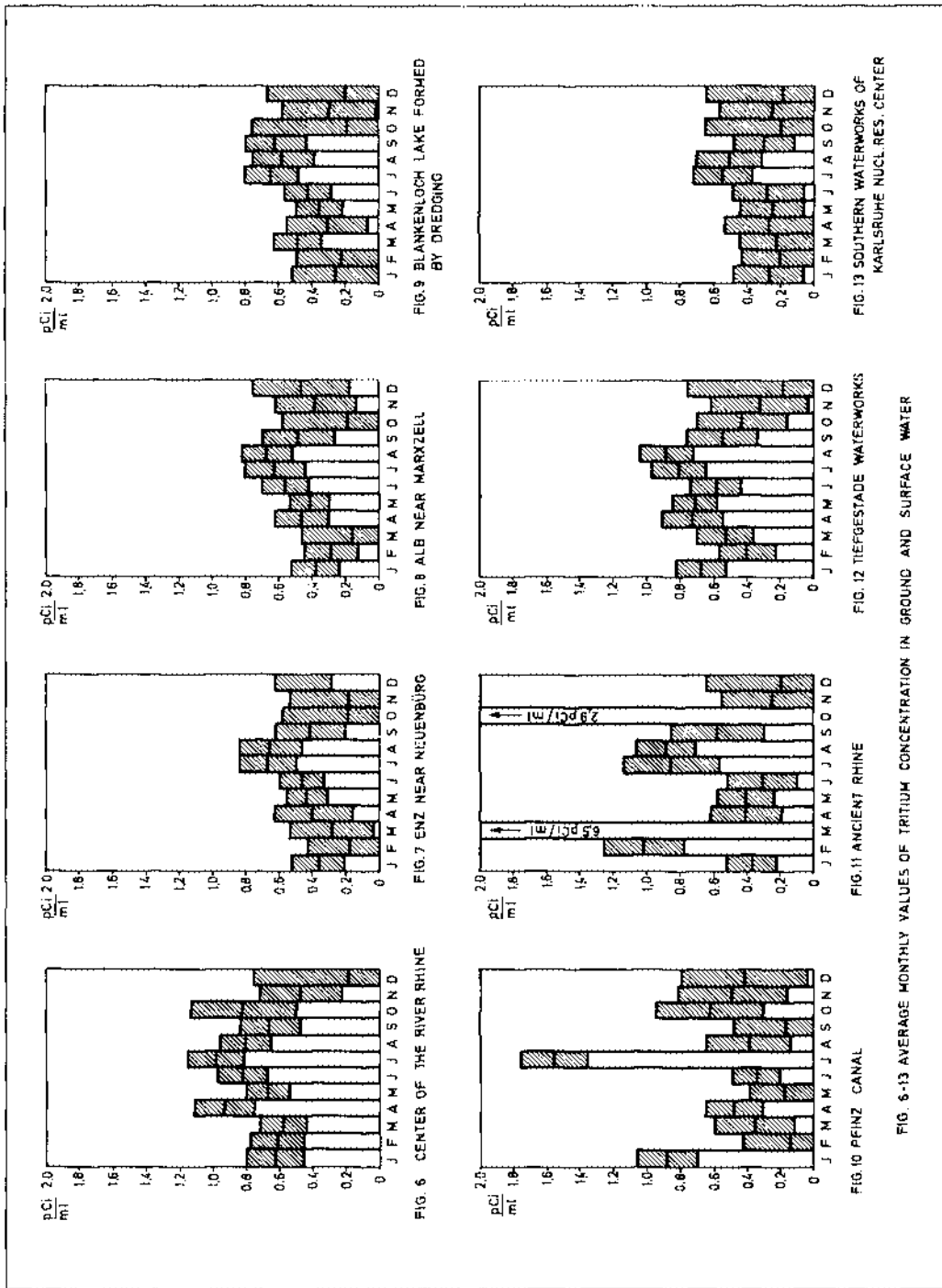


FIG. 6-13 AVERAGE MONTHLY VALUES OF TRITIUM CONCENTRATION IN GROUND AND SURFACE WATER

precipitation with an expected time lag.

In Figs. 12 and 13 the ground water activities are shown for two sampling points. The Tieggestade waterworks of the Karlsruhe Nuclear Research Center is located in close vicinity of the river Rhine. Here the concentration values are slightly inferior to that in the Rhine water. However, the values determined for the southern waterworks of the Nuclear Research Center are markedly inferior. The comparison of the Figs. 6, 12 and 13 indicates that the major part of the drinking water from the Tieggestade waterworks is Rhine water filtered on the border. It must be expected that this portion is subject to variations

in time. Among others also a dependence on the amount of water carried by the Rhine is conceivable. According to an estimate the water taken from this waterworks should contain an average of about 50% of Rhine water.

Fig. 1 contains a table stating the nature of the samples, the number of sampling points, and the frequency of sampling. The frequency distributions indicated in this table for three kinds of water with annual averages of the tritium concentration, which are identical within certain intervals of measuring values, makes evident the different tritium concentrations of drinking water, surface water and precipitation.

On the assumption that the tritium concentration in the human body corresponds on an average to the tritium concentration of drinking water, an estimate value for the radiation burden of 0.07 mR/a is obtained for concentrations as encountered in the drinking water from the southern waterworks of the Nuclear Research Center. A radiation burden in the range of 0.1 mR/a would have to be assigned to drinking water mainly taken from surface waters. Since it must be assumed that the tritium concentration in plants exposed to the free atmosphere is by no means lower than in surface waters, the water uptake from vegetable food should not lead to a reduction of the tritium concentration in the human body so that the estimated values indicated above must be considered to be realistic. As a matter of fact they agree with the values given in [4].

#### References

- [1] L.A. König, Umweltbelastung durch Tritium, Report KFK 1560 (1972)
- [2] L.A. König, M. Winter, J. Amann, H. Schüler, Contributions to Annual Report 1972 of the Health Physics Division, Report KFK 1818 (1973), edited by H. Kiefer and W. Koelzer
- [3] L.A. König, M. Winter, Über die Tritiumkontamination der Umwelt, Symposium on Radioecology ..., Rome, Sept. 7-10 (1971), EUR 4800
- [4] E. Fischer, S. Mlinko, T. Szarvas, Bestimmung des natürlichen Tritiumgehalts von Lebensmitteln, GDCH-Meeting on Spurenanalyse (Tracer Analysis), Erlangen, April 2-5, 1973

ANALYSIS OF AN ANALYTICAL TECHNIQUE FOR  
DISTRIBUTING AIR SAMPLING LOCATIONS AROUND NUCLEAR FACILITIES

David A. Waite  
Battelle  
Pacific Northwest Laboratories  
Richland, Washington 99352

Abstract

A new analytical mechanism for distributing air sampling locations around nuclear facilities, including reactors, fuel fabrication, fuel reprocessing and research centers, has been devised. This method was developed to facilitate the efficient incorporation of past experiences into environmental surveillance programs for new installations. The technique provides an initial distribution of air samplers around a site which correlates well with placements on sites which have been occupied by long-established programs with their inherent evolution and refinements. Sensitivities of calculated sampler distributions to various meteorologic and demographic input parameters will be discussed. The applicability of this approach has been examined by comparing sampling locations in well-established air monitoring programs scattered worldwide with location distributions recommended by this mechanism. Results of these comparisons will be detailed.

Introduction

In recent years public pressure has compelled an increase in attention to environmental surveillance around industrial complexes of all types. Information involving probabilities of detecting short duration releases in the environment have been widely circulated. It is seldom noted, however, that in contrast to emergency monitoring, the worth of systems for routine environmental surveillance is judged on the basis of long-term productivity. Even though this fundamental difference exists between monitoring accident and routine conditions, the publicity in some cases has had an adverse influence on industrial confidence in data obtainable from routine surveillance programs. This lack of confidence is reflected in some environmental surveillance programs.

An examination of existing programs indicates a need for the development of surveillance system design techniques which can be easily and economically applied by environmental surveillance personnel to yield sampling location distributions which correlate well with placements on sites which have been occupied by long-established programs. Even though there are recognized techniques for designing surveillance systems around nuclear installations,<sup>1,2</sup> there is little evidence to show that these are being widely applied by those responsible for operating the programs. These more sophisticated design methods are generally regarded as expensive to utilize.

In an attempt to provide a consistent, uncomplicated, economical mechanism for the examination of surveillance systems, an evaluation was begun of variables which influence a program's ability to assess population exposures and which could be incorporated into a workable sampling location distribution procedure. Acceptability of the procedure would be based, it was decided, upon: (a) its ability to provide an initial distribution of air samples around a site which correlated well with placements on sites which have been occupied by long established programs with their inherent evolution and refinements, and (b) respectable agreement between distributions derived by well-recognized techniques and this method.

An examination of parameters involved in release mechanisms, transport models and exposure pathways for airborne releases from nuclear fuel cycle facilities, revealed that population exposure magnitudes were most directly related to population distribution and contaminant transport characteristics of the local environment involved.

### Procedure

Examination of existing surveillance programs indicated that distances of environmental media sampling locations from the source of contamination are generally selected to correspond to plant boundaries, maximum potential concentration points, population centers and relatively unaffected areas and that the most needed guidance was in the radial partitioning of sampling locations. The relationship,

$$\text{Weighting Factor (W)} = \frac{\text{fraction of total population}}{\text{distance}} + \frac{\text{fraction of time sector is downwind of sources,}}{\text{distance}}$$

associating demographic and meteorological variables was found to yield useful recommendations when applied to a site on an octant basis.

The application of this method to sites in coastal or agricultural areas requires only minor modification of the basic procedure.<sup>3</sup> In coastal zones it is usually appropriate to adjust the number of radial divisions to the number required to cover the surrounding inhabited land mass. For agricultural areas an equivalent population index is necessarily applied. This index is derived by multiplying the number of people who are direct recipients of produce, dairy products, etc., from the area by the biological discrimination factor for the critical nuclide in the exposure pathway involved.

The sum of eight calculated weighting factors is scaled to equal the desired number of sampling locations. The scaling factor, when multiplied by each octant weighting factor, yields the number of sampling locations within that octant when rounded to the nearest integer. For an octant which is downwind of the source twenty percent of the time and has fifty percent of the regional population at fifteen miles from the site, the calculation of its portion of a seventeen sampling location program would go as follows:

$$W = \frac{.50}{15} + .2 = .23$$

Assuming the sum of eight weighting factors is 1.0, the scaling factor (SF) is  $17/1.0 = 17$ . Number of locations per octant =  $W \times SF = 3.97$  which would be rounded to 4 to indicate the number of locations to be allotted to that sector.

### Analysis

To test acceptability of the method on the basis of its ability to provide distributions which correlate well with placements on sites with well-established programs, eleven mature sites were selected as models for testing. The sites were located in the United States, India, Japan, and Germany. The facilities included power reactors, uranium enrichment facilities, fuel fabrication plants and nuclear research establishments. Sampling locations per site varied from eight to forty. Results of sampling location distribution comparisons are shown in Figure I below. The deviation noted on the abscissa is the difference between the calculated and actual number of sampling locations present per octant. It should be noted that over thirty percent of the octants had zero deviations. Most of the greater deviations can be attributed to the relatively arbitrary divisions formed by the octant boundaries and to criteria applied in the rounding off of calculated values.

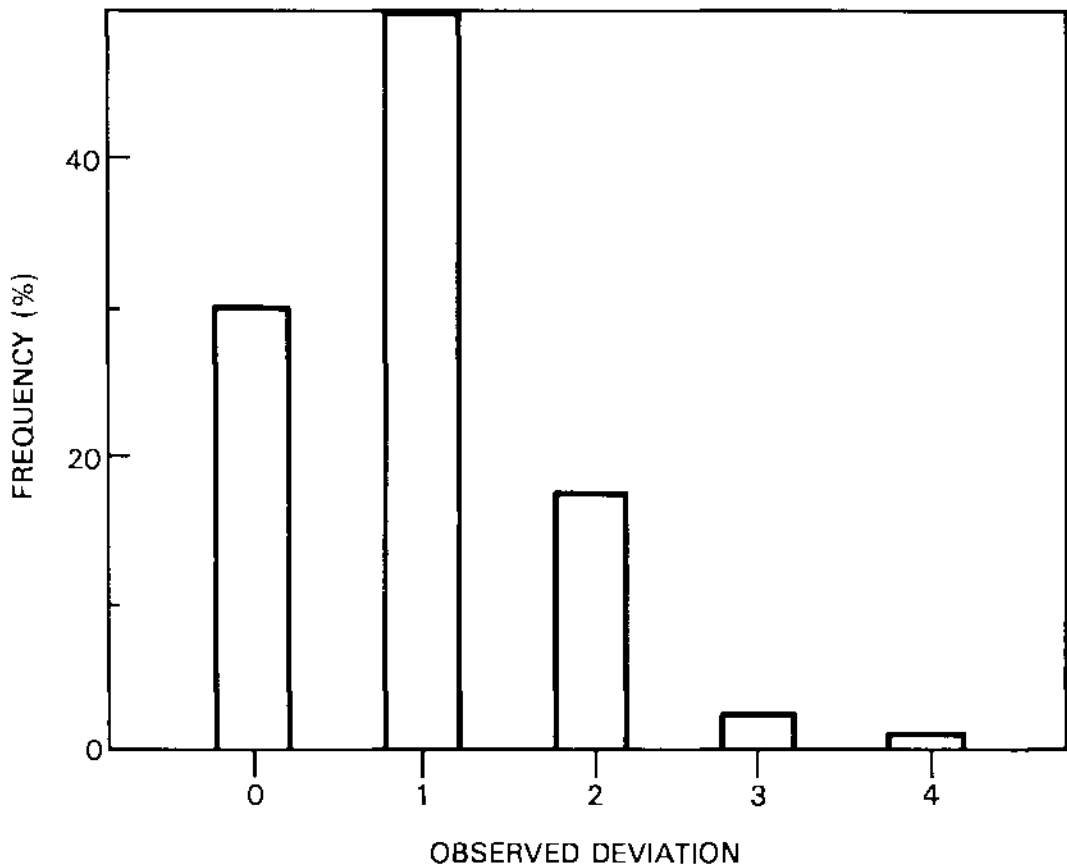


FIGURE I: Deviations in Location Distributions

The model site chosen to test agreement of this calculational method with distributions derived by recognized techniques with the Nine Mile Point Power Reactor.<sup>4</sup> The land environmental monitoring system includes five inner and six outer stations. The determination of the necessary number of on-site stations was based on meteorological data collected over a two-year period. The inner stations are located onsite at points in each land sector calculated to give the maximum average annual ground concentration. The outer stations are designed to measure background for comparison with onsite measurements. The layout of the land program is indicated in Figure II below.

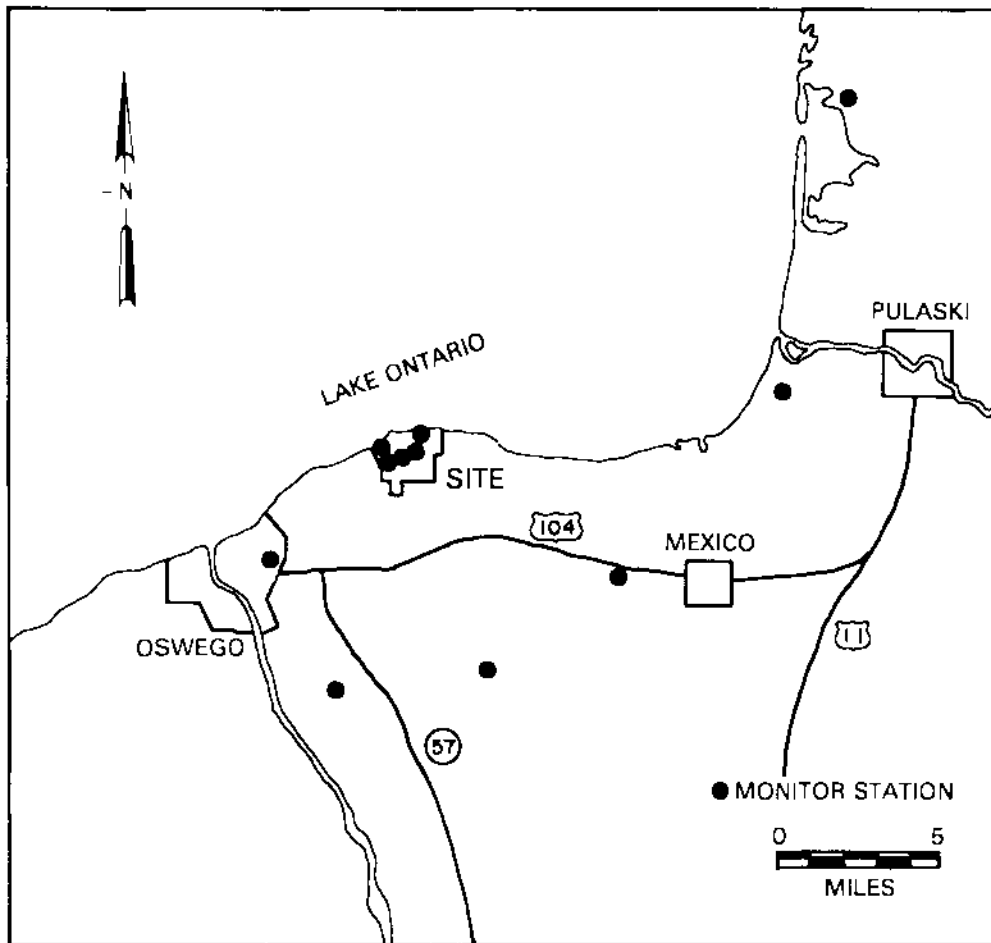


FIGURE II: Land Monitoring Station Locations

Applying the weighting factor procedure to demographic and meteorologic data for this site to distribute eleven sampling locations yielded the following comparison of programs:

<u>Octant No.</u>	<u>Model Sampling Locations</u>	<u>Calculated Sampling Locations</u>	<u>Deviation</u>
1	0	0	0
2	1	1	0
3	2	3	1
4	2	2	0
5	4	3	1
6	2	2	0
7	0	0	0
8	0	0	0

On the basis of this favorable comparison and the eleven mature sites comparison, the acceptability criteria were met.

## References

1. Pelletier, C.A. The Sensitivity of Environmental Surveys to Atmospheric Releases from Nuclear Facilities, Univ. of Michigan, 1966. (Ph.D. thesis)
2. Pelletier, C.A. "Performance and Design of an Environmental Survey," Environmental Surveillance in the Vicinity of Nuclear Facilities, Charles C. Thomas, Publ., Springfield, Ill., 1970.
3. Waite, D.A. An Analytical Technique for Distributing Air Sampling Locations Around Nuclear Facilities, BNWL-SA-4534, Battelle, Pacific Northwest Labs., Richland, Wash., May, 1973.
4. Philbin, T.W. and G.H. Whipple. "An Environmental Survey for the Nine Mile Point Power Reactor," Environmental Surveillance in the Vicinity of Nuclear Facilities, Charles C. Thomas, Publ., Springfield, Ill., 1970.





Les fuites de circuits, à l'état de gaz ou de vapeur d'eau, entraînent des rejets atmosphériques notables qui ont atteint, pour l'année 1972, 31 300 Ci en gaz rares et 23 mCi en produits volatils, iode presque exclusivement.

L'importance de l'activité rejetée et surtout l'augmentation par rapport à l'année 1971 qui s'est encore accusée dans les premiers mois de 1973 a conduit à rechercher les sources principales de ces rejets.

#### Méthodes de détection de fuites de gaz (octobre 1971)

Deux moyens de détection ont été utilisés :

- une chambre d'ionisation différentielle sur le circuit de ventilation générale, de seuil  $2 \cdot 10^{-6}$  Ci/m<sup>3</sup>, dont les indications pouvaient être comparées à l'équipement original de mesure de l'activité rejetée qui est constitué par quatre compteurs G.M. dans la veine de la cheminée,
- des détecteurs d'aérosols radioactifs portables (EAR 10), de seuil  $10^{-10}$  Ci/m<sup>3</sup>, dont le tuyau d'aspiration a été placé aux points critiques.

Pour tester la méthode, des fuites d'eau primaire (1 à 2 l/h) sur le circuit du réservoir de contrôle volumétrique ou de gaz (150 à 600 l/h) sur le réservoir de collecte des gaz primaires ont été provoquées. Lors de l'essai de fuites d'eau, le détecteur de gaz sur la ventilation générale n'a détecté la fuite qu'après arrêt des ventilations des autres étages (figure 1), le débit n'étant alors que 29 000 m<sup>3</sup>/h. A la reprise générale des ventilations (sauf la ventilation permanente de la caverne du réacteur), une augmentation nette a été perçue, également par les compteurs G.M. de la cheminée, suivie d'une stabilisation qui n'était pas encore parfaite au moment de la reprise de la ventilation permanente. La réponse des compteurs G.M. a été beaucoup moins nette dans ce dernier cas, en raison de leur contamination résiduelle.

Il a été ainsi vérifié que les fuites pouvaient être détectées par des détecteurs d'aérosols ou de gaz radioactifs, avec une sensibilité d'autant meilleure que la dilution de la fuite est faible. Compte tenu des périodes courtes des descendants solides des gaz en particulier, il est préférable de ne pas modifier les conditions de ventilation, cette modification étant de nature à changer les niveaux d'équilibre et à rendre illusoire la comparaison des résultats pour des situations de débit différentes. En conséquence, il a été jugé utile d'équiper les gaines de ventilation des centrales en construction (Fessenheim et Bugey) de trappes permettant l'introduction d'un embout de prélèvement pour détecteurs portables d'aérosols ou de gaz radioactifs. Cette solution est préférable à celle qui consisterait à faire des manœuvres sur la ventilation pour voir l'influence sur le détecteur fixe du rejet final. Les trappes doivent évidemment être prévues en amont des filtres.

#### Localisation des fuites à la Centrale de Chooz

En octobre 1971, les fuites de gaz se répartissaient assez également entre la caverne du réacteur et la caverne des auxiliaires. Des investigations complémentaires ont mis en évidence des fuites au pressuriseur dans le premier cas et sur le circuit d'alimentation en hydrogène du réservoir de contrôle volumétrique dans le deuxième cas.

Avec la croissance des rejets d'iode 131 au début de l'année 1973, il est apparu utile de rechercher l'origine de la fuite ainsi que la forme physico-chimique de l'iode, pour préciser les caractéristiques d'une filtration éventuelle.

Plusieurs sources possibles ont été prospectées ; des contaminations notables n'ont été mesurées que sur les pompes de charge ou dans la caverne du réacteur. Au premier emplacement, le faible débit de ventilation du local entraîne que la contribution de cette fuite dans le rejet général est faible.

Par contre, la contamination de la caverne du réacteur est importante et a été suivie pendant deux mois avec deux types de prélèvement :

- un prélèvement hebdomadaire sur un ensemble de filtres en série (papier - charbon - papier - charbon),
- un prélèvement sur un ensemble identique, où le filtre charbon amont a été conservé en service pendant deux mois, les autres filtres étant changés chaque semaine et tous étant mesurés à cette occasion.

De cette expérimentation, on a pu conclure que la quasi totalité de l'activité rejetée en iode provenait de la caverne du réacteur où elle se trouvait sous forme volatile. Le débit d'activité de la ventilation générale est généralement plus faible que celui mesuré sur la ventilation permanente de la caverne du réacteur, probablement en raison de la présence des filtres.

#### Forme de l'iode et efficacité d'une filtration éventuelle

La comparaison des activités mesurées en iode 131 sur les filtres amont et aval en service continu ou hebdomadaire tend à montrer que l'efficacité en utilisation continue d'une couche de charbon actif imprégné d'iodure de potassium d'épaisseur 35 mm diminuait de 99,9 % à 98 % en trois semaines et moins de 90 % en cinq semaines. Ces résultats mériteraient d'être confirmés en testant en continu l'efficacité d'un filtre d'épuration, comportant une épaisseur de charbon de 50 mm, à placer sur le circuit de ventilation permanente de la caverne du réacteur.

Cette situation a conduit à rechercher la forme chimique de l'iode par le moyen de May Pack, série de filtres de cuivre imprégnés ou non d'argent, suivie d'une série de filtres en charbon de noix de coco imprégnés ou non de TEDA. La première série permet d'évaluer l'activité retenue sous forme moléculaire, la deuxième série retient également les formes organiques et moléculaires.

Ces mesures exécutées à notre demande par le Service d'Etudes de Sureté Radiologique du Commissariat à l'Energie Atomique, ont montré que 90% de l'activité de l'iode correspondait à une forme organique quel que soit le lieu de prélèvement. Ceci explique la constatation faite précédemment sur la baisse d'efficacité du filtre. Des essais avec injection d'iode en un temps limité et circulation continue d'air montrent en effet que l'efficacité diminue beaucoup plus vite avec l'iode organique qu'avec l'iode moléculaire.

Si ces résultats sont confirmés par des mesures ultérieures, il apparaîtra que le meilleur moyen de protection est la réduction de la fuite à l'origine, la filtration n'étant en effet efficace que sous la condition d'un remplacement fréquent des filtres. On peut également envisager l'utilisation cyclique de deux filtres en parallèle, l'un étant en service, et l'autre en décroissance.

## Evolution de la contamination en cas d'arrêt de la ventilation

Un arrêt de la ventilation de la caverne du réacteur a été provoqué en juin 1972.

### Arrêt de la ventilation avec fuite de gaz

Après arrêt de la ventilation, on a observé une augmentation de la contamination de l'ordre d'un facteur 10, variable selon les nucléides (figure 2). L'activité volumique atteinte à l'équilibre a été mesurée, ou calculée dans le cas où la stabilisation n'avait pu être obtenue. A partir de la valeur d'équilibre et de la période du radioélément, on a évalué la fuite d'activité dans la caverne du réacteur, de volume libre 25 000 m<sup>3</sup>.

La comparaison de la répartition des radioéléments gazeux par rapport au <sup>133</sup>Xe montre que la contamination ne pouvait pas provenir d'une fuite directe d'eau primaire et que les radioéléments à vie longue étaient favorisés. On a également observé que les manoeuvres sur la phase vapeur du pressuriseur agissait sur l'évolution de l'activité, l'échantillonnage continu stabilisant les périodes longues, l'aspersion augmentant l'apport des périodes courtes surtout (figure 2).

Une fuite a effectivement été trouvée sur la phase vapeur du pressuriseur et, à partir des rapports entre le taux de la fuite en <sup>133</sup>Xe et <sup>135</sup>Xe et entre les contaminations volumiques de ces éléments dans l'eau primaire, on l'a évaluée à 13 kg/h.

Au cours de cet essai, il n'est absolument pas apparu d'iode. La contamination en tritium a atteint 7.10<sup>-6</sup> Ci/m<sup>3</sup> pour 0,2 Ci/m<sup>3</sup> dans le primaire.

### Arrêt de la ventilation avec fuite d'eau provoquée

Pour obtenir une contamination atmosphérique mesurable en iode et permettre d'évaluer un dépôt éventuel sur les surfaces intérieures du bâtiment il a fallu provoquer une fuite d'eau et la vaporiser par une plaque chauffante.

La contamination volumique s'est stabilisée très vite pour <sup>133</sup>I mais n'avait pas encore atteint son niveau d'équilibre pour <sup>131</sup>I après 80 heures de fuite. La période apparente a néanmoins pu être évaluée à 16,5 h pour <sup>131</sup>I et 10 h pour <sup>133</sup>I, ce qui, compte tenu des périodes radioactives de ces nucléides, conduit à une période de dépôt de 18 heures pour les conditions de l'essai. Il faut noter qu'aucune détermination de la forme chimique de l'iode n'a été faite à l'époque et que ces résultats ont été obtenus avec la ventilation interne en service (45 000 m<sup>3</sup>/h).

## Conclusion

Le souci de réduire l'activité rejetée par les fuites dans les centrales à eau impose de prévoir les moyens de détection, sous forme de détecteurs portables d'aérosols ou de gaz radioactifs destinés à prélever dans les gaines des circuits de ventilation grâce à des trappes prévues à la construction.

Dans les locaux isolables, l'arrêt de la ventilation permet de détecter des fuites très faibles. Dans les conditions de la centrale de Chooz, il est possible de déceler une fuite de gaz dans la caverne du réacteur représentant un débit d'eau primaire de 2 l/j, pour une activité de cette eau primaire de 1 Ci/m<sup>3</sup>.

En cas de fuite de vapeur, entraînant une libération d'iode dans l'atmosphère des locaux, le dépôt sur les structures est important. La

période de dépôt a été trouvée égale à 18 heures, mais cette détermination mériterait d'être reprise en s'intéressant à la forme chimique de l'iode. Une fuite de  $10^{-9}$  Ci/s conduit, à Chooz, à une contamination volumique à l'équilibre, à l'arrêt de la ventilation en circuit ouvert, de  $2,5 \cdot 10^{-9}$  Ci/m<sup>3</sup> en iode 131.

En l'absence d'incident particulier, la majeure partie de l'iode se trouve sous forme organique et volatile. L'efficacité des filtres en charbon de noix de coco imprégné d'iodure de potassium semble décroître rapidement lorsqu'ils sont traversés en permanence par l'air contaminé. Sous réserve de confirmation de cette conclusion, il apparaît que l'épuration continue de l'iode des effluents gazeux par ce procédé n'est pas susceptible d'apporter une réduction importante des rejets, sauf par permutation cyclique de filtres disposés en circuits parallèles.

La mesure d'activité des gaz rejetés à la cheminée par chambre d'ionisation à circulation s'est avérée beaucoup plus représentative des rejets que la mesure par compteurs G.M., dont la réponse est affectée par la contamination des structures internes de la cheminée et du gaz porteur par les descendants des gaz rares.

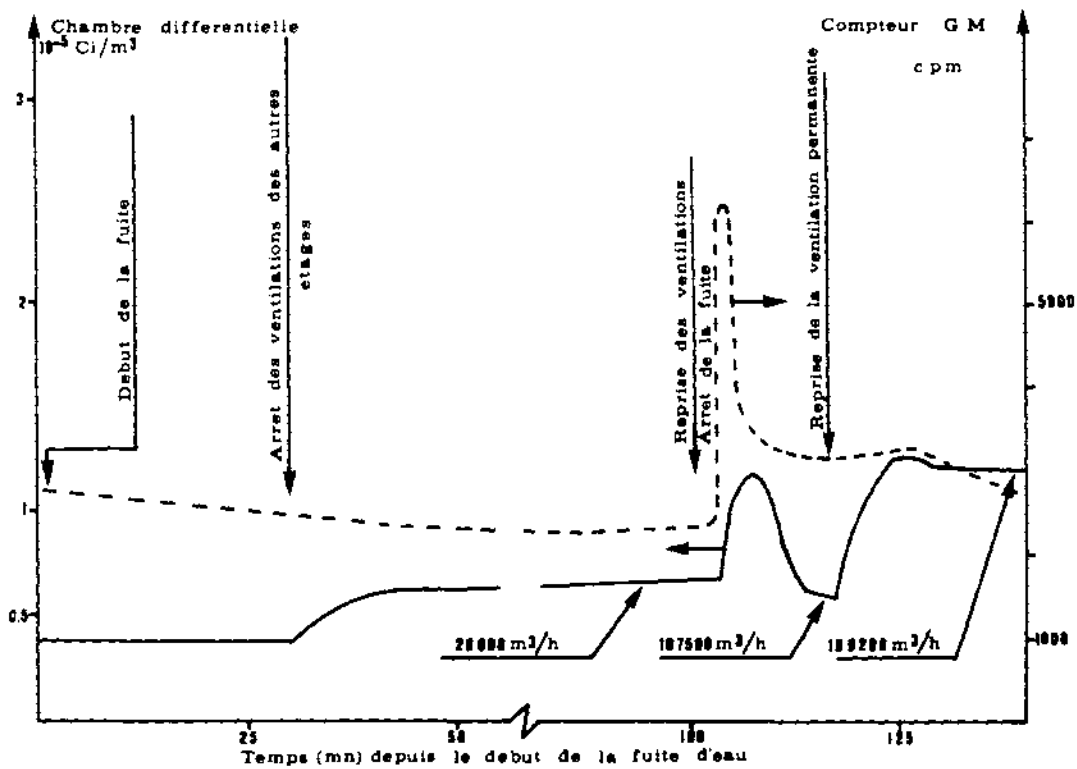


Figure 1 - Réponse des détecteurs de la cheminée pour une fuite d'eau.

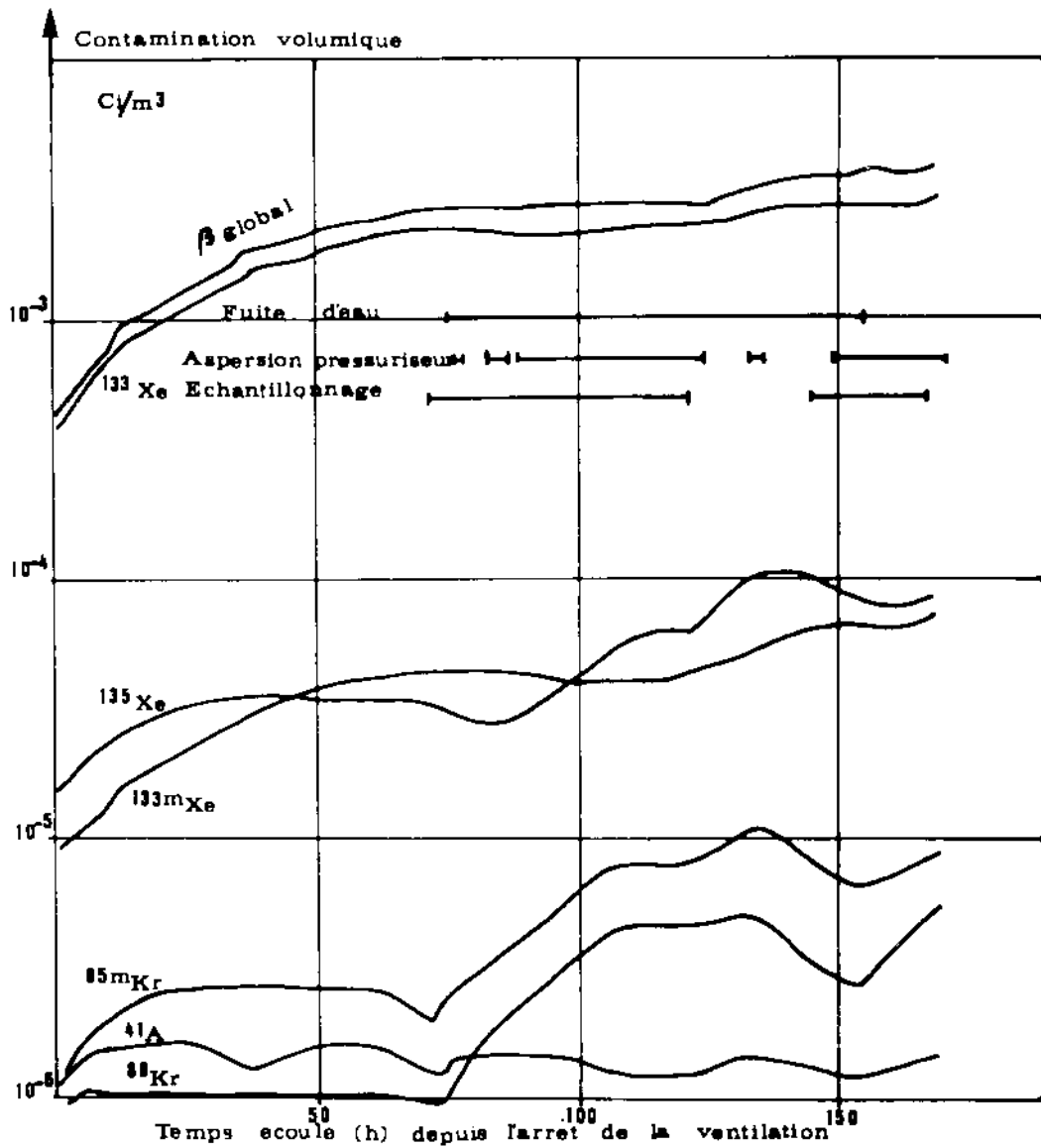


Figure 2 - Evolution de la contamination atmosphérique dans la caverne du réacteur après arrêt de la ventilation.

## LATE RADIATION EFFECTS

### MALIGNANCY RISK TO HUMANS FROM TOTAL BODY $\gamma$ -RAY IRRADIATION\*

Charles W. Mays, Ray D. Lloyd  
Radiobiology Division, University of Utah,  
Salt Lake City, Utah, U.S.A. 84132

and John H. Marshall  
Center for Human Radiobiology, Argonne National  
Laboratory, Argonne, Illinois, U.S.A. 60439

ABSTRACT: Based on data from the A-bomb survivors and radiobiological studies on the dose-rate effect, the following estimates are made of the cumulative deaths from induced malignancies in a natural population of 1,000,000 persons of mixed ages each receiving a total body dose of 1 rad from  $\gamma$ -rays:

	RADIATION-INDUCED DEATHS	
	at HIGH dose-rate (over 10 rad/min)	at LOW dose-rate (under 0.01 rad/min)
LEUKEMIAS		
Higher linear estimate	40	20
Preferred linear estimate	25	5
Lower linear estimate	14	1
Dose squared estimate	0.1	0.004
FATAL CANCERS (excluding leuk.)		
Higher linear estimate	150	75
Preferred linear estimate	100	20
Lower linear estimate	50	5
Dose squared estimate	0.4	0.016

The linear estimates are for use in radiation protection work, while the dose squared estimates illustrate the radiobiological possibility, based on the observed incidence of leukemia at Nagasaki, that the dose-response to  $\gamma$ -irradiation may be sigmoid, rather than linear.

Caution: These estimates are provisional and subject to future revision as more information is acquired.

### INTRODUCTION

We have carefully reviewed previous estimates of risk including the BEIR Report,<sup>1</sup> UN Reports,<sup>2</sup> Marinelli,<sup>3</sup> Dolphin,<sup>4</sup> and ICRP Publications 14<sup>5</sup> and 8<sup>6</sup>. Each of these prior reviews has provided a valuable step toward the ultimate goal of a better quantification of the effects of radiation on mankind.

The most valuable information on the effects of total body irradiation of humans is from the A-bomb survivors.<sup>1-7</sup> At the time of burst they were a population of mixed sexes in which fetuses, children, and adults were represented. The average age at the time of burst was 29 years (BEIR Report,<sup>1</sup> pg. 148). We assume that an average survival time of about 40 years after exposure can be used to obtain a reasonable approximation to the average risk, with

\* Work supported by the U.S. Atomic Energy Commission.

the realization that some of these A-bomb survivors have died sooner and were at risk a shorter time, while others will die later and be at risk a longer time. Our present risk estimates are averaged over the entire population of A-bomb survivors, which undoubtedly consists of some individuals more sensitive than average, as well as other individuals who are less sensitive.

We have analyzed the data in such a way so as to minimize interference from neutrons on the derived risk estimates for  $\gamma$ -rays. Our preferred estimates of lifespan risk involve future projections based on the observed trends of decreasing mortality rates from induced leukemia and increasing mortality rates from induced cancer. Yet despite the differences between our approach and that of the BEIR Report, there is reasonable biological agreement between our linear estimates and the BEIR linear estimates for high dose-rates.

To indicate the uncertainties in our "preferred" linear estimates, we have also derived "higher and lower" linear estimates. However, there is a strong possibility that the actual dose-response relationships may be sigmoid,<sup>1,7-11</sup> rather than linear. Therefore, alternative estimates of risk are given based on a dose-squared model.

Other reports have discussed the lesser overall effectiveness per rad expected at low dose-rates of  $\gamma$ -irradiation. We have now assembled sufficient radiobiological evidence to offer what may be the first realistic estimates of the actual risk at low dose-rates of total body  $\gamma$ -irradiation. This may be the most important contribution of the present report.

However, it must be emphasized that this is an interim report. Most of the irradiated subjects are still alive and must continue to be studied. Some uncertainties exist in the tissue doses actually received. We still lack fundamental knowledge of how malignant neoplasms are induced by irradiation. Therefore, it must be emphasized that the estimates of risk presented here are provisional and may require revision in the light of future information.

Now an outline of our analysis will be given. First, the risk for leukemia induction at high dose-rates will be estimated from the Nagasaki A-bomb data, taking advantage of the small exposure to neutrons. Next, the risk for cancer induction at high dose-rates will be obtained by multiplying the Nagasaki leukemia risk by the projected ratio of total induced cancer/total induced leukemia. Finally, the risks for leukemia and cancer induction at low dose-rates will be obtained by multiplying the risks at high dose-rates by appropriate effectiveness factors.

#### LEUKEMIA RISK AT HIGH DOSE-RATES

The mortality rate from leukemia in the A-bomb survivors at Hiroshima and Nagasaki from 1950 to 1970 is given in Table 1. The exposure is the "tissue kerma in air", which is the kinetic energy released per unit mass from the  $\gamma$ -rays and neutrons interacting with a small bit of tissue suspended 3 feet above ground.<sup>12</sup> The effect of shielding provided by buildings is included. The "tissue kerma in air" has also been called the "field free dose" or the "air dose," and will be referred to in this paper simply as the "kerma." The  $\gamma$ -ray dose within a person may be slightly less than the kerma from A-bomb  $\gamma$ -rays. However, the neutron dose within a person is much less than the kerma from A-bomb neutrons. This is due to a greater attenuation by the body of the neutrons than of the  $\gamma$ -rays. Hopefully, reliable estimates of the actual dose distributions within the human body may soon become available. In the meantime we shall assume provisionally that the  $\gamma$ -ray doses received by the A-bomb survivors are approximately equal to the  $\gamma$ -ray "tissue kerma in air." For conciseness, both the dose in rads and the dose equivalent in rems will

usually be referred to by the general term "dose" throughout this paper.

Table 1. LEUKEMIA MORTALITY IN A-BOMB SURVIVORS (1950-1970)  
(Jablon and Kato, BEIR Report p. 108, 1972, corrected)

Tissue kerma in air (rads)			No. of Persons	Person yr At risk	Leukemia Deaths	Leuk./yr (10 <sup>6</sup> persons)
Total	Gamma	Neutron				
HIROSHIMA						
200+	269.3	93.9	1460	26700	27	1010
100-199	108.5	30.1	1677	30200	10	331
50-99	56.9	13.3	2665	48300	7	145
10-49	17.6	4.3	10707	195400	17	87
0-9	0.9	0.3	43730	795600	34	43
NAGASAKI						
200+	329.1	5.6	1310	24300	15	616
100-199	144.3	1.4	1229	23000	3	130
50-99	70.3	0.2	1231	22900	0	0
10-49	21.3	0.0	3700	67600	2	30
0-9	2.3	0.0	11404	209900	11	52

In the fitting of dose-response curves, an important constraint is that the curves pass through (or at least near) control incidence. This is because no radiation effects occur at zero dose. The lowest dosage-group at each city is the most appropriate control for that city since it was followed-up similarly to the higher dosage-groups and the average kerma to the lowest groups was negligible (a total kerma of 1.2 rads at Hiroshima and 2.3 rads at Nagasaki). For each city, dose-response curves were started at the incidence rate of the lowest dosage group (regarded as zero rads) and were given a slope such that the predicted sum of leukemias exactly equalled the observed total, using the curve-fitting procedure of Mays and Lloyd.<sup>9</sup> The slope for Hiroshima (2.23 leuk. per yr/10<sup>6</sup> person rad) was much steeper than the slope for Nagasaki (0.88 leuk. per yr/10<sup>6</sup> person rad), primarily due to the greater neutron component from the Hiroshima weapon and the greater potency of neutrons relative to  $\gamma$ -rays in inducing malignancy. By trial and error it was found that the slopes for the two cities became equal at 0.8 leuk. per yr/10<sup>6</sup> person rem when an average neutron potency factor\* of 9 was assumed. The insensitivity of the Nagasaki risk rate coefficient to changes in the assumed neutron potency factor is shown in Table 2.

Table 2. NEUTRON POTENCY FACTOR and RISK RATE COEFFICIENT

Neutron Potency Factor	Leukemias/year (10 <sup>6</sup> person rem)	
	Nagasaki	Hiroshima
1	0.88	2.23
9	0.80	0.79
20	0.72	0.42

\*The neutron potency factor is defined as the ratio of  $\gamma$ -ray kerma/neutron kerma for equal biological effect. It is not equal to the neutron RBE which is the ratio of absorbed doses, since attenuation within the body causes the neutron dose in the body to be much less than the neutron kerma in air.



Wide ranges in the assumed neutron potency factor (1-20) cause only a small variation ( $\pm 10\%$ ) in the calculated risk rate coefficient for Nagasaki. This is due to the small neutron component of the Nagasaki weapon. The conclusion that the neutron potency factor increases as the kerma decreases<sup>13</sup> is probably correct, but has a negligible influence on the average risk rate coefficient for Nagasaki.

The preferred linear estimate will be taken as 0.80 leuk. per yr/ $10^6$  person rem for the years 1950-1970 (5 to 25 years after irradiation). However, there is some statistical uncertainty due to the small number of 20 leukemia cases in the Nagasaki population exposed to 10 rads and over. From Poisson statistical tables,<sup>14</sup> a 10% chance of having 20 or fewer cases corresponds to an expectation value of 27.1 cases (higher limit), whereas a 10% chance of having 20 or more cases corresponds to an expectation value of 14.5 cases (lower limit). In the exposed Nagasaki population the 20 cases at 10 rads and over, correspond to 12.8 induced cases plus 7.2 natural cases. The higher limit corresponds to 27.1-7.2 = 19.9 induced cases, while the lower limit corresponds to 14.5-7.2 = 7.3 induced cases. The higher linear estimate is  $(19.9/12.8)(0.80) = 1.25$  leuk. per year/ $10^6$  person rem, while the lower linear estimate is  $(7.3/12.8)(0.80) = 0.45$  leuk. per yr/ $10^6$  person rem.

The non-linear appearance of the plotted dose-response curve for Nagasaki raises reasonable doubt on whether the dose-response is really linear (Fig. 1). Among the 4931 persons exposed at Nagasaki to 10-99 rads (Table 1), 7.2 total cases of leukemia are predicted (4.7 natural plus 2.5 induced according to the "preferred" linear estimate), whereas only 2 leukemia cases were actually observed. A linear relationship predicting 7.2 cases when only 2 were observed

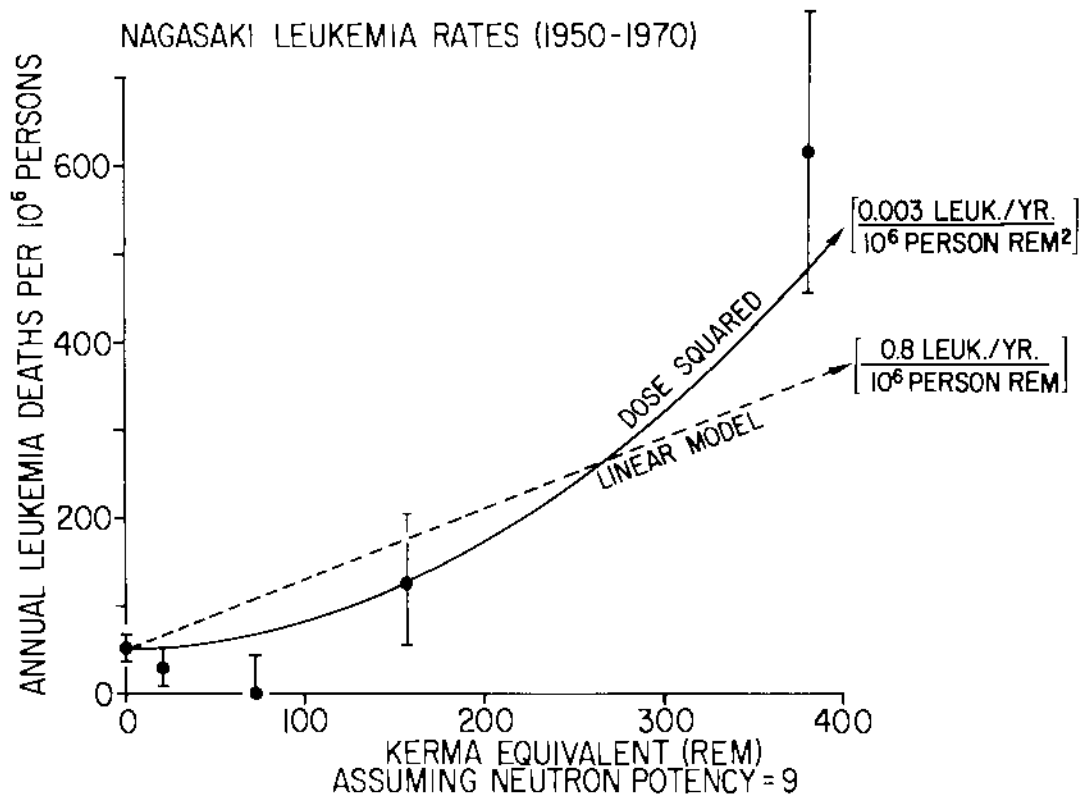


Fig. 1. Linear and dose-squared models fitted to the leukemia rates observed at Nagasaki (1950-1970). The dose-squared model makes a much better fit. The error bars are  $\pm 1$  standard deviation.

Table 3. INDUCED MALIGNANCIES IN HIGH DOSE A-BOMB SURVIVORS EXPOSED TO 200 RADS AND OVER AT HIROSHIMA AND NAGASAKI\* (Estimates in parentheses are for unavailable data)

Years After Irrad.	Years In Interval	Leukemia		Cancer	
		Deaths/yr 10 <sup>6</sup> persons	Deaths 10 <sup>6</sup> persons	Deaths/yr 10 <sup>6</sup> persons	Deaths 10 <sup>6</sup> persons
0 to 5	5	(800)	(4000)	(0)	(0)
5 to 10	5	1300	6500	600	3000
10 to 15	5	900	4500	200	1000
15 to 20	5	500	2500	300	1500
20 to 25	5	500	2500	2000	10000
25 to 30	5	(400)	(2000)	(3700)	(18500)
30 to 35	5	(300)	(1500)	(5400)	(27000)
35 to 40	5	(300)	(1500)	(7100)	(35500)
Total			25000		96500

\*The net rates of induced leukemia, and cancer excluding leukemia, within the observed intervals of 5-25 years were scaled from Fig. 16 of Jablon and Kato.<sup>7</sup>

Induced leukemia mortality before the start of the ABCC study (0 to 5 yr) is unavailable but is assumed reasonably close to the average rate during the observed intervals. Assumed leukemia mortality rates after 25 years are based on present trends. For this heavily irradiated population, exposed to a tissue kerma in air of 200 rads and over from  $\gamma$ -rays and neutrons at Hiroshima and Nagasaki, 25000 total deaths from induced leukemia/10<sup>6</sup> heavily irradiated persons is predicted (2.5% mortality from induced leukemia).

Induced cancer mortality during 0 to 5 years is assumed virtually equal to zero, due to the long latent periods typical for non-leukemic malignancy. Beyond 25 years, the mortality rates for induced cancer are tentatively assumed to increase by roughly the same amount in each successive 5-yr interval as the observed increase of 1700 cases per yr/10<sup>6</sup> persons which occurred between the intervals 15 to 20 yr and 20 to 25 yr. Under this provisional assumption 96500 total deaths from induced cancer/10<sup>6</sup> heavily irradiated A-bomb survivors is predicted (about 10% mortality from induced cancer excluding leukemia).

If the mortality rate from induced cancer remained constant at 2000 cases per yr/10<sup>6</sup> persons, then 45500 total deaths from induced cancer/10<sup>6</sup> heavily irradiated A-bomb survivors would be predicted. Conversely, if after 25 years the increase in each successive 5-year interval were 3400 cases per year/10<sup>6</sup> persons (or double the observed increase between 15 to 20 yr and 20 to 25 yr) 147500 total deaths from induced cancer/10<sup>6</sup> heavily irradiated A-bomb survivors would be predicted.

is rejected significantly ( $P = 0.03$ ). An excellent fit to the Nagasaki incidence rate is made by the fitted dose squared relationship of 0.003 induced leuk. per year/10<sup>6</sup> person rem<sup>2</sup>, starting at a natural incidence rate of 52 leuk. per yr/10<sup>6</sup> persons, and assuming an average neutron potency factor of 9. This dose squared relationship will be used to provide alternative estimates of risk.

Now, the lifetime risks will be estimated for leukemia induced by total body  $\gamma$ -ray irradiation at high dose-rates (10-1000 rem/min) such as received by the A-bomb survivors. Assuming the average death rate from induced

leukemia was the same in the unobserved interval 0 to 5 years after irradiation as in the observed 5 to 25 yr interval, the total incidence during the first 25 years following irradiation based on the preferred linear model would be  $(25 \text{ yr})(0.8 \text{ leuk per yr}/10^6 \text{ person rem}) = 20 \text{ leuk}/10^6 \text{ person rem}$ . Based on present trends (see Table 3 and Fig. 2.) about 80% of the lifetime leukemia risk should be expressed at 25 years. Therefore, the preferred linear estimate for the lifetime risk from leukemia is  $(20 \text{ leuk.}/10^6 \text{ person rem})/0.8 = 25 \text{ leuk.}/10^6 \text{ person rem}$ . The higher and lower linear estimates and the dose squared estimate were calculated similarly, and are shown in Table 4.

Table 4. LIFETIME RISK FROM LEUKEMIA AT HIGH DOSE-RATES  
(From a total body  $\gamma$ -ray dose "D")

Higher linear estimate	= (40 leuk./ $10^6$ person rem) D
Preferred linear estimate	= (25 leuk./ $10^6$ person rem) D
Lower linear estimate	= (14 leuk./ $10^6$ person rem) D
Dose squared estimate	= (0.1 leuk./ $10^6$ person rem <sup>2</sup> ) D <sup>2</sup>

Our linear estimates compare favorably with linear estimates derived from other sources. When we made a similar analysis on the excess leukemias in British patients given x-ray therapy for the treatment of ankylosing spondylitis,<sup>9,15</sup> we obtained 21 leukemias/ $10^6$  person rem averaged over the total marrow. (The average dose to the spinal marrow was taken as 880 rads,<sup>4</sup> and since about 40% of the active bone marrow was irradiated, the mean dose to the total

INDUCED MALIGNANCY RATES (OBSERVED & PREDICTED)  
IN A-BOMB SURVIVORS EXPOSED TO OVER 200 RADS

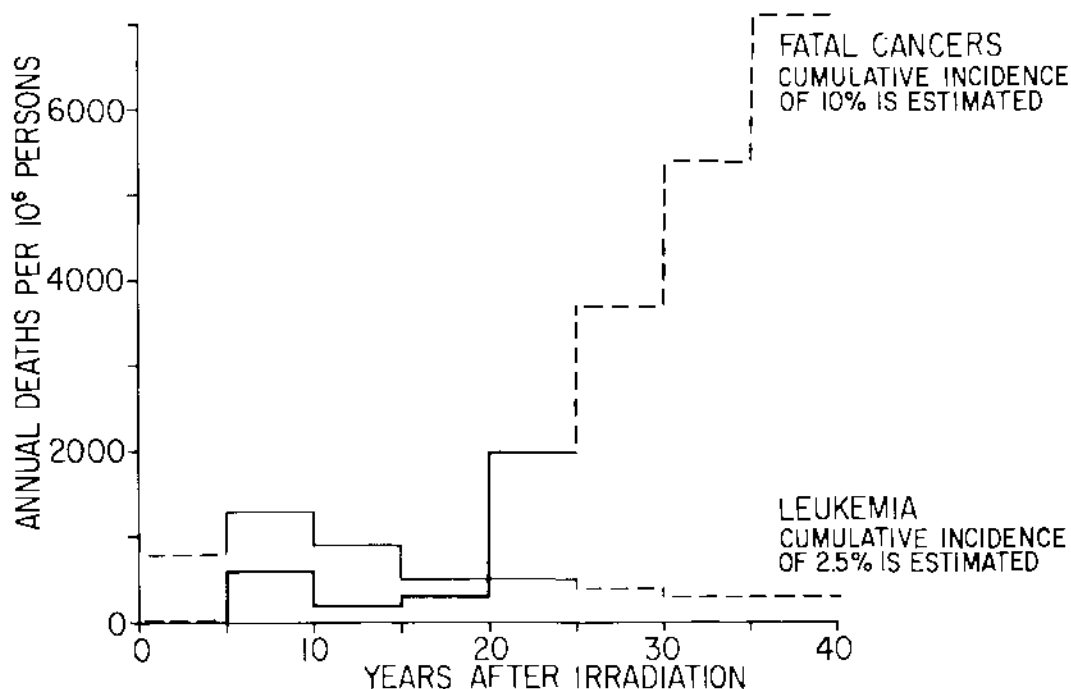


Fig. 2. Death rates from induced malignancy in the A-bomb survivors exposed to 200 rads and over (observed rates are shown as solid lines; predicted, as dashed lines). The rates from induced leukemia are decreasing, while those from induced cancer are increasing. See Table 3 for details.

marrow was taken as 350 rads). The BEIR Report<sup>1</sup> estimate (p. 169) of 516-738 leukemias/yr in 200,000,000 persons receiving 0.1 rem/yr corresponds to 26-37 leuk./10<sup>5</sup> person rem. Dolphin and Marley<sup>4</sup> estimate a lifetime risk of 20 leuk./10<sup>5</sup> person rem. Our preferred linear estimate of 25 leuk./10<sup>6</sup> person rem seems a reasonable estimate (presuming linearity) for the induction of leukemia at high dose-rates, and will be used in deriving linear risk estimates for cancer induction.

#### CANCER RISK AT HIGH DOSE-RATE

The lifetime risk from fatal cancers (fatal malignant neoplasms excluding leukemia) induced at high dose-rates will now be estimated as the projected ratio of fatal cancers/fatal leukemias, multiplied by the lifetime risk from leukemia. The combined high dosage groups at Hiroshima and Nagasaki exposed to 200 rads and over are of special interest because their incidences of leukemia and cancer are clearly elevated above normal values.<sup>7</sup> As detailed in Table 3 and Fig. 2, about 2.5% of the people in this highly exposed group are expected to die of radiation-induced leukemia, whereas if the mortality rate from induced cancer continues to increase at the present trend, a cumulative total of about 10% of these highly exposed persons are predicted to die of radiation-induced cancer. Assuming 4 induced cancer deaths per induced leukemia, the preferred linear estimate of the lifetime risk from fatal cancer becomes (4 cancers/leuk.)(25 leuk./10<sup>6</sup> person rem) = 100 fatal cancers/10<sup>6</sup> person rem.

However, it is uncertain whether or not the death rate from induced cancers will continue upward exactly according to the present trend. If it increases at twice the present trend, a cumulative induction of about 6 fatal cancers/leukemia is projected, for which the lifetime risk would be (6 fatal cancers/leuk.)(25 leuk./10<sup>6</sup> person rem) = 150 fatal cancers/10<sup>6</sup> person rem, which tentatively we regard as a plausible higher linear estimate. On the other hand, if the death rate from induced cancer will plateau at its 1965-1970 level, a cumulative induction of about 2 fatal cancers/leukemia is indicated, for which the corresponding lifetime risk would be (2 fatal cancers/leuk.)(25 leuk./10<sup>6</sup> person rem) = 50 fatal cancers/10<sup>6</sup> person rem, which we regard as a provisional lower linear estimate. There is much uncertainty in these projections since they involve not only the applicability of the models used, but what fraction of the future cancers will become fatal, considering new advancements in medical treatment. The next follow-up should be of exceptional interest.

For our dose squared model, the lifetime cancer risk is taken as (4 fatal cancers/leuk.)(0.1 leuk./10<sup>5</sup> person rem<sup>2</sup>) = (0.4 fatal cancers/10<sup>6</sup> person rem<sup>2</sup>).

Risk estimates from fatal cancers at high dose-rate are shown in Table 5.

---

Table 5. LIFETIME RISK FROM FATAL CANCERS AT HIGH DOSE-RATES  
(From a total body γ-ray dose "D")

Higher linear estimate	= (150 fatal cancers/10 <sup>6</sup> person rem) D
Preferred linear estimate	= (100 fatal cancers/10 <sup>6</sup> person rem) D
Lower linear estimate	= (50 fatal cancers/10 <sup>6</sup> person rem) D
Dose squared estimate	= (0.4 fatal cancers/10 <sup>6</sup> person rem <sup>2</sup> ) D <sup>2</sup>

---

Corresponding linear estimates converted from pp. 168-169 of the BEIR Report<sup>1</sup> range from 60 to 420 with a best estimate of about 140 fatal cancers/10<sup>6</sup> person rem, excluding leukemia. Dolphin and Marley<sup>4</sup> estimate 80 fatal cancers/10<sup>6</sup> person rem. Unfortunately, the existing cancer results from ankylosing spondylitic patients receiving partial body x-ray therapy directed at selected

regions of the skeleton are of limited usefulness in deriving numerical estimates of cancer risk because of uncertainties in soft-tissue doses, and because of the possibility that spondylitic disease may enhance the incidence of certain forms of cancer.<sup>5,15</sup> The risk from induced cancer is more uncertain than that from induced leukemia.

CANCER AND LEUKEMIA RISK AT LOW DOSE-RATES

$\gamma$ -rays, x-rays, and  $\beta$ -particles interact with matter so as to transfer nearly all of their energy to electrons. These moving electrons produce a relatively sparse distribution of ionizations and excitations along their paths, and therefore,  $\gamma$ -rays, x-rays, and  $\beta$ -particles are known as radiations of low LET (linear energy transfer). The cell culture work of Elkind and Sutton<sup>16</sup> showed quite conclusively that considerable repair of the damage from low LET radiation was possible if sufficient time was allowed between successive irradiations. The implication from the "Elkind effect" is that in general, the residual damage from a given dose of low LET radiation should decrease as the dose-rate is lowered, due to increased available time for biological repair between successive local radiation events.

Table 6 presents ten comparisons of the dose-rate effectiveness factor (dose at high dose-rate/dose at low dose-rate for equal biological effect) for life shortening and the induction of neoplasms by low LET radiations.<sup>17</sup> The

Table 6. <span style="font-size: small;">{ Effectiveness at LOWER dose-rate Effectiveness at HIGHER dose-rate }</span>	
Life shortening in beagles, Andersen, <sup>18</sup> Casarett <sup>19</sup> (0.006-0.06 R/min vs. 8 R/min)	0.08
Life shortening in RF male mice, Upton <sup>20</sup> (0.004-0.06 rad/min vs. 80 rad/min)	0.07
Life shortening in RF female mice, Upton <sup>20</sup> (0.0004-0.07 rad/min vs. 7 rad/min)	0.45
Leukemia in RF male mice, Upton <sup>20</sup> (0.004-0.06 rad/min vs. 80 rad/min)	0.14
Leukemia in RF female mice, Upton <sup>20</sup> (0.0004-0.07 rad/min vs. 7 rad/min)	0.26
Leukemia in CBA and C57 Bl mice, R. H. Mole <sup>21</sup> (0.02 rad/min vs. 1.35 rad/min)	0.15
Leukemia in LAF <sub>1</sub> female mice, Grahn <sup>22</sup> (0.01-0.06 R/min vs. 2-20 R/min)	0.2
Bone sarcomas in CFl female mice, Finkel <sup>23,24</sup> (0.0001-0.01 rad/min vs. 0.02-0.09 rad/min)	0.05
Mammary tumors in S.D. female rats, Shellabarger <sup>25</sup> (0.03 R/min vs. 10 R/min)	0.68
Thyroid tumors in Lister and Long-Evans rats, Doniach <sup>26</sup> ( $\approx$ 1 rad/min vs. 150 rad/min)	0.1
----- Normal mean $\pm$ standard deviation	0.22 $\pm$ 0.20
Log-normal mean $\pm$ std. deviation	0.16 $\left\{ \begin{array}{l} + 0.20 \\ - 0.09 \end{array} \right.$

levels of effect ranged from slight to severe. Usually the low dose-rates were below 0.1 rad/min, and usually the high dose-rates were above 1 rad/min. Assuming a normal distribution of effectiveness ratios, the mean  $\pm$  std. dev. was  $0.22 \pm 0.20$ . Assuming a log-normal distribution, the corresponding mean was 0.16 with std. deviations of + 0.20 and - 0.09.

Tentatively, a preferred estimate for the overall effectiveness of low vs. high dose-rates from sparsely-ionizing radiation is taken as 0.2 for the summed impact of delayed somatic effects in humans, with somewhat arbitrary bounds of 0.1 to 0.5 for this overall effectiveness factor. The individual effectiveness factors will of course vary with biological endpoint and species (for example, 0.05 for bone sarcomas in CF1 mice vs. 0.68 for mammary tumors in Sprague-Dawley rats), and may vary with the incidence level at which comparison is made.

The estimated risk coefficient at low dose-rates is taken as that at high dose-rates multiplied by the indicated effectiveness factors shown in Table 7.

Table 7. POPULATION RISK FROM SPARSELY IONIZING RADIATION  
(Deaths per 1,000,000 persons receiving 1 rem)\*

	at HIGH dose-rate (over 10 rem/min)	Eff. Factor	at LOW dose-rate (under 0.01 rem/min)
LEUKEMIA			
Higher linear estimate	40	0.5	20
Preferred linear estimate	25	0.2	5
Lower linear estimate	14	0.1	1
Dose squared estimate	0.1	0.2	0.004**
CANCER (Exc. Leuk.)			
Higher linear estimate	150	0.5	75
Preferred linear estimate	100	0.2	20
Lower linear estimate	50	0.1	5
Dose squared estimate	0.4	0.2	0.016**

\* At extremely low doses from  $\gamma$ -rays, multiple ionizations within microscopic volumes of tissue are infrequent. Therefore, as the dose approaches zero, the effectiveness at high dose-rate should approach that at low dose-rate.

\*\* In the dose squared estimates, the doses are squared. Therefore, to convert a dose squared risk at high dose-rate to a dose squared risk at low dose-rate, the dose-rate effectiveness factor (which is a ratio of doses) must also be squared. It is uncertain which dose-rate effectiveness factor is most appropriate for the dose squared model. Tentatively, the average factor of 0.2 obtained from linear intercomparisons<sup>17</sup> has been used.

The following example illustrates the numerical calculation of risk. From Table 7, the predicted number of leukemias plus fatal cancers induced in a population of  $10^6$  persons receiving 10 rem of  $\gamma$ -irradiation to the total body at low dose-rate would be:

Preferred linear estimate

$$\left[ \frac{5 \text{ leuk.} + 20 \text{ fatal cancer}}{10^6 \text{ person rem}} \right] [10^6 \text{ persons}] [10 \text{ rem}] = 250 \text{ cases}$$

Dose squared estimate

$$\left[ \frac{0.004 \text{ leuk.} + 0.016 \text{ fatal cancer}}{10^6 \text{ person rem}^2} \right] [10^6 \text{ persons}] [10 \text{ rem}]^2 = 2 \text{ cases}$$

## DISCUSSION

For purposes of radiation protection, the use of the preferred linear estimates is recommended. For radiobiological predictions of actual effects, it might be desirable to give some consideration to the possibility that the dose-response relationship may be curvilinear.

For the purpose of applying these risk estimates to humans, we somewhat arbitrarily consider a "low" dose-rate to be below 0.01 rem/min and a "high" dose-rate to be above 10 rem/min. Thus,  $\gamma$ -ray exposures from background radiation, from properly operating nuclear reactors, and from most routine occupational situations can be regarded as occurring at low dose-rates; whereas high dose-rates typically apply to the A-bomb survivors, to patients exposed to medical x-rays, and to persons acutely exposed in radiation accidents. With more data it may be possible to estimate the dose-rate effectiveness in the "gap" between 0.01 and 10 rem/min.

These risk estimates apply to the "average" person in a general population of mixed ages, and may require modification to be applied to special groups, such as fetuses or patients with diseases, such as polycythemia vera, which can alter the susceptibility to radiation-induced malignancy.<sup>3,11,27</sup> The risk to populations in other parts of the world may differ somewhat from that to the A-bomb survivors. As a future refinement, it may be desirable to analyze the mortality and survival times of each age group separately rather than assuming an average post-irradiation survival time of 40 years for the total exposed population.

The lifespan risk estimates given for low dose-rates apply to uniform total body irradiation from  $\gamma$ -rays, x-rays,  $\beta$ -particles, but not for radiations of high LET, such as neutrons and  $\alpha$ -particles, since at least under some conditions the effectiveness of high LET radiation increases as the dose-rate is lowered.<sup>20,29</sup>

In the future, a better understanding of the dose-response relationships and the dose-rate effect is expected which should permit more reliable estimates of the actual risks from radiation. Until then, it is hoped that the risk estimates in this report may provide interim guidance.

## DEDICATION

This article is dedicated to the memory of John C. Bugher, M.D., at whose urging this analysis was undertaken.

## REFERENCES

1. BEIR Report, The effects on populations of exposure to low levels of ionizing radiation, National Academy of Sciences National Research Council, Wash., D. C. (1972).
2. United Nations Scientific Committee on the Effects of Atomic Radiation, Reports to the General Assembly: 17th Session, Supplement 16(A/5216) in 1962; 19th Session, Supplement 14(A/5814) in 1964; 21st Session, Supplement 14(A/6314) in 1966; 24th Session, Supplement 13(A/7613) in 1969; and 27th Session, Supplement 25(A/8725) in 1972, United Nations, New York.
3. L. D. Marinelli, Estimates of the radiation-induced leukemic risk in man, Argonne National Laboratory Report 7760, Part 2, pp. 133-153 (1970).
4. G. W. Dolphin and W. G. Marley, Risk evaluation in relation to the

protection of the public in the event of accidents at nuclear installations, in Environmental Contamination by Radioactive Materials, International Atomic Energy Agency, Vienna, pp. 241-254 (1969).

5. ICRP Publication 14, Radiosensitivity and Spatial Distribution of Dose, Pergamon Press, Oxford (1969).
6. ICRP Publication 8, The evaluation of risks from radiation, Pergamon Press, Oxford (1966).
7. S. Jablon and H. Kato, Studies of the mortality of A-bomb survivors. 5. Radiation dose and mortality, 1950-1970, *Radiat. Res.* 50, pp. 649-698 (1972).
8. R. E. Rowland, Patricia M. Failla, A. T. Keane, and A. F. Stehney, Tumor incidence for the radium studies, Argonne National Laboratory Report ANL-7860, Part 2, pp. 1-8 (1970).
9. C. W. Mays and R. D. Lloyd, Bone sarcoma risk from  $^{90}\text{Sr}$ , in Biomedical Implications of Radiostrontium Exposure, Ed. by M. Goldman and L. K. Bustad, USAEC Symposium Series 25, CONF 710201, Springfield, Virginia, pp. 352-375 (1972).
10. C. W. Mays and R. D. Lloyd, Predicted toxicity of  $^{90}\text{Sr}$  in humans, in Second International Conference on Strontium Metabolism, Chairman J. M. A. Leefhan, USAEC CONF-720818, Springfield, Virginia, pp. 181-205 (1972).
11. C. W. Mays, Cancer induction in man from internally-deposited radioactivity, *Health Physics*, 25, pp. 585-592 (1973).
12. J. A. Auxier, J. S. Cheka, F. F. Haywood, T. D. Jones, and J. H. Thorngate, Free-field radiation dose distributions from the Hiroshima and Nagasaki bombings, *Health Physics* 12, pp. 425-429 (1966).
13. A. M. Kellerer and H. H. Rossi, RBE and the primary mechanism of radiation action, *Radiat. Res.* 47, pp. 15-34 (1971).
14. E. C. Molina, Poisson's Exponential Binomial Limit, 5th Printing, Van Nostrand Co., New York (1949).
15. W. M. Court-Brown and R. Doll, Mortality from cancer and other causes after radiotherapy for ankylosing spondylitis, *Brit. Med. J.* 2, pp. 1327-1332 (1965).
16. M. N. Elkind and Harriet Sutton, Radiation response of mammalian cells grown in culture, *Radiat. Res.* 13, pp. 556-593 (1960).
17. C. W. Mays, R. D. Lloyd, and J. H. Marshall, Dose-rate effect of low LET radiation in lifespan shortening and the induction of neoplasms, manuscript in preparation (1973). Presented by CWM as an invited refresher course at the Radiation Research Meeting at St. Louis, Missouri on 3 May 1973.
18. A. C. Anderson and L. S. Rosenblatt, The effect of whole-body x-irradiation on the median lifespan of female dogs (beagles), *Radiat. Res.* 39, pp. 177-200 (1969).
19. G. W. Casarett and H. E. Eddy, Fractionation of dose in radiation-induced male sterility, in Dose Rate in Mammalian Radiation Biology, Ed. by D. G.



- Brown, R. G. Cragle, and T. R. Noonan, USAEC CONF-680410, National Technical Information Service, Springfield, Virginia, pp. 14.1 to 14.10 (1968).
20. A. C. Upton, M. L. Randolph, and J. W. Conklin, Late effects of fast neutrons and gamma-rays in mice as influenced by the dose-rate of irradiation: induction of neoplasia, *Radiat. Res.* 41, pp. 467-491 (1970).
  21. R. H. Mole, Patterns of response to whole-body irradiation: The effect of dose intensity and exposure time on duration of life and tumor production, *Brit. J. Radiol.* 32, pp. 497-501 (1959).
  22. D. Grahn, R. J. M. Fry, and R. A. Lea, Analysis of survival and cause of death statistics for mice under single and duration-of-life gamma irradiation, *Life Sciences and Space Research X*, Akademie-Verlag, Berlin, pp. 175-186 (1972).
  23. Miriam P. Finkel and Birute O. Biskis, The induction of malignant bone tumors in mice by radioisotopes, *Acta Union Internationale Contre le Cancer* 15, pp. 99-106 (1959), supplemented by additional data from Miriam Finkel to C. W. Mays (1971).
  24. Miriam P. Finkel and Birute O. Biskis, Experimental induction of osteosarcomas, in Progress in Tumor Research, Ed. by F. Homburger (Karger, Basel) 10, pp. 72-111 (1968).
  25. C. J. Shellabarger and R. D. Brown, Rat mammary neoplasia following  $^{60}\text{Co}$  irradiation at 0.03 R or 10 R per minute, *Radiat. Res.* 51, Abstract ED-3 (1972), supplemented by additional data from C. J. Shellabarger to C. W. Mays (16 Jan. 1973).
  26. J. Doniach, Effects including carcinogenesis of  $^{131}\text{I}$  and x-rays on the thyroid of experimental animals: A review, *Health Physics* 9, pp. 1357-1362 (1963).
  27. B. Modan and A. M. Lilienfeld, Polycythemia vera and leukemia -- The role of radiation treatment, *Medicine* 44, pp. 304-344 (1965).
  28. H. Spiess and C. W. Mays, Protraction effect on bone sarcoma induction of  $^{224}\text{Ra}$  in children and adults, in Radionuclide Carcinogenesis, Ed. by C. L. Saunders et al., AEC Symposium Series 29, CONF 720505, National Technical Information Service, Springfield, Virginia, pp. 437-450 (1973).
  29. A. Luz, W. A. Müller, W. Gössner, B. Hindringer, and O. Hug, The role of dose and dose-protraction for the induction of osteosarcomas after incorporation of short-lived  $\alpha$ -emitting bone-seekers, 9th Annual Meeting of the European Society for Radiation Biology, Rome (26-28 Sept. 1972).

## THE RISK OF NEW CANCER IN WOMEN RADIATED FOR BREAST CANCER

Leonard A. Sagan  
Palo Alto Medical Clinic  
Palo Alto, California 94301

George Linden  
California Tumor Registry  
California State Department of Health  
Berkeley, California 94704

### Abstract

Using the records of the California Tumor Registry, the appearance of new cancers was studied among women treated with radiation for cancer of the breast. Comparison was made with cases treated with surgery only. Results do not demonstrate any excess new cancers in the radiation treated group that can be attributed to radiation therapy.

### Introduction

Although ionizing radiation is known to be carcinogenic, the magnitude of risk involved among patients treated with radiotherapy is not well defined. Furthermore the few available studies show inconsistencies both within these studies as well as with the experience of atomic bomb survivors.

The study of rheumatoid spondylitis by Court, Brown and Doll<sup>1</sup> demonstrates a clearly excessive risk of leukemia following radiation therapy. A lesser relative increase in risk was noted for cancer sites which were included in the heavily irradiated area, the predominant increase being in pulmonary tissues.

Hutchinson,<sup>2</sup> who conducted an international survey of leukemia frequency among women with cervical cancer treated with radiation had found no increase in leukemia. He attempted to reconcile this finding with the spondylitic study by speculating that the smaller, more heavily irradiated marrow in this group of patients was no longer capable of a neoplastic response.

Wagoner<sup>3</sup> and Doll<sup>4</sup> have also carried out studies of women treated for pelvic disease with radiation therapy. In both cases, an increase in leukemia was found, a difference that is difficult to reconcile with the Hutchinson study.

Two studies have been found in which women treated for breast disease with radiation have been studied for subsequent neoplasia. Mettler et al.<sup>5</sup> surveyed 606 women treated for acute postpartum mastitis with air doses ranging from 75 to more than 1000 rads. They found a total of 28

malignancies at all sites compared with an expectation of 21.67, but if breast cancers were removed, then the expected numbers very closely approximated those observed i.e. 15.79 vs. 15. Without an adequate control he was unable to distinguish between radiation causality or an association between mastitis and breast malignancy.

Schottenfeld and Berg,<sup>6</sup> utilizing the records at Memorial Sloan-Kettering, followed up 9,792 women treated for breast cancer between the years 1949 and 1962. The frequency of all malignancies was twice expectation, but the excess was entirely accounted for by second breast cancers. At individual sites, there were increases in ovarian, thyroid and sarcomatous malignancies, both of the soft parts and bone; however the numbers were small and therefore of doubtful significance. The authors do not state whether or not any of these cancers were treated with radiation.

There are no reports in the literature of radiation associated carcinogenesis among patients treated with radiation for carcinoma of the breast. Such a study was carried out utilizing records of the California Tumor Registry, making comparison with women with breast cancer treated surgically. It is concluded that there is small excess of cancer in the radiation treated group that is probably due to chance variation or to confounding variables. The evidence does not support the hypothesis of radiation as the etiologic factor.

#### Methodology

The California Tumor Registry is a central registry of 48 hospitals in California. It functions as one section of the San Francisco Bay Area Resource for Cancer Epidemiology of the California State Department of Health. Each of the participating hospitals abstracts and submits to the Registry all cases of reportable neoplasm including data on age, sex, race, marital status, diagnostic information, stage of disease, treatment and survival. The hospitals included in the Registry system diagnose about one-third of all cancer cases in California.

Since the initiation of the Registry in 1942 to a cut-off date of December 1969, 29,540 cases of breast cancer among white females were reported to the Registry as having been treated by surgery and/or radiation. These are shown in Table I. Eighty four percent of the 10,154 "radiation" cases received both surgery and radiation whereas those listed as surgical cases were treated with surgery alone ("Treatment" refers to first course of treatment).

Expected numbers of second cancers were generated by applying age-specific cancer incidence rates from the population based Alameda County Cancer Registry to the accumulated person years in each age group.

The observed numbers of cases (except for breast cancer) are generally lower than the expected numbers of cases. This is probably due to the portion of subsequent cancers developed by the study group which were diagnosed and treated in hospitals outside the California Tumor Registry system. The incomplete ascertainment of all subsequent cancers lessens the reliability of the data and makes necessary cautious interpretation of the study results. This point is more fully discussed in a previous publication.

Table I

Observed<sup>1</sup> and Expected<sup>2</sup> Cancer Cases  
 Site of First Subsequent Primary Cancer  
 by Treatment<sup>3</sup> of Breast Cancer, White Females

California Tumor Registry, 1942-1969

<u>Site of First Subsequent Primary Cancer</u>	<u>Treatment (Breast Cancer)</u>			
	<u>Surgery (19,386)</u>		<u>Radiation (Incl. Surg + Rad) (10,154)</u>	
	<u>Observed</u>	<u>Expected</u>	<u>Observed</u>	<u>Expected</u>
All sites	886	929	361	327
Buccal cavity and pharynx	23	23	4	8
Digestive organs and peritoneum	167	268	74	91
Respiratory system	21	42	10	15
Lung	17	38	9	14
Breast	426	247	172	89
Genital organs	146	177	49	65
Corpus uteri	60	79	22	29
Urinary organs	26	37	16	12
Lymphatic and hematopoietic system	27	54	13	19
Leukemia	10	23	5	8
Other and unspecified sites	50	81	23	28
Thyroid	17	12	7	5

1 Observed cases include first subsequent primary cancer following diagnosis of breast cancer and occurring prior to closing date of 12/69.

2 Expected cases were obtained by applying Alameda County Cancer Registry incidence rates to the person-years population.

3 First course of tumor-directed treatment, or series of treatments, initiated within four months after diagnosis.

Note: Observed and expected cancer cases exclude in situ cases and basal and squamous cell carcinomas of the skin.

The number of cases in each group is shown in parentheses.

Making the assumption that the degree of incomplete ascertainment of subsequent cancers was the same for both treatment groups, the observed to expected ratios of the radiation treated cases were compared with the observed to expected ratios of the surgically treated group:

$$R \text{ (ratio)} = \frac{\frac{O_r}{E_r}}{\frac{O_s}{E_s}}$$

### Results

Table II shows the above noted ratios. For all sites combined the radiation group shows a 16 percent excess compared to those treated by surgery alone. Cancer of the breast constitutes the largest number of subsequent cases for both treatment groups and these might be questioned because of the difficulty in determining whether the subsequent cancer is a new primary or a recurrence; however, exclusion of breast cancer from the total for all sites combined has little effect on the ratio.

The urinary organs show the highest risk - 90% above the surgery group - but this may be due to chance variation based on small numbers.

The leukemia experience is of unusual interest because of the known increase in leukemia risk following radiation exposure, and because of reports of excess leukemia in women treated for breast cancer with radiotherapy. Carey et al. in a comprehensive review of the literature found both carcinoma of the breast and acute leukemia in 18 cases, 15 of which had received radiotherapy. They added six cases of their own, in which only one had received radiotherapy and suggested that there was a spontaneous association between the two diseases not explained by radiation exposure. Five cases were observed in the present study. Three of the five were of the myelogenous variety and two were of unspecified cell type. There were no cases of lymphatic leukemia.

In order to examine further the hypothesis of radiation as an etiologic factor, some further data tabulations were carried out. It was postulated that, if the excess cancers among the radiated group were, in fact, due to radiation exposure such an effect should have been more prominent in the second half of the 1942-1969 study period when megavoltage therapy had replaced orthovoltage techniques, producing a total dose approaching a two-fold increase. Table III shows that comparison.

There is a slight decrease in the radiation/surgery ratio for all sites combined and for most individual sites, but the ratio is essentially unchanged in the two periods. The lymphatic and hematopoietic tissues show a higher ratio in the first time period but the numbers of cases in the radiation groups are small. There were a total of nine observed radiation cases in 1942-54. The diagnoses in these nine cases are as follows:

Table II

Ratios<sup>1</sup> for Subsequent Cancer  
Among White Women Treated for Breast Cancer

<u>Site of First Subsequent Primary Cancer</u>	<u>Ratio<sup>1</sup></u>	
All sites	1.157	(361)
Buccal cavity and pharynx	0.500	(4)
Digestive organs and peritoneum	1.305	(74)
Respiratory system	1.333	(10)
Lung	1.437	(9)
Breast	1.120	(172)
Genital organs	0.914	(49)
Corpus uteri	0.999	(22)
Urinary organs	1.897	(16)
Lymphatic and hematopoietic system	1.368	(13)
Leukemia	1.438	(5)
Other and unspecified sites	1.331	(23)
Thyroid	0.988	(7)

$$^1\text{Ratio} = \frac{\frac{\text{Observed number of second cancers (Radiation Group)}}{\text{Expected number of new cases among white women of same age}}}{\frac{\text{Observed number of second cancers (Surgical Group)}}{\text{Expected number of new cases among white women of same age}}}$$

Note: numbers in parentheses are the observed number of second cancers among the women treated with radiation.

Table III

Ratios<sup>1</sup> for Subsequent Cancer  
Among White Women Treated for Breast Cancer  
by Year of Treatment for Breast Cancer

<u>Site of First Subsequent Primary Cancer</u>	<u>Year of Treatment</u>	
	<u>1942-54</u>	<u>1955-69</u>
All sites	1.160 (133)	1.141 (228)
Buccal cavity and pharynx	0.667 (2)	0.400 (2)
Digestive organs and peritoneum	1.320 (27)	1.281 (47)
Respiratory system	1.900 (3)	1.118 (7)
Lung	2.250 (3)	1.154 (6)
Breast	1.156 (63)	1.084 (109)
Genital organs	0.831 (18)	0.967 (31)
Corpus uteri	1.142 (10)	0.898 (12)
Urinary organs	2.040 (6)	1.786 (10)
Lymphatic and hematopoietic system	2.557 (9)	0.659 (4)
Leukemia	3.667 (3)	0.686 (2)
Other and unspecified sites	0.642 (5)	1.904 (18)
Thyroid	1.000 (2)	0.909 (5)

$$^1 \text{Ratio} = \frac{\frac{\text{Observed number of second cancers (Radiation Group)}}{\text{Expected number of new cases among white women of same age}}}{\frac{\text{Observed number of second cancers (Surgical Group)}}{\text{Expected number of new cases among white women of same age}}}$$

Note: Numbers in parentheses are the observed number of second cancers among the women treated with radiation.

Reticulum cell sarcoma	1
Lymphosarcoma	2
Lymphoblastoma	1
Multiple myeloma	2
Acute myelogenous leukemia	1
Leukemia cell type unspecified	<u>2</u>
Total	9

The variety of histological cell types does not support the thesis of a single etiologic agent.

A second approach to evaluation of the excess second cancers among the radiation treated group was to examine the latent period between treatment and appearance of second cancers. With the exception of leukemia, the risk of which tends to appear early and then fall among both Japanese A-bomb survivors and irradiated spondylitics, the appearance of solid tumors tends to slowly increase with time and is only now becoming apparent in both of these studies. Therefore, the interval between first and second cancers among radiation and surgically treated groups was compared at five year intervals, with the expectation that radiation induced cases would tend to increase with increasing intervals of time. Tabulations are shown in Table IV. Second cancers at all sites do not show any trend, the ratios by five year intervals following first diagnosis of breast cancer being 1.13, 1.22 and 1.08. Although cancer of the lymphatic and hematopoietic tissues shows a rising frequency with time, the numbers of cases are very small.

#### Discussion

Data presented demonstrate that among some 10,154 women treated for breast cancer with radiation therapy, there is a 16% excess of second cancers in comparison with women with breast cancer who are treated with surgery alone. Detailed examination of the data, however, is otherwise inconsistent with an etiology solely or even partially related to radiation. The inconsistencies are as follows:

1. Radiation induced neoplasia would be expected to appear in tissues which are in or near the treated organ, i.e. thyroid, lung, esophagus and the other breast. Such an effect was not found.

2. Since radiation dose used for treatment of breast cancer was considerably greater during the second half of the total study period 1942-1969 because of the introduction of megavoltage therapy, one would expect the excess to appear more prominently in the second half of this period than in the first. In fact, the excess was similar in the two periods. (It is possible, of course, that such an effect might appear in the 1955-1969 cases after longer intervals of 15 or 20 years following radiation treatment.)

3. Radiation induced neoplasia would be expected to appear increasingly with increasing intervals following therapy. In fact, the excess was found equally at all intervals studied.

An alternative explanation is that the 16% excess cancer frequency noted among women treated with radiation is the result of chance variation, confounding variables or the incomplete ascertainment of all secondary cancers.



Table IV

Ratios<sup>1</sup> for Subsequent Cancer  
 Among White Women Treated for Breast Cancer  
 by Interval Between Diagnosis of Breast Cancer and Second Cancer

Site of First Subsequent Primary Cancer	Interval (Years)					
	0-4		5-9		10+	
All sites	1.127	(237)	1.217	(81)	1.076	(43)
Buccal cavity and pharynx	0.173	(1)	0.000	(0)	4.000	(3)
Digestive organs and peritoneum	1.141	(44)	1.751	(19)	1.301	(11)
Respiratory system	0.862	(3)	2.292	(5)	1.600	(2)
Lung	1.000	(3)	2.222	(4)	1.750	(2)
Breast	1.112	(119)	1.057	(33)	1.099	(20)
Genital organs	1.078	(36)	0.742	(9)	0.506	(4)
Corpus uteri	1.238	(14)	0.900	(6)	0.577	(2)
Urinary organs	1.810	(10)	2.083	(5)	1.333	(1)
Lymphatic and hematopoietic system	1.208	(7)	1.591	(5)	1.833	(1)
Leukemia	1.200	(3)	1.500	(2)	0.000	(0)
Other and unspecified sites	1.484	(17)	1.528	(5)	0.375	(1)
Thyroid	1.633	(7)	0.000	(0)	0.000	(0)

$$^1 \text{Ratio} = \frac{\frac{\text{Observed number of second cancers (Radiation Group)}}{\text{Expected number of new cases among white women of same age}}}{\frac{\text{Observed number of second cancers (Surgical Group)}}{\text{Expected number of new cases among white women of same age}}}$$

Note: Numbers in parentheses are the observed number of second cancers among the women treated with radiation.

Ignoring the variation in the data and accepting the 16% excess cancers as radiation induced allows comparison of this experience with that found among other exposed human populations. The recent report of the National Academy of Sciences<sup>10</sup> estimates one to two cases of leukemia and six cases of all other cancer per million man rems per year of experience. The radiation treated group reported here, assuming an average treatment dose of 4000 rads would contain at least 175,000,000 person-rem years of experience. Using the National Academy of Science estimate referenced above, one would have expected an excess of 175 cases of leukemia and six times as many cases of all cancers. The absence of any significant excess of leukemia cases supports Hutchinson's contention that the entire marrow must be exposed, as in the spondylitics and Japanese survivors, in order for the leukemogenic effect to be seen.

There is another possible explanation of the discrepancy between this experience and that of the two major population studies on which the NAS estimates were made and that is the very large difference in dose. In the latter cases, where leukemia was observed, doses were in the order of hundreds of rads whereas in the breast cancer group, treatment dose was in the order of several thousand rads. A number of animal studies as well as some human data suggests that there is an optimal carcinogenic dose and that doses exceeding those levels will fail to produce such an effect, and thus present a clear deviation from the widely postulated linear dose response relationship.

Certainly the same explanation must hold for the experience with both lung and thyroid, tissues considered to be radiosensitive and both of which are included, at least partially, in the treatment field when the cancerous breast is irradiated.

In summary, a study of the appearance of second cancers among women treated with radiation for breast cancer demonstrated a small excess of carcinogenesis when compared to women treated with surgery alone; however, the data do not implicate radiation as a causal factor.

#### References

1. Court Brown, W. M. and Doll, R.: Mortality from cancer and other causes after radiotherapy for ankylosing spondylitis. *Brit. Med. J.* 2, 1327-1332, 1965.
2. Hutchison, G. B.: Leukemia in patients with cancer of the cervix uteri treated with radiation. A report covering the first five years of an international study. *J. Nat. Cancer Inst.* 40, 951-982, 1968.
3. Waggoner, J. K.: Leukemia and other malignancies following radiation therapy for benign gynecological disorders. Unpublished. Paper presented at American Public Health Association Annual Meeting, November 12, 1969.
4. Doll, R. and Smith, P. G.: The long-term effects of x irradiation in patients treated for metropathia haemorrhagica. *Brit. J. Radiol.* 41, 362-368, 1968.
5. Mettler, F. A., Jr., Hempelmann, L. H., Dutton, A. M., Pifer, J. W., Toyooka, E. T. and Ames, W. R.: Breast neoplasms in women treated

- with x-ray for acute postpartum mastitis. A pilot study. J. Nat. Cancer Inst. 43, 803-811, 1969.
6. Schottenfeld, D., and Berg, J. W.: Incidence of multiple primary cancers. IV. Cancers of the female breast and genital organs. J. Nat. Cancer Inst. 46, 161-170, 1971.
  7. Dunn, J. E. Jr., Bragg, K. U., Sautter, C. and Gardipee, C.: Breast cancer risk following a major salivary gland carcinoma. Cancer 29, 1343-1346, 1972.
  8. Sakka, M.: Leukemia and ionizing radiation in Japan. Tokoku J. Exp. Med. 85, 12-15, 1965.
  9. Carey, R. W., Holland, J. F., Sheehe, P. R., and Graham, S. As association of cancer of the breast and acute myelocytic leukemia. Cancer 20, 1080-1088, 1967.
  10. Advisory Committee on the Biological Effects of Ionizing Radiations: The effects on populations of exposure to low levels of ionizing radiation. Natl. Acad. Sci., Natl. Res. Council, Washington, D.C. 72.

## RADIOSENSITIVITY AND DOSIMETRY OF THE TISSUES OF BONE

### IMPLICATIONS FOR SETTING MAXIMUM PERMISSIBLE LEVELS OF BETA EMITTERS

F. W. Spiers  
Department of Medical Physics  
The University of Leeds  
England

#### Abstract

Calculations are made of the risk to bone from incorporated  $\beta$  emitters, based on calculations of absorbed dose rates to bone marrow and endosteal tissues for a body burden of  $1 \mu\text{Ci}$ . The risk of leukaemia is based on human data and that of osteosarcoma and other bone tumours on a comparison of the incidence of these malignancies with the leukaemia incidence in animals continuously irradiated by  $\beta$  particles from ingested  $^{90}\text{Sr}$ . Maximum permissible body burdens are then derived by a comparison of these risk values with a total risk estimated to correspond with an annual dose of 5 rad to the whole body.

#### Introduction

Although the maximum permissible levels of bone-seeking radionuclides are at present related to the human data on carcinogenesis from  $^{226}\text{Ra}$ , there are unavoidable difficulties in this procedure. The comparison of any given radionuclide with  $^{226}\text{Ra}$  is based on the energy absorbed per unit mass of mineral bone, but the absorbed dose delivered to the cells of bone depends on the nature of the ionizing particle and its range. In making the comparison with  $^{226}\text{Ra}$  it is necessary therefore to adopt a quality factor for  $\alpha$  particles relative to  $\beta$  particles, but there is little, if any, evidence on which to set this value for late effects in bone. It has also been necessary to include a relative damage factor for those radionuclides which differ from radium in their pattern of deposition and, in the absence of quantitative data, this has to be given an arbitrary value.

In recent years, however, there has been a measure of agreement on the identity and location of the tissues at risk in irradiated bone and this has enabled dose parameters to be chosen that are more relevant to risk calculations than the absorbed dose determined simply for the bone matrix. Calculations of the relevant absorbed doses have been published for bone-seeking radionuclides that are distributed in volume throughout bone<sup>1,2</sup> and, very recently, the corresponding absorbed doses for radionuclides that are deposited on bone surfaces have been determined<sup>3</sup>.

Suggestions were made in the ICRP Report 11 that, in the case of high energy  $\beta$  emitters, the skeletal burden would be set by the limiting annual dose of 5 rem to bone marrow and that, for low energy  $\beta$  emitters, the limitation would be the dose to endosteal tissues in bone. In the intermediate energy range, however, the doses to bone marrow and bone surfaces set comparable limitations and, if the irradiation of both

marrow and endosteal tissues is regarded as undesirable, some better synthesis of the risks to both tissues should be sought.

It is worthwhile attempting to calculate the total risk in the case of bone because, although bone contains tissues that differ in their radiosensitivity, bone-seeking radionuclides irradiate only the tissues of bone, with practically no irradiation of other parts of the body. The problem of setting permissible levels of dose to organs of differing radiosensitivity, when more than one organ is irradiated, has been discussed in the Reports of two Task Groups in ICRP Publication 14<sup>5</sup>. A similar problem is encountered in bone because different radiosensitivities are assigned to the different tissues in bone, tissues that, under most circumstances, receive very different doses from an incorporated radionuclide. The present approach differs from that followed in the ICRP Publication 14 in that the risk of leukaemia from the irradiation of bone marrow is taken from human data as in the UNSCEAR 1972 Report<sup>6</sup>, while risk factors for tumours of other bone tissues are derived from data on the comparative risks of these tumours and leukaemia in animals irradiated continuously by high energy  $\beta$  particles from incorporated  $^{90}\text{Sr}+^{90}\text{Y}$ . A total risk for all bone tissues is then calculated on a dosimetric basis for a body burden of  $1\ \mu\text{Ci}$  of any given bone-seeking radionuclide and this is compared with an estimated occupational risk to give a maximum permissible body burden. The magnitude of the occupational risk is again based on UNSCEAR data and chosen to correspond approximately to an annual dose of 5 rad whole body irradiation at low LET. The maximum permissible body burdens calculated on these risk data compare interestingly with the present values of the ICRP.

#### Tissues at Risk and Late Effects in Bone

It was concluded in the ICRP Report 11<sup>4</sup> that the tissues at risk in irradiated bone were the active bone marrow with respect to leukaemia, the osteoprogenitive tissues of the endosteum with respect to osteosarcoma, and certain epithelial tissues adherent to bone in cranial air sinuses with respect to carcinoma. Considerations put forward more recently by Loutit and Vaughan<sup>7</sup> suggest that tumours may also arise from the reticulo-endothelial and supporting tissues in the marrow spaces in trabecular bone. With regard to the location of osteoprogenitive cells near endosteal surfaces, Sissons<sup>8</sup> and Vaughan<sup>9</sup> have concluded that the majority lie within a zone extending from the endosteal surface out to a distance of not more than  $10\ \mu\text{m}$ . Osteoprogenitive cells must also be present near endosteal surfaces in cortical bone but, because proliferative activity is so much less on these surfaces than on trabecular surfaces, it is assumed in this paper that the radiosensitivity of cortical endosteum is less than that of trabecular endosteum.

The late effects taken into account in the risk calculations, together with the corresponding tissues and their location and the relevant dose parameters, are summarized in Table 1. The associated risk factors are also designated for later reference.

#### Principles of the Calculation of Total Risk to Bone

The risk factors in Table 1 are defined as the probability of tumour occurrence per year for "continuous" irradiation and are expressed conveniently as cases  $/10^6/\text{yr}$  for an absorbed dose rate of 1 rad/yr to the relevant tissues. Numerically the value for a given risk rate is the same as the total number of cases per million occurring in the years following a single dose of 1 rad, - as given for example in the conclusions of the UNSCEAR Report<sup>6</sup>. Assuming that the risk factors in Table 1 are known, the total risk  $R_T$  to bone for a body burden of  $1\ \mu\text{Ci}$  can be written:

$$R_T = (r_m + r_o) \bar{D}_M + r_a \bar{D}_S + r'_o \bar{D}'_S$$

Table 1

Late Effect	Tissue and Location	Relevant Dose	Risk Factor
Leukaemias	Haemopoietic bone marrow in trabecular spaces	Average dose to red marrow, $\bar{D}_M$	$r_m$
Osteogenic sarcomas	Endosteal tissue layer, 10 $\mu\text{m}$ thick, on:	Average dose to endosteal layer:	
	(1) trabecular surfaces	(1) trabecular, $\bar{D}_S$	$r_o$
	(2) cortical surfaces	(2) cortical, $\bar{D}'_S$	$r'_o$
Angiosarcomas reticulum-cell tumours, some fibrosarcomas	Reticulo-endothelial and supporting tissues in trabecular spaces	Average dose to tissues in trabecular spaces, $\bar{D}_M$	$r_a$

where the absorbed dose rates  $\bar{D}_M$ ,  $\bar{D}_S$  and  $\bar{D}'_S$  are calculated from the corresponding dose factors  $\bar{D}_M/D_o$  etc. The parameter  $D_o$  is the absorbed dose rate to a very small soft-tissue inclusion in bone such that the irradiation is under conditions of particle equilibrium. The value of  $D_o$  is calculated in rad/yr for a body burden of 1  $\mu\text{Ci}$ , taking values of  $f_2$ , for the fraction of the body burden in the skeleton, from the Report of Committee II of ICRP, Publication 2<sup>10</sup>. The mass of bone matrix in which the radionuclide is distributed is taken as 5000 g.

If now some level of personal occupational risk,  $R_{MPL}$ , is assumed, the corresponding maximum permissible body burden,  $q$ , will be given by the equation:

$$qR_1 = R_{MPL} \quad (2)$$

where it is assumed that only the tissues of bone are irradiated and that the risk to the person is that to bone alone.

The dosimetric data on which the calculations are made are shown in Fig. 1 for volume-seeking radionuclides and in Fig. 2 for those that are deposited on surfaces. The dose factors are skeletal averages in the case of trabecular bone; for cortical bone the dose factors refer to the resorption cavities and Haversian canals of a section of a human femur, the only bone for which calculations are so far available. The calculations for surface-seeking radionuclides are based on the assumption that the retained radionuclide is distributed uniformly in a layer of very small thickness on the endosteal surfaces. Integration of the dose over the 10  $\mu\text{m}$  zone containing the cells at risk results, of course, in a finite average dose factor. It is convenient also to adopt the convention of expressing the dose factors for the surface seekers in terms of the parameter  $D_o$ . To do this, the retained radionuclide is considered to be distributed either over the total endosteal surface area  $A$ , or distributed through a bone volume  $V$ , and for numerical convenience the ordinate in Fig. 2 is given as  $\bar{D}(V/A \times 10^2 \times D_o)^{-1}$ . In this paper  $V$  is taken as approximately 2500  $\text{cm}^3$  and  $A$  as 10  $\text{m}^2$ ;  $V/A \times 10^2$  then has the value 2.5.

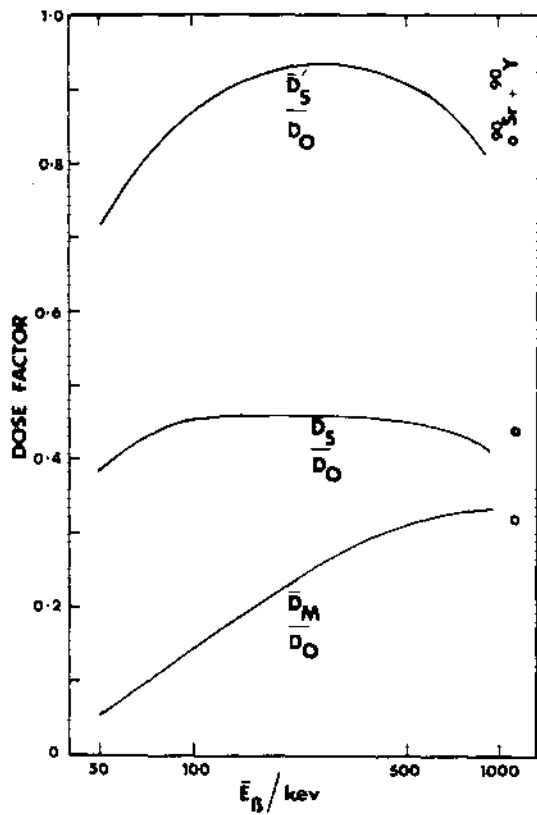


Fig. 1 Dose factors for volume-seeking  $\beta$  emitters.

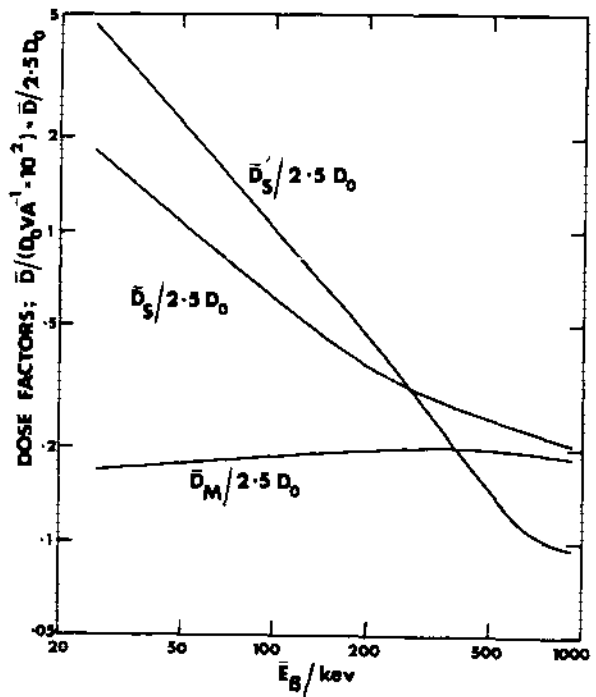


Fig. 2 Dose factors for surface-seeking  $\beta$  emitters; curve for  $D'_S$  only approximate.

### Choice of Risk Factors

#### Bone Marrow Risk

The bone marrow risk is considered to be leukaemia and the risk factor is taken from the upper limit of the range of 15-40 cases per rad per million persons exposed as given in the UNSCEAR REPORT<sup>6</sup>. The value of  $r_m$ , for a dose rate of 1 rad/yr, is therefore assumed to be:

$$r_m = 40/10^6/\text{yr} \quad (3)$$

#### Osteogenic Sarcoma Risk

There is not much evidence of the induction of bone tumours in humans by low LET external radiation and, in the ICRP Report 14<sup>5</sup>, bone tumour risk is passed as an order of magnitude less than leukaemia. On the other hand, where beagles<sup>11</sup> and miniature swine<sup>12</sup> have been subjected to continuous irradiation of bone and bone marrow from ingested  $^{90}\text{Sr}$ , both osteogenic sarcoma and myeloid and lymphoid leukaemias have occurred. It is therefore probably more relevant to the radiation protection situation to

take into account this evidence from continuous irradiation by a bone-seeker and to estimate a ratio of osteogenic sarcoma incidence to leukaemia incidence from animal data. Only data relating to irradiation by the energetic  $\beta$  particles of  $^{90}\text{Sr}+^{90}\text{Y}$  have been considered because it is only in this case that the bone marrow and the surfaces of trabecular bone receive comparable doses - see Fig. 1. The evidence from the beagles and swine is numerically not very concordant; in the beagles 19 osteogenic sarcomas have been recorded against 14 myeloid leukaemias whereas in the swine only 7 sarcomas have occurred, compared with 23 myeloid and 17 lymphoid leukaemias. In both cases, however, bone tumours show a greater prevalence in relation to myeloid leukaemia than suggested by the human data for external irradiation, received instantaneously or over a short period of time. It is therefore prudent for the present purpose to assume that, for the continuous internal irradiation of bone, osteogenic sarcomas are about half as prevalent as leukaemia and hence to assume a risk rate of  $20/10^6/\text{yr}$  for a dose rate of 1 rad/yr. We then have:

$$r_o + r'_o = 20/10^6/\text{yr} \quad (4)$$

The division of the risk between trabecular surfaces ( $r_o$ ) and cortical surfaces ( $r'_o$ ) can only be conjectured at present, but at least a clue is given by data from the radium poisoning cases. Here only two mid-shaft tumours have been recorded in a total of 30 sarcomas of the six long bones<sup>13</sup>. Considerations of the relative cortical and trabecular surface areas in the mid and outer thirds of these bones indicates that, despite some differences in dosimetry and isotope retention, the probability of tumour occurrence per unit area of cortical bone is unlikely to be more than one half of that of trabecular bone. Since the total surface areas of cortical and trabecular bone in the skeleton are roughly equal the following values of  $r_o$  and  $r'_o$  have been assumed:-

$$r_o = 13/10^6/\text{yr} \text{ and } r'_o = 7/10^6/\text{yr} \quad (5)$$

#### Angiosarcoma and other Bone Tumour Risk

Only animal data are available to suggest what magnitude should be attached to the risk of angiosarcoma, reticulum-celled tumours and other tumours that might arise in tissues of the marrow spaces. In the beagles continuously irradiated<sup>11</sup>, tumours of this class were only about one tenth as frequent as osteogenic sarcomas; similarly low ratios were shown in beagles given multiple injections of  $^{90}\text{Sr}$  at high dose<sup>14</sup> and in mice given high dose  $^{90}\text{Sr}$  injections<sup>15</sup>. On the other hand in beagles given low dose injections<sup>14</sup>, in beagles given single injections<sup>16</sup> and in mice given low dose injections, the ratio of angio and other tumours to osteogenic sarcomas ranged from 0.5 to 0.8. The low dose data have therefore been given the greater weight and the angio-group tumours have been taken to be half as prevalent as the osteosarcomas. The value of  $r_a$  has been assumed to be:

$$r_a = 10/10^6/\text{yr} \quad (6)$$

The overall result of these choices of risk factors is to make the bone tumour risk approach more nearly to the leukaemia risk than the human data for external irradiation suggests. It is probably better to make these assumptions, both on grounds of caution and because the animal data represent all that is available for the internal irradiation of bone by incorporated  $\beta$  emitters.

#### Risk Factor for Occupational Dose Level

The maximum permissible annual occupational dose is 5 rem for whole-body irradiation and this presumably represents a maximum permissible occupational risk. The conclusions of



the UNSCEAR Report<sup>6</sup> are that an absorbed dose of 1 rad may result (on the linear dose-effect hypothesis) in from 30 to 140 cases of certain specified cancers per 10<sup>6</sup> persons irradiated; there is a further estimate of some possible 40 cases of thyroid cancer but no figure is given for bone cancer. Great uncertainty necessarily attaches to these estimates and for the purpose of the present calculations the risk level is taken in round figures as 100 cases/10<sup>6</sup> persons and hence an "acceptable" maximum risk corresponding to 5 rad/yr is :

$$R_{MPL} = 5 \times 100 \times 10^{-6} = 5/10^4/\text{yr} \quad (7)$$

#### Total Risk to Bone and Calculation of the MPBB

Using the values of the risk factors given in Eqs. (3) (5) and (6), the total risk for a body burden of 1  $\mu\text{Ci}$  is:-

$$R_1 = (50\bar{D}_M + 13\bar{D}_S + 7\bar{D}'_S)/10^6/\text{yr} \quad (8)$$

and the value of the MPBB,  $q$ , is:-

$$q = R_{MPL}/R_1 = \frac{500}{50\bar{D}_M + 13\bar{D}_S + 7\bar{D}'_S} \mu\text{Ci} \quad (9)$$

This formula can be applied to both volume-seeking and surface-seeking  $\beta$  emitters if the values of the dose rates are calculated from the data in figs. 1 and 2.

#### Results for Volume-Seeking Beta Emitters

The risks calculated for six volume-seeking radionuclides are set out in Table 2 which gives the separate contributions from the trabecular spaces, the trabecular surfaces and the cortical surfaces. The total risk is then given, followed by the resulting value of the MPBB according to Eq. 9. The ICRP value quoted in the last column (from ICRP Publication 2<sup>10</sup>) is either that for bone as the critical organ (<sup>45</sup>Ca, <sup>89</sup>Sr, <sup>32</sup>P and <sup>90</sup>Sr + <sup>90</sup>Y) or bone as a reference organ (<sup>14</sup>C and <sup>18</sup>F). In the case of <sup>14</sup>C there is a large contribution to the dose to the trabecular spaces from the radionuclide present in marrow fat, because red marrow has been estimated to contain 53% fat<sup>16</sup>. In the case of <sup>18</sup>F the contribution to the total risk from the annihilation radiation is almost as great as that from the  $\beta$  radiation. If only  $\beta$  radiation from the radionuclides incorporated in the bone matrix were considered, the risk values for <sup>18</sup>F and <sup>14</sup>C would be lower and the calculated MPBB values much higher. The ratio of the present calculations of MPBB to those given by the ICRP would then range from 2 for high energy  $\beta$  emitters to about 4 for the low energy  $\beta$  emitter <sup>14</sup>C. This increase towards low energies is to be expected from the data in Fig. 1 where  $\bar{D}_M/D_0$  falls as the  $\beta$ -particle energy decreases. The dose factor  $\bar{D}'_S/D_0$  also falls at low energies but to a less marked extent.

#### Results for Surface-Seeking Beta Emitters

The risks and values of the MPBB calculated for five surface-seeking radionuclides are presented in Table 3 in the same form as in Table 1. These radionuclides have been considered to be surface-seekers either because they are known to deposit on endosteal surfaces or because they belong to the same chemical groups as known surface-seeking elements. Thus the three group IIIA radionuclides, <sup>171</sup>Tm, <sup>143</sup>Pr and <sup>90</sup>Y, are taken to deposit on resorbing and quiescent surfaces as does the lanthanide element yttrium<sup>17</sup>. The two group IIIB radionuclides, <sup>115</sup>In and <sup>204</sup>Tl, are considered to behave like the element gallium which deposits in regions of osteogenic activity<sup>17</sup>. As with the volume-seeking radionuclides the ratio of the present calculation of the MPBB to

Table 2: Volume-Seeking Beta Emitters

Radio-nuclide	Energy $\bar{E}_\beta$ /MeV	Risks as cases/10 <sup>6</sup> /yr for 1 $\mu$ Ci			BB Total risk	MPBB $\mu$ Ci	ICRP $\mu$ Ci
		Trabecular spaces	Trabecular surfaces	Cortical surfaces			
<sup>14</sup> C	0.050 $\beta^-$	1.58	0.35	0.10	2.03	250	300(fat) 400(bone)
<sup>45</sup> Ca	0.077 $\beta^-$	1.51	1.59	1.63	4.73	110	30
<sup>18</sup> F	0.25 $\beta^+$	6.60	3.08	3.43	13.11)	20	20
	0.51 $\gamma$	8.00	2.08	1.12	11.20)		
<sup>89</sup> Sr	0.55 $\beta^-$	33.90	12.61	14.03	60.54	8	4
<sup>32</sup> P	0.70 $\beta^-$	24.65	8.39	8.30	41.34	12	6
<sup>90</sup> Sr+ <sup>90</sup> Y	1.13 $\beta^-$	71.65	25.62	26.05	123.3	4	2
<sup>226</sup> Ra	5.65 $\alpha$	317	2077	1229	3623)	0.14	0.1
	0.42 $\beta^-$	21	8	8	37)		

Table 3: Surface-Seeking Beta Emitters

Radio-nuclide	Energy $\bar{E}_\beta$ /MeV	Risks as cases/10 <sup>6</sup> /yr for 1 $\mu$ Ci			BB Total risk	MPBB $\mu$ Ci	ICRP $\mu$ Ci
		Trabecular spaces	Trabecular surfaces	Cortical surfaces			
<sup>171</sup> Tm	0.026 $\beta^-$	1.70	4.80	6.68	13.18	40	90
<sup>115</sup> In	0.147 $\beta^-$	2.88	1.76	1.37	6.01	80	60
<sup>204</sup> Tl	0.242 $\beta^-$	1.95	0.85	0.50	3.30	150	100
<sup>143</sup> Pr	0.307 $\beta^-$	12.40	4.86	2.38	19.64	25	20
<sup>90</sup> Y	0.927 $\beta^-$	65.90	18.50	4.59	88.99	6	3

that given by the ICRP is 2 for the high energy  $\beta^-$  emitter <sup>90</sup>Y, but the ratio falls towards lower energies and is about 0.5 for <sup>171</sup>Tm. This trend is also in line with the data of Fig. 2 because, whereas the curve for  $\bar{D}_M$  is now almost independent of  $\beta^-$ -particle energy, the curves for the surface dose parameters rise steeply at low energies.

#### Results for <sup>226</sup>Ra

Dosimetric data are also available for <sup>226</sup>Ra uniformly distributed in bone<sup>18</sup> and risk values have also been calculated for this radionuclide and its retained daughter products. The results are given in the last section of Table 2. Risk factors appropriate to <sup>226</sup>Ra have been derived from the linear extrapolation given in the report of the U. S. Advisory Committee on the Biological Effects of Ionizing Radiations (BEIR Report 1972)<sup>19</sup>.

In this Report, 48 cases of osteogenic sarcoma are recorded in a group of 775 persons subjected to a mean dose of 1700 rad, i.e. a tumour rate of 36 cases/ $10^6$  for a dose of 1 rad to the bone mass. Because the dose to trabecular endosteal tissues is about one third of the dose to bone (and only a few tumours have been identified as originating in the cortical mid-thirds of long bones<sup>13</sup>) the risk rate for osteogenic sarcomas can be put at  $105/10^6/\text{yr}$  for a dose rate of 1 rad/yr. The value of  $r_0$  can then be taken as  $70/10^6/\text{yr}$  and  $r'_0$  as  $35/10^6/\text{yr}$ . The radium case data, reviewed in the BEIR Report, also include a further 20 cases of carcinoma of air sinuses for which the risk rate can be similarly deduced as  $40/10^6/\text{yr}$  for a dose rate of 1 rad/yr to a layer of tissue adjacent to bone of the air sinus cavities. The dose factor for this situation will be approximately the same as for the trabecular surfaces and so this risk can be added to the value of  $r_0$  for calculation purposes. No data are available for the incidence rate of angiosarcoma etc., except that these tumours seem to have occurred very rarely in radium cases; no value has been given therefore to the factor  $r_a$ . Only a nominal value of 200 has been given to the bone marrow risk factor  $r_m$  because, as can be seen from Table 2 the very low dose to bone marrow makes the total bone risk insensitive to this factor. The nominal value of 200 was taken as 5 times the value used for  $\beta$  emitters because the risk factor for osteogenic sarcoma,  $105/10^6/\text{yr}$ , was about 5 times greater than the value of  $20/10^6/\text{yr}$  chosen for  $\beta$  emitters in Eq. (4). The value of the total risk for a volume-seeking  $\alpha$  emitter is then given by:

$$R_T = (200\bar{D}_M + 110\bar{D}_S + 35\bar{D}'_S)/10^6/\text{yr} \quad (10)$$

In calculating the value  $q$  for the MPBB, the same value of  $R_{MPL}$  is used and a small contribution is added for the  $^{226}\text{Ra}$   $\beta$  particles, according to Eq. 8. The result,  $q = 0.14 \mu\text{Ci}$  is surprisingly close to the ICRP value, based on quite other considerations.

### Conclusions

This paper attempts to calculate maximum permissible body burdens for a number of  $\beta$  emitters that are deposited in bone either throughout the bone matrix as volume-seekers or on bone surfaces as surface-seekers. The principles are different from those used hitherto, in that the dose parameters are the absorbed doses in rad to the relevant tissues and risk parameters are derived from the commonly accepted linear extrapolations of available human data on tumour incidence, combined with incidence ratios for other tumours from animal investigations. The values of MPBB so obtained are higher than present ICRP values, but not so much higher as to rule the method out of court. Concordance with ICRP values for high energy  $\beta$  emitters could be obtained by altering the choice of risk corresponding to that for occupational exposure. If this were done then at low  $\beta$ -particle energies, the new calculations would generally give higher values for the MPBB for the volume-seekers and lower values for the surface-seekers, compared with present ICRP values. A more formal approach to the problem would be to use the dosimetric data in a similar manner, but simply choose the risk factors to follow ICRP dose levels, i.e. put the total risk for bone tumours at one third of that adopted for the bone marrow risk, - in accordance with a dose of 5 rem to bone marrow and 15 rem to a single organ. It is suggested, however, that the method used in this paper is preferable and the concordance obtained when the method is applied to  $^{226}\text{Ra}$  gives encouragement that eventually all the bone-seeking radionuclides may be treated in this way, without recourse either to a relative damage factor or to an overt specification of a quality factor.

### References

1. Joan R. Whitwell and F. W. Spiers, in Proceedings of the Fifth Congress of the French Society for Radioprotection, Grenoble, (1971).
2. F. W. Spiers, G. D. Zanelli, P. J. Darley, J. R. Whitwell and M. Goldman, in Biomedical Implications of Radiostrontium Exposure, Eds. M. Goldman and L. K. Bustad, AEC Symposium Series 25, CONF-710201, 130, (1972).
3. Joan R. Whitwell and F. W. Spiers, unpublished data.
4. International Commission on Radiological Protection, ICRP Publication 11, Pergamon Press, Oxford, England, (1968).
5. International Commission on Radiological Protection, ICRP Publication 14, Pergamon Press, Oxford, England, (1969).
6. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), Official Records of the General Assembly, Supplement 25 (A/8725), United Nations, New York, (1972).
7. J. F. Loutit and Janet M. Vaughan, Brit. J. Radiol., 44, 815, (1971).
8. H. Sissons, Medical Research Council, London, Sub-Committee on Permissible Levels, PIRC/PL/70/4, (1970).
9. J. Vaughan, Medical Research Council, London, Sub-Committee on Permissible Levels, PIRC/PL/70/4, (1970).
10. International Commission on Radiological Protection, ICRP Publication 2, Pergamon Press, New York, USA, (1959).
11. R. R. Pool, R. J. R. Williams and M. Goldman, in Biomedical Implications of Radiostrontium Exposure, Eds. M. Goldman and L. K. Bustad, AEC Symposium Series, CONF-710201, 277, (1972), and D. L. Dungworth, M. Goldman, J. W. Switzer and D. H. McKelvie, Blood, 34, 610, (1969).
12. W. J. Clarke, R. H. Busch, P. L. Hackett, E. B. Howard, M. E. Frazier, B. J. McClanahan, H. A. Ragan and G. S. Vagt, in Biomedical Implications of Radiostrontium Exposure, Eds. M. Goldman and L. K. Bustad, AEC Symposium Series, CONF-710201, 242, (1972).
13. C. W. Mays, personal communication.
14. M. P. Finkel, B. O. Biskis, I. Greco and R. W. Camden in Biomedical Implications of Radiostrontium Exposure, Eds. M. Goldman and L. K. Bustad, AEC Symposium Series, CONF-710201, 285, (1972).
15. A. Nilsson, Acta Radiol., 9, 155, (1970).
16. B. E. Roberts, D. W. Miles and C. G. Woods, Brit. J. Haemat., 16, 75, (1969).
17. F. C. McLean and A. M. Budy, Radiation, Isotopes and Bone, Academic Press, New York, USA, 34-35, (1964).
18. Joan R. Whitwell and F. W. Spiers, unpublished data.
19. Report of the Advisory Committee on the Biological Effects of Ionizing Radiations, National Academy of Sciences, National Research Council, Washington, D.C., 128, (1972).

## A THEORY OF RADIATION RISK BASED ON MICRODOSIMETRY

Roger D. Caldwell, Ph.D.  
University of Pittsburgh  
Pittsburgh, Pennsylvania 15261

### Abstract

This paper demonstrates how microdosimetry can be applied to a theory of radiation risk. Many recent radiobiological experiments show that the relative biological effect (RBE) between two different radiations increases as the dose decreases. The historically developed quantities of radiation dosimetry, the absorbed dose and the dose equivalent, fail to account for this phenomenon. Also, because the absorbed dose is an averaged quantity, it provides no physical basis for other than linear extrapolation of biological data to the small radiation exposures encountered in radiation protection. Microdosimetry suggests that a consistent theory of radiation risk can not be linear for all radiations.

Several models which fit the distributions of microdosimetry to biological data are possible. Because none of them can be linear, it is necessary to process the individual datum of a microdosimetric distribution in order to produce an index of risk. As a demonstration of the feasibility of the procedure, a Rossi-type tissue equivalent proportional counter was interconnected with a PDP-8I computer and exposed to several different types of radiation. The results show that single-valued indices of radiation risk can be directly measured and, thus, one of the hurdles to the application of microdosimetry to radiation protection appears to be solvable.

### Introduction

Perhaps the central issue in radiation protection is how biological effect data obtained at high doses should be extrapolated to the low dose range of maximum permissible limits and radiation protection guides. If the important biological effects of radiation occurred with statistically sufficient frequency at the doses of interest, it would be possible to investigate the dose-effect relation experimentally. However, the important effects, such as the induction of neoplastic disease and genetic mutation, occur with low probability even at high dose rates.<sup>1</sup> It has not been possible to date to reliably estimate the shape of the dose-effect curve below dose levels at least ten times greater than present guide lines. Furthermore, as Rossi<sup>2</sup> has pointed out, the experimental approach is usually self-defeating. There is an understandable tendency to reduce permissible doses well below the level where biological effects can be observed. Consequently, the radiobiological experimenter is always pursuing the retreating mirage of lower dose effects. It is therefore clear that extrapolations must be made on some theoretical basis.

Most extrapolations to the low dose range have a biological basis. Dose response schemes<sup>1,3</sup> which make use of thresholds are based on the accepted notions of biological repair processes and sublesion damage. The well-known sigmoid LD<sub>50</sub> curve for experimental whole-body irradiation of mammals is a simple example.

Linear and curvilinear relationships have been proposed<sup>1,3</sup> which are derived from physical and molecular considerations. At sufficiently low doses and dose rates the linear model has been attractive to many workers because the spatial and temporal separation of ionizing particles is large enough so that effects are caused principally by single tracks through cells. Since interactions between tracks in this case would be negligible and the density of tracks is linear with dose, a linear dose-effect relationship is a necessary result. The major difficulty with this reasoning is that the estimates of risk, which are linearly extrapolated to low doses are based on biomedical evidence obtained at high doses where, for low LET radiations, there are multiple traversals of ionizing electrons through cells and cell nuclei.

The linear non-threshold hypothesis has been used in radiation protection standards because it is easy to apply, gives clear-cut estimates of risk and is generally thought to be conservative. However, Baum<sup>4</sup> has presented evidence that linear extrapolations are not necessarily conservative for heterogeneous human populations. He shows how a dose-effect relation represented by a simple power function of dose with an exponent of less than one can occur when subdivisions of a population have differing sensitivities. An important result of a power function with an exponent less than one is that the effectiveness per rad is greater at low doses than at high doses. However, Baum assumes a linear dose-effect relation for each of his subdivisions of the considered population with different thresholds for each subdivision, and consequently, even in his hypothesis, the linear dose-effect relation is fundamental.

Inherent in existing radiation protection guides is the assumption of linearity of effect with dose for all radiations. This is a consequence of a constant quality factor for all doses of a given radiation. But, increasing evidence shows that the relative biological effect (RBE) of any type of radiation is not a constant, but increases with decreasing dose. Furthermore, consideration of microdosimetry data suggests that the risk of effect can not be linear for all radiations. For example, the dose-effect curves for neutron and gamma radiation cannot both be linear. If the curve is linear for neutrons, it cannot be linear for gamma radiation over the entire dose range of biological interest.

This paper presents the biological and physical evidence that supports linearity of dose-effect for densely ionizing radiations (i.e., high LET-radiation) and decreasing effectiveness with decreasing doses of sparsely ionizing radiations (i.e., low-LET radiation). It also reviews the potential uses of microdosimetry in radiation protection and discusses how microdosimetric indices of risk may be measured directly.

#### RBE as a Function of Dose

The ICRP<sup>5,6</sup> originally intended the LET-QF relation to follow the LET-RBE relation. But this intention was shown to be in error when results of track segment experiments began to appear. In these experiments<sup>7,8,9</sup> accelerated heavy particles pass through monolayers of cells. By adjusting the amount of absorber in front of the target cells, the experimenter could select the portion of the heavy particle track (and thus the LET) he wished for irradiation. These experiments, pioneered by Barendsen, are ideally designed to test the

efficacy of LET as a quality factor. The results offered little support for the LET-QF relation. RBE-LET curves differed from one biological system to another. RBE did not always increase with increasing LET; for lower organisms, the RBE decreased with high LET radiation. For mammalian cell killing the RBE does increase at high LET, but not linearly. Furthermore, the RBE-LET curve for cell killing goes through a maximum at about 100 keV/micron and falls off at higher LET values. Similar experiments with chromosome abnormalities as an end point<sup>10</sup> showed a similar RBE-LET curve with a maximum.

By itself, the RBE-LET track segment evidence only appears to place the QF conservatively high as a function of LET. This is not a serious problem, since the risk from radiation exposure would never be under estimated. However, the difficulty with constant quality factors became more significant when a number of radiobiological experiments indicated that RBE was a function of dose over the whole range of LET. Barendsen's now classic, track segment experiments were the first to show this. The peak RBE had a value of 7.5 for 80% cell survival, 4.5 for 20% survival, 3.5 for 5% survival and 3.0 for 1% survival. Neither the shape of the curves or the location of the maxima (110 keV/micron) vary, but the RBE increases for all values of LET when the survival increases (and thus when the dose decreases).

Barendsen's finding on the dose-dependence of the RBE was soon supported by other workers. Bateman,<sup>11</sup> measuring opacities in mouse eye lens for neutrons of different energies, also found the RBE varying inversely with dose. The slope of the RBE-LET curve increases as the neutron energy is decreased from 14.3 MeV to a maximum at about 340 keV and then decreases for lower energies. The dependence of RBE on dose was extended to the induction of mammary cancer in Sprague-Dawley rats by Vogel<sup>12</sup> and Shellabarger.<sup>13</sup> An apparent difference in their results can be attributed to differences in neutron energy spectrum.<sup>13</sup> Otherwise, their results are consistent with those of other workers for cell killing, chromosome aberrations and cataracts. The same increase in RBE occurs as the dose is decreased and the slope of the RBE-dose curve for mammary cancer data on a log-log plot is similar to and consistent with the data on other effects. The significance of Vogel's and Shellabarger's data cannot be over emphasized. Neoplastic disease constitutes the major concern of those setting radiation protection standards. Cell killing and chromosome aberrations clearly originate with damage within a single cell, while cancer can not so readily be traced to an alteration of a single cell. The similar RBE-dose kinetics for cell killing and mammary cancer suggest that neoplastic disease may also originate within a single cell. Even if this simplification does not actually occur, the results may still be used in an empiric fashion so that the impact on radiation protection will remain in any case. Kellerer and Rossi<sup>14</sup> have summarized the available data which show the RBE-dose dependence. They used the data to support an hypothesis that biological effects originate in cellular lesions which are produced by one high LET particle but which require at least two electrons. Recently, Hall, et. al.,<sup>15</sup> have reported exhaustive studies on the RBE-dose dependence for effects on *Vicia faba*. These studies completely confirm all aspects of the other studies quoted.

What is to be made of these data? They show that RBE is unity, or close to it, at high doses over the range of LET. Then for a variety of effects (cell survival, lens opacification and cancer induction) as the dose decreases, the RBE begins to increase, at different rates for radiations of different LET. Does the RBE increase indefinitely at lower doses? If so, what does that mean for radiation protection? Would this imply drastically lowered limits for densely ionizing radiations? Or does one re-evaluate limits for x-and gamma radiation?

A report by Sparrow, et. al.<sup>16</sup> suggests a possible resolution of this problem. They obtained dose-response curves for somatic mutation in *Tradescantia* stamen hairs exposed to 0.43 MeV neutrons and 250 kvp x-rays. All the neutron data fit a linear plot. The x-ray data was curvilinear down to five rads. From 5 to 100 rads the RBE (neutron/gamma) decreased with increasing dose from about 50 to about 15. Below x-ray doses of 5 rads, the RBE remained constant because the x-ray data were linear. Both the neutron and gamma curves peak and decline at higher doses so that the RBE does not approach unity as other RBE-dose data. The study shows, admittedly for a non-mammalian cell, but nevertheless for a eucaryotic cell, that the RBE does not increase indefinitely. Furthermore, since the neutron response is linear throughout the range, while the x-ray response is only linear at very low doses, it appears that x-rays are less effective per rad at low doses.

#### Some Concepts of Microdosimetry

If the biological effectiveness between different radiations did not vary with dose, the absorbed dose, and with an appropriate quality factor, the dose equivalent, would serve the purpose of radiation protection without difficulty. Since the dose-dependence of RBE is now well established, it is apparent that the physical parameters now used for the estimation of risk, the absorbed dose and linear energy transfer, need to be re-examined.

When a small animal, e.g., a mouse, is irradiated, the absorbed dose is constant over the whole animal (neglecting, of course, the differences in atomic composition and density of bone, fat, etc.). This constancy of dose remains for any tissue mass larger than several milligrams. Thus, if our mouse received 100 rads, every gram absorbs 10,000 ergs, every 100 milligrams very close to 1000 ergs and every 10 milligrams about 100 ergs. But when the sample of mouse gets down to the size of the cell (micrograms), or the cell nucleus (tenths of ug), the energy density (E/m) will seldom equal the absorbed dose. This results from the discontinuous nature of radiation energy deposition; matter exposed to ionizing radiation receives its energy from charged particles which dissipate their kinetic energy in discrete tracks of ionization and excitation. While a gram of tissue may be traversed by a vast number of charged particles when absorbing a dose of one rad, a cell nucleus in that gram of tissue might be traversed by only a few particles, or even none at all. Since it is widely supposed that biological effects originate in cells, it would seem prudent to investigate the deposition of energy on a microscopic scale. Any given amount of energy absorbed in a material will, if the sample size is small enough, produce distributions of microscopic values of the energy density. The study of these microscopic distributions has come to be called microdosimetry.

Historically, microdosimetry originated with Rossi's use of tissue equivalent, spherical proportional counters to determine LET distributions.<sup>17</sup> He soon realized that the proportional counter spectra were interesting in themselves.<sup>18,19</sup> The counter, operating with tissue equivalent gas at pressures of a few millimeters of mercury, could simulate very small volumes of tissue. For example, a 7.5 inch counter can simulate a volume of effective diameter of 6 microns when operated at 40 mm Hg, 3 microns at 20 mm Hg and 1.5 microns at 10 mm Hg. In each case the mass of the gas is equal to that of the small simulated tissue sphere at unit density. The practical limit to which this technique has been pushed is about 0.1 micron.<sup>20</sup>

Of course there is a magnification of the particle fluence in the simulated sphere because the number of particles crossing the cavity is proportional to the cross-sectional area of the cavity. Corrections for this effect and other technical details have been extensively discussed by Rossi.<sup>21</sup>



Pulses can be sorted as they are produced in the counter (i.e., sent directly to a pulse height analyzer) or collected for a given time and then sorted. In the first case, a distribution of single events is generated, and in the second case, distributions of multiple events occur.

When a charged particle traverses the proportional counter and is detected, the process is called an event. Hopefully, this experimental event mimics the microscopic event, the passage of a charged particle through a biological structure. To determine the magnitude of an event, the size of the biological structure must be specified. The quantity of energy,  $E$ , deposited in a single event, divided by the microscopic diameter was first called the event size,  $Y$ , and later redefined as the lineal energy density,  $y$ .<sup>22</sup>

Single event distributions do not have to be limited to the lineal energy density. The event energy can also be divided by the volume or mass of the biological structure. The ratio of  $E/m$  has been defined as the specific energy,  $z$ .<sup>22</sup>  $f_1(z)$  is the differential distribution of single events in specific energy  $z$ .

Another single event distribution is the event frequency,  $\phi(y)$ , the mean number of events with size in excess of  $y$  per unit absorbed dose.  $\phi(0)$  is the frequency of events of all sizes per unit absorbed dose.

Single event distributions are unique for each type of radiation. The range of magnitude included in the distribution is a characteristic of each radiation type. For example,  $^{60}\text{Co}$  radiation produces an  $f_1(z)$  distribution in one micron spheres which includes events from 1 to 200 rads, while Pu-Be neutrons produce events from 80 to 7000 rads. Single event distributions increasingly overlap as the sample size is increased and increasingly separate as the sample size is decreased. The ability to "fingerprint" the radiation type had led to attempts, unsuccessful to date, to use the single event distributions of microdosimetry as an index of radiation quality.

It is widely accepted that energy is responsible for biological changes. Work must be done on a biological system to cause any change. This acceptance of energy's role accounts for the fundamental position that the absorbed dose holds in radiation science.

Microdosimetry does not abdicate the fundamental concept of energy input into a biological system as being a necessary condition for radiation effect. Rather, it requires that the energy input be known with more detail. Instead of the energy input to a body organ, averaged over the whole organ, the question asked is: what is the distribution of energy inputs to the microscopic structures in the cell?

The absorbed dose is a single valued quantity, while the specific energy,  $z$ , is random valued and must always be known in its complete distribution. The absorbed dose is thus the mean value of the differential distribution of  $z$ . The symbol for the  $z$  distribution is  $f(z;D)$  indicating that the distribution is also a function of absorbed dose.

The event frequency must exceed one for any sphere size, provided the dose is high enough. The smaller the sphere size or the higher the LET, the greater the dose must be for multiple events to occur. It also follows that for any combination of radiation type and sphere size that the event frequency can be less than one. This means that no event occurs in some fraction of the samples. When this zero component is large, the  $f(z;D)$  distribution is nearly equivalent to the  $f_1(z)$  distribution. All energy depositions come from single events in this case. This occurs at low doses, but at different absolute absorbed

doses for different radiations. Generally, densely ionizing radiations, such as alphas, protons and neutrons, produce single event distributions in the absorbed dose range of interest for radiation protection. Electron and photon radiation usually produces multiple event distributions. At very low doses, or also with very small sphere sizes, electrons and photons can produce single event energy density distributions. The dose below which this occurs for 250 kVp x-rays in one micron spheres is 20 rad, in 3.5 micron spheres two rads. For the same spheres, 0.34 MeV neutrons produce single event distributions up to 1430 rads and 143 rads, respectively.

Comparing specific energy distributions for various doses of low LET and high LET radiations shows that high LET spectra increase in area with increasing dose but do not shift in value because multiple events have low probability. Low LET radiation spectra on the other hand show strong shifts up the scale with increasing dose because of multiple events. At sufficiently high doses the low LET and high LET spectra overlap completely. It's this phenomenon that appears to account for the dependence of the RBE on dose.

#### The Microdosimetric Implications of the Absorbed Dose

As a single valued, averaged quantity, the absorbed dose concept assumes that ionizations are distributed homogeneously and randomly throughout the irradiated material. As a consequence, when the dose is lowered, the concentration of ionizations is assumed to be lowered. Each ionization would, in this case, have the same probability of contributing risk at low doses that it did at high doses. Thus, only linear extrapolation is possible as long as the absorbed dose is believed to be fundamental.

Of course, ionizations are not randomly distributed in irradiated material. They are associated with ionizing particles which are randomly distributed. Recognizing the reality of microscopic energy distributions, the ICRU<sup>23</sup> provided an alternative definition of the absorbed dose:

$$D = \bar{z} = \int_0^{\infty} z f(z) dz$$

If the absorbed dose were the correct quantity with which to correlate biological effect, then this non-uniform deposition of energy in biological structures must be reflected logically and consistently in the biological effects of radiation. Since effects originate within cells, then the absorbed dose concept presumes that the probability of an effect occurring in a cell is linearly related to the energy density in the cell. However, if this were true, there would be no RBE between different radiations and no RBE dependence on dose. Furthermore, the results of track segment experiments cited earlier are consistent with a linear z-effect relation. These experiments show that a limit of effectiveness is reached with increasing energy density. More important, the effectiveness per rad falls off rapidly with decreasing energy density.

The deficiencies of the absorbed dose are not fully compensated for by multiplication with LET-related quality factors. Because of the RBE dependence on dose, it is clear that a non-linear function must be found to weight the energy density distribution.

## A Model for the Extrapolation of Risk to Low Doses

Consider two types of radiation which have quite different  $f_1(z)$  distributions, say fast neutrons and gamma radiation. Recall that the  $f(z;D)$  distribution for fast neutrons was similar to the  $f_1(z)$  distribution over most of the dose range of biological interest. That is, the  $z$  values remain constant with dose, and only the probability of each  $z$  increases with increasing dose. Because the area under the  $zf(z;D)$  distribution in this case is directly proportional to the dose, you would expect the dose effect curve to be linear over that range of dose, no matter what the  $z$ -effect function was. The  $f(z;D)$  distribution for gamma radiation can also approximate its  $f_1(z)$  distribution, but at much smaller doses. At very small doses, the gamma dose effect curve would be linear but reduced in effectiveness per rad, provided that the  $z$ -effect function had values greater than zero within the range of values included in the  $f_1(z)$  distribution for gamma radiation. This is likely to be the case since the  $f_1(z)$  distributions for fast neutrons and gamma radiation overlap for sphere sizes such as the cell nucleus and since all fast neutron events appear to be effective in cell killing experiments and to be linearly proportional for cataract and cancer induction. For an absolute dose threshold to exist, an effective energy density threshold in the particular cellular target, e.g. the cell nucleus, must occur.

A great deal of evidence exists to show that the cell nucleus is the appropriate sphere size for microdosimetry. For example, the cell killing experiments with track segments show probability cross-sections which are geometrically equivalent to the cell nucleus cross-section. Other evidence includes kinetic analysis by Kellerer and Rossi.<sup>24</sup> A typical diameter for a mammalian cell nucleus is 3.5 microns. For a sphere of this diameter there is considerable overlap of  $f_1(z)$  distributions for low LET and high LET radiations. To provide enough separation to account for the RBE's of 50 or more which appear to be reached at low doses, it is necessary to assume very diminished effectiveness for low values of specific energy, so that an effective threshold of energy density in the mammalian cell nucleus appears to be required for biological effect.

The significance of this model is far reaching. It demonstrates that it is not physically or biologically feasible to extrapolate linearly from high doses of gamma or x-radiation to low doses. It also gives support to the conclusion that risk for low doses of radiation may be significantly over estimated.

Two of the important features of the extrapolation need numerical values: Firstly, the dose at which the RBE becomes constant is the dose where the reference radiation, 250 kVp x-rays, becomes a single event distribution. For target volumes 3.5 microns in diameter single event distribution occur at doses less than 2 rads. Secondly, if one made use of Kellerer and Rossi's equation for the RBE,<sup>14</sup> one would project a maximum RBE of about 400. This maximum RBE, because it occurs at low doses, is of obvious significance for estimating risk to populations. This value of the RBE should not be used to reduce the neutron fluence to which a population should be exposed. Rather, since electron and photon radiation is so much less efficient than previously assumed from linear extrapolations from high doses, the permissible photon fluence may be increased substantially.

## The Direct Measurement of Indices of Risk

If, as suggested here, the measurement of cell nuclei energy density distributions is important, it will become necessary to perform on-line processing of the large amount of data contained in a microdosimetric

distribution. Conceptually, this offers no barrier. Miltenberger<sup>25</sup> has interconnected a TE proportional counter to a PDP-8I computer and obtained single-valued indices of risk from a variety of radiations and doses. There are, however, some difficult obstacles, not the least of which is the fluence magnification mentioned earlier. In a sense the dose reduction phenomenon, caused by the density differences in the TE gas and counter wall allows accumulation of microdosimetric distributions within reasonable times. However, high intensity fields may not be measureable since the counting time, already below the millisecond range for low dose rates, will be too short even for electronic timing. Because the technique is most important for low dose levels, this fundamental deficiency may not inhibit the practical use of the technique.

The procedure used by Miltenberger was as follows: The TE proportional counter was connected to a pulse amplifier. From there the amplified pulse was directed to an analog to digital converter. The numerical value from the converter was then fed into a Digital PDP-8I computer. The numerical value was then weighted by the computer according to the function assumed and added as an increment of risk to the sum index.

It should be possible to develop analog circuits to perform the weighting function and thus allow the eventual development of a reasonably sized and priced instrument.

#### References

1. Report of the Advisory Committee on the Biological Effect of Ionizing Radiations, The Effects on Populations of Exposure to Low-Levels of Ionizing Radiation, National Academy of Sciences/National Research Council, Washington, D.C. 20006, November, 1972.
2. Rossi, H.H., The Effects of Small Doses of Ionizing Radiation, Phys. Med. Biol., Vol. 15, No. 2, pp. 255-262, 1970.
3. International Commission on Radiological Protection, Publication 14, Radiosensitivity and Spatial Distribution of Dose, Oxford, Pergamon Press, 1969.
4. Baum, J.W., Population Heterogeneity Hypothesis on Radiation Induced Cancer, Health Physics, Vol. 25, pp. 97-104, 1973.
5. International Commission on Radiological Protection, Report of the RBE Committee to the International Commissions on Radiological Protection and on Radiological Units and Measurements, Health Physics Vol. 9, p. 357, 1963.
6. International Commission on Radiological Protection, Recommendation of the International Commission on Radiological Protection, 1965, ICRP Publication 9, Pergamon Press, Oxford, 1966.
7. Barendsen, G.W., Impairment of the Proliferative Capacity of Human Cells in Culture by Particles with Differing Linear-energy Transfer, Int. J. Rad. Biol. Vol. 5, p. 453, 1964.
8. Barendsen, G.W., Relative Biological Effectiveness as a Function of Linear Energy Transfer, Proceeding of the First Symposium on Microdosimetry, Ispra (Italy), Euratom, 1967.

9. Todd, P.W., Heavy-Ion Irradiation of Cultured Human, Rad. Res. Supp. 7, p. 196, 1967.
10. Skarsgard, L.D., B.A. Kihlman, C.M. Pujara and S. Richardson, Survival, chromosome abnormalities and Recovery in Heavy-Ion and X-irradiated Mammalian Cells, Rad. Res. Supp. 7, p. 208, 1967.
11. Bateman, J.L. and M.R.S. Snead, Current Research in Neutron RBE on Mouse Lens Opacity, in Symposium on Neutrons in Radiobiology, CONF-691106, 1969.
12. Vogel, H.H. and R. Zaldiver, Experimental Mammary Neoplasms: A Comparison of Effectiveness between Neutrons, x- and Gamma-radiation, in Symposium on Neutrons in Radiobiology, CONF-691106, 1969.
13. Shellabarger, C.J., Unpublished data, given in Radiation Carcinogenesis at Low Doses by H.H. Rossi and A. Kellerer, Science, Vol. 175, pp. 200-202, 14 Jan. 1972.
14. Kellerer, A.M., and H.H. Rossi, RBE and the Primary Mechanism of Radiation Action, Rad. Res. Vol. 47, pp. 15-34, 1971.
15. Hall, E.J., H.H. Rossi, A.M. Kellerer, L. Goodman and S. Marino, Radiobiological Studies with Monoenergetic Neutrons, Rad. Res. Vol. 54, pp. 431-443, 1973.
16. Sparrow, A.H., A.G. Unterbrink and H.H. Rossi, Mutations Induced in Tradescantia by Small Doses of X-rays and Neutrons: Analysis of Dose-Response Curves, Science, Vol. 176, pp. 916-918, 26 May, 1972.
17. Rossi, H.H. and W. Rosenzweig, Advice for the Measurement of Dose as a Function of Specific Ionization, Radiology, Vol. 64, pp. 404-410, 1955.
18. Rossi, H.H., Specification of Radiation Quality, Rad. Res., Vol. 10, p. 522, 1959.
19. Rossi, H.H., M.H. Biavati and W. Gross, Local Energy Density in Irradiated Tissues. I. Radiobiological Significance, Rad. Res. Vol. 15, p. 431, 1961.
20. Rossi, H.H., Experimental Limitations of Microdosimetry, Second Symposium on Microdosimetry, Stresa, Italy, EUR 4452.
21. Rossi, H.H., Microscopic Energy Distributions in Irradiated Matter in Radiation Dosimetry, Volume 1, F.H. Attix and W.C. Roesch, eds., Academic Press, 1967.
22. Rossi, H.H. and A.M. Kellerer, Summary of Quantities and Functions Employed in Microdosimetry, Second Symposium on Microdosimetry, Stresa, Italy, EUR 4452.
23. International Commission on Radiological Units and Measurements, Radiation Quantities and Units, ICRU Report 19, Washington, D.C., 1971.
24. Kellerer, A.M., and H.H. Rossi, The Theory of Dual Radiation Action, Current Topics in Radiation Research, Volume 8, No. 2, October, 1972.
25. Miltenberger, R.P., Measurement of the Microdosimetric Quantities  $\bar{z}_e$  and  $\phi(z)$ , Masters Thesis, University of Pittsburgh, Graduate School of Public Health, 1973.

A COMMON MOLECULAR MECHANISM IN RADIOBIOLOGY  
ITS IMPLICATIONS IN RADIOLOGICAL PROTECTION

K.H. Chadwick and H.P. Leenhouts.  
Association Euratom-ITAL, Postbus 48,  
Wageningen, The Netherlands.

Abstract

The paper sketches the development of a theoretical model which attempts to describe the biological action of ionizing radiation starting from a molecular mechanism through the cellular effects to the effects occurring in animals. The model is based on the action of radiation on the DNA molecule in the cell and it is assumed that radiation induced DNA double strand breaks are the most critical lesions in cellular radiobiology.

In the first part of the paper the molecular mechanism of DNA double strand breakage is described and related to cellular effects such as radiation induced cell death and radiation induced mutations. Evidence is presented to show that cell death, mutations and DNA double strand breaks all have the same dose kinetics.

The effects of dose rate and LET on DNA double strand breakage and on cell death and mutation induction are considered and the original equations can be modified to give the form expected at low dose and low dose rate. Using the basic equations a limiting relative biological effectiveness is defined which is dose and dose rate independent and is particularly relevant in radiological protection.

In the second part of the paper the cellular effects are tentatively related to effects occurring in animals and organs such as animal survival and radiation induced cancer. The theoretical equations are compared with experimental data in the literature.

The consequence of the mechanism for the genetic effects of radiation are briefly discussed with regard to the radiation dose kinetics.

It is concluded that the model gives dose relationships which are non-linear but contains a means of extrapolating from high dose, high dose rate results to obtain parameters which could be of importance for radiological protection. It suggests that a basic mechanism may be common to the various radiobiological effects and to all eukaryotic cells and it would thus provide a basis for the extrapolation of results in animals to man. The model also provides a common link between the biochemistry of DNA, the metabolism of the cell, radiobiology and radiological protection.

Introduction

Recently a molecular theory of cell survival has been proposed<sup>1</sup> which attempts to explain the dose relationship of cell survival following ionizing radiation on the assumption that radiation induced DNA double strand breaks are the most critical radiation damage. The application of this molecular model to the dose rate effect and the analysis of the survival of cells synchronized at different phases of the cell cycle has given results which can be interpreted on the basis of the biochemistry and behaviour of the DNA molecules in the cell.

It is somewhat surprising that if the molecular model is extended to describe radiation effects on more complex systems certain interesting correlations can be found which indicate that a common molecular mechanism may be at the root of these radiation effects.

The model is still in its infancy, further developments and refinements are necessary and it is clear that more experimental work must be carried out to correlate the molecular mechanism more closely with the observed effect. However, an attempt will be made here to sketch the relationship of the various radiation induced effects via the induction of the common primary damage, to show that these dose relationships are compatible with experimental results and to indicate that the interpretation of the relationships has a biological significance. Finally, the implications of the molecular model for radiological protection are inferred.

### The proposed Molecular Mechanism

It is generally, though not universally, accepted that the DNA molecule is the most critical target for radiation. The DNA occurs in the nucleus of the cell as chromosomal DNA in the form of a long double helix structure and the induction of single strand breaks and double strand breaks in the DNA helix by radiation is a well documented scientific fact<sup>2,3</sup>.

The basic assumption of the theoretical model presented in this paper is that the double strand break is the critical lesion which eventually leads to the biological end effects.

Radiation can induce a double strand break in the DNA double helix in two ways

1. a double strand break in one radiation event,
2. a double strand break as a result of the combination of two independent breaks from two independent radiation events.

Following a radiation dose  $D$  the number of double strand breaks produced in one radiation event in a cell is proportional to dose, say  $\alpha D$ . The number of double strand breaks produced in two independent radiation events in a cell is proportional to the square of the dose, say  $\beta D^2$ . Thus the total number of DNA double strand breaks in a cell is given by

$$N = \alpha D + \beta D^2. \quad (1)$$

In the original derivation of this expression<sup>1</sup> the coefficients  $\alpha$  and  $\beta$  contain parameters which have a radiobiological significance and it is important that in the analysis of the experiments this radiobiological significance be borne in mind.

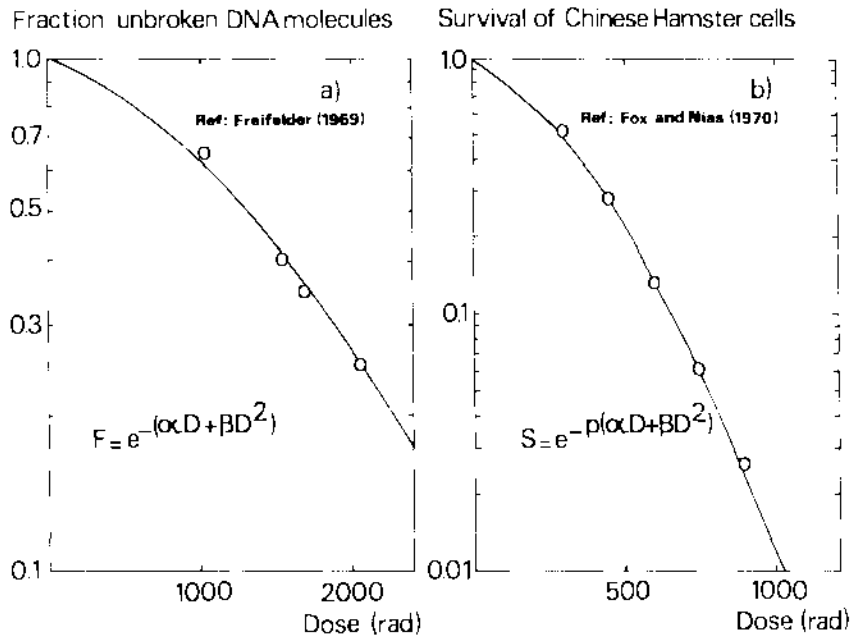
### Cellular Effects

#### Cell Survival

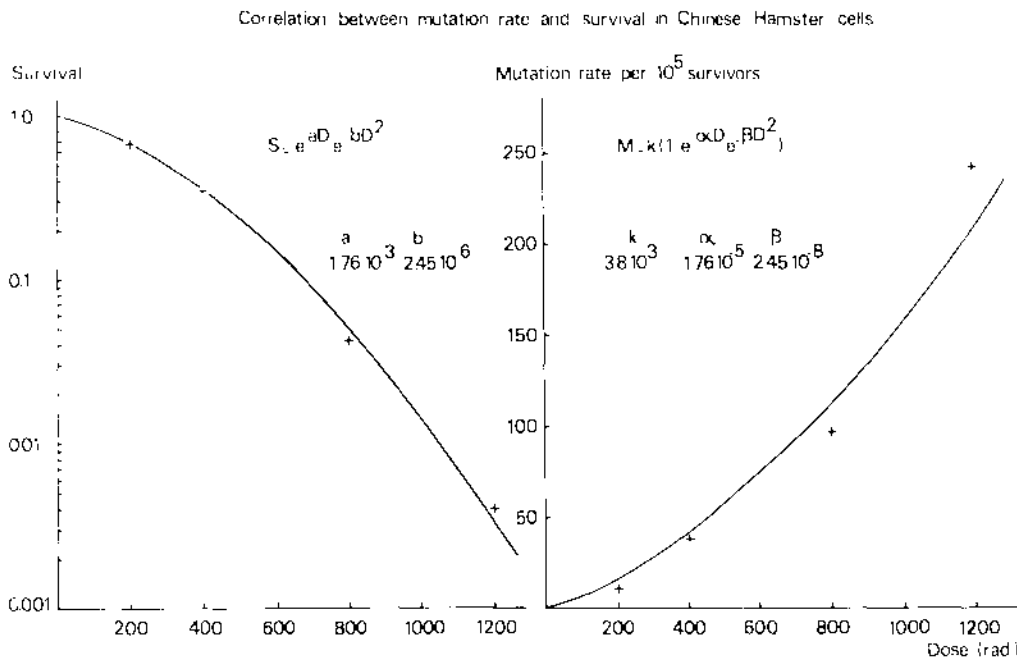
In a population of cells the number of DNA double strand breaks  $N$  will be the mean value of a poissonian distribution. If each double strand break has a probability  $p$  of leading to cell reproductive death, then poissonian statistics can be used to derive the probability that  $N$  double strand breaks lead to cell reproductive death and cell survival is given by

$$S = e^{-pN} = e^{-p(\alpha D + \beta D^2)} \quad (2)$$

In figure 1 the induction of DNA double strand breaks and cell survival are shown analysed according to the model presented here. The figure illustrates that the dose kinetics are the same. If the coefficients are calculated to give the chance for a double strand break per nucleotide pair then the magnitude of the coefficients is also very similar ( $\alpha^1 = 1.6 \cdot 10^{-12}$ ,  $\alpha^1 p^1 = 5.3 \cdot 10^{-13} \text{ rad}^{-1}$ ;  $\beta^1 = 1.6 \cdot 10^{-15}$ ,  $\alpha^1 \beta^1 = 9.3 \cdot 10^{-16} \text{ rad}^{-2}$ ).



1. Correlation between dose kinetics for radiation induced DNA double strand breaks and cell survival (ref. 2, 21).



2. Correlation between dose kinetics and coefficients for radiation induced mutations and survival in chinese hamster cells. (ref. 4).



## Mutation Induction

Radiation damage of the DNA in the cell may lead to a change in the hereditary material which may be expressed as a mutation when the cell divides. A DNA single strand break can be perfectly repaired as the repair enzymes can copy the undamaged complementary strand. The repair of a DNA double strand break however, is, if it is possible, more likely to be accompanied by a fault and is thus a possible source of mutations. An unrepaired double strand break may lead to a loss of DNA and also to a mutation.

If each DNA double strand break in the cell has a probability  $q$  of leading to a specific mutation then the probability that  $N$  double strand breaks in the cell lead to the incidence of specific mutations is

$$M = 1 - e^{-qN} = 1 - e^{-q(\alpha D + \beta D^2)} \quad (3)$$

Figure 2 demonstrates the correlation between cell survival and mutation induction in chinese hamster cells<sup>4</sup>. The coefficients derived for cell survival can be used to give the curve for mutation induction. The dose kinetics and the correlation with cell killing suggest that the mutations arise from a DNA double strand break.

## Dose rate and LET effects

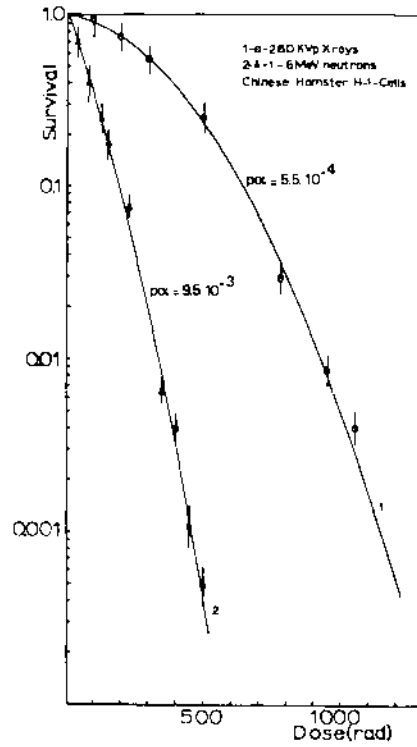
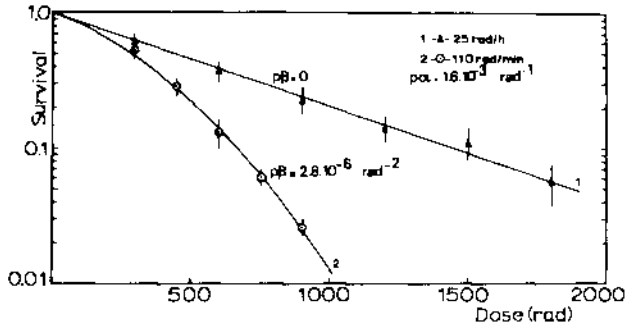
According to the model the dose rate and LET effects occurring in cell survival and mutation induction are directly related to the dose rate and LET effects occurring in the production of DNA double strand breaks. These effects can be explained briefly in the following way. The induction of double strand breaks in one radiation event is independent of any dose rate effect. The occurrence of double strand breaks arising from two single strand breaks will be dependent on the dose rate of the radiation if the DNA single strand breaks can be repaired during irradiation. In general cells have the ability to repair a single strand break accurately by the action of specific enzymes. Consequently, as the radiation dose is protracted more single strand breaks will repair and fewer double strand breaks will arise from two independent radiation events. This means that as the dose rate is decreased the coefficient  $\beta$  will decrease and at low enough dose rates will eventually become zero and equation (2) and (3) can be modified accordingly. This effect is illustrated in Figure 3a for cell survival.

The induction of double strand breaks in one radiation event whilst independent of dose rate is not independent of radiation quality and more densely ionizing radiation will have a greater chance of breaking both strands in one event than sparsely ionizing radiation. This effect is illustrated in Figure 3b for cell survival. The neutron irradiation is more efficient in breaking the double strand in one event and the  $\beta$  coefficient is consequently larger.

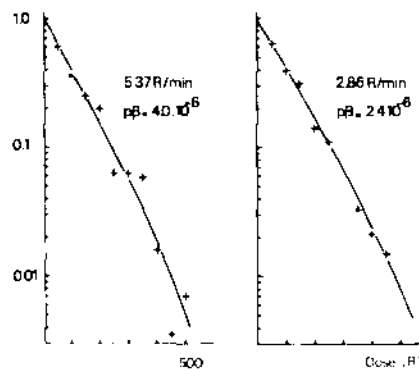
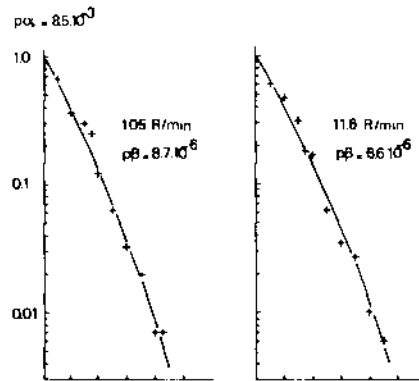
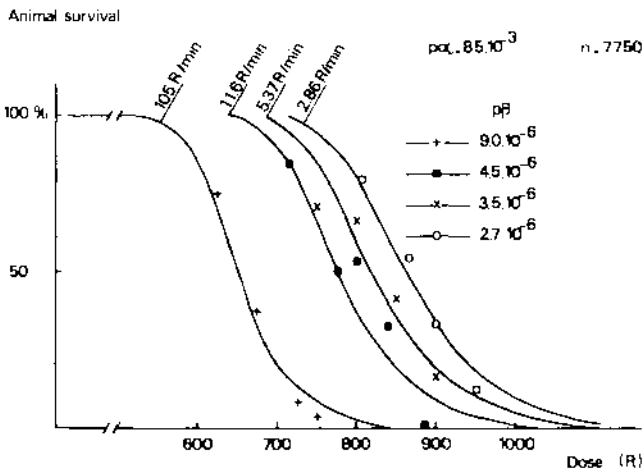
The model implies that the coefficient  $\beta$  or  $q$  is of prime importance for radiological protection as it is the coefficient which determines the biological effect at low doses and at low dose rates. The model based on the mechanism of DNA double strand breakage presented here offers the possibility of estimating the  $\beta$  or  $q$  coefficient from experiments made at high doses and high dose rates. It is possible to define a limiting relative biological effectiveness ( $RBE_0$ ) using the  $\alpha$  coefficients derived for two different radiations as

$$RBE_0 = \frac{\alpha_{\text{test radiation}}}{\alpha_{\text{standard radiation}}} \quad (4)$$

This  $RBE_0$  is especially relevant at low doses and dose rates, is dose and dose rate independent and its value is important in determining appropriate Quality Factor values.



3. a. Dose rate effect on cell survival (ref. 21).  
 25 rad/h  $p\alpha = 1.6 \times 10^{-3}$   $p\beta = 0$   
 110 rad/min  $p\alpha = 1.6 \times 10^{-3}$   $p\beta = 2.8 \times 10^{-6}$
- b. (right) Effect of radiation quality on cell survival (ref. 22).  
 280 kVp X  $p\alpha = 0.55 \times 10^{-3}$   $p\beta = 4.8 \times 10^{-6}$   
 1-6 MeV n  $p\alpha = 9.5 \times 10^{-3}$   $p\beta = 12.3 \times 10^{-6}$   
 $RBE_0 = 17$



4. Fit of the equation  $L = 1 - 1 - e^{-p(\alpha D + \beta D^2)}$  to the survival of female mice irradiated with X-rays at different dose rates (see ref. 7).
5. (right) Fit of equation  $S = e^{-p(\alpha D + \beta D^2)}$  to the survival of transplanted mouse bone marrow cells irradiated in vivo in recipient mice at different dose rates.

## Multicellular Effects

In this section the associations made between the molecular mechanism and the cellular effects are tentatively extended to develop dose relationships for multicellular effects. In this extension it is necessary to assume that the multicellular effect arises from the accumulation of damage at the cellular level and that in a homogeneous population the chance that the primary radiation damage develops to the ultimate biological effect is the same for all the animals in the experiment.

### Animal Survival

If it is assumed that:

1. the death of an animal following radiation is associated with a specific radiation syndrome which is connected with the killing of cells from a critical organ. For instance the LD 50/30 in mice is associated with the bone marrow syndrome which is related to the survival of haemopoietic stem cells<sup>5,6</sup>.
2. the death of the animal results because the specific cell pool is reduced below a critical level. In a homogeneous animal population this amounts to killing a certain critical number of the specific cells.

Then, if the specific cell survival is given by

$$S = e^{-p(\alpha D + \beta D^2)}$$

and  $n$  is the critical number of cells, the chance that the animal will survive is given by

$$L = 1 - (1 - e^{-p(\alpha D + \beta D^2)})^n \quad (5)$$

The survival curve for animals is thus defined by three coefficients,  $p\alpha$ ,  $p\beta$  and  $n$ , two of which define cell survival, and are related to the induction of double strand breaks in the DNA molecule.

A practical application of this theoretical expression to the survival of animals and of the correlation with cell survival is demonstrated in Figures 4, 5 and 6. Figure 4 presents the fit of equation (5) to the survival of female B6D2F1 mice aged 10-14 weeks and weighing 18-24 g following X-irradiation at different radiation dose rates<sup>7</sup>. Figure 5 presents the survival of transplanted bone marrow cells, irradiated *in vivo* in recipient mice at the same dose rates, fitted with equation (2). The analysis has been made as follows:

1. A best fit of equation (2) was made to the cell survival curves. The  $p\alpha$  coefficient was found to be approximately constant:  $(7.9 - 8.9) \times 10^{-3}$ .
2. A mean value for  $p\alpha$  of  $8.5 \times 10^{-3}$  was used and the best  $p\beta$  coefficients were determined for the cell survival.
3. Using  $p\alpha = 8.5 \times 10^{-3}$  the animal survival curves were best fitted for different  $n$  values to see if the  $p\beta$  values were in the same order as those found for cell survival.
4. This was indeed found and  $n$  was fixed at  $n = 7750$ ,  $p\alpha = 8.5 \times 10^{-3}$  and the best  $p\beta$  values were determined.

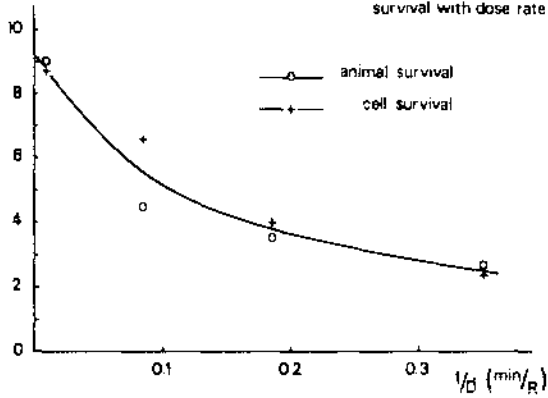
Figure 6 gives the comparison between the  $p\beta$  values determined in the cell survival studies and in the animal survival studies at the different dose rates.

### Radiation Induced Cancer

If it is assumed that a somatic mutation is the radiation induced process which eventually leads to the development of a radiation induced cancer<sup>8,9,10</sup> and that the mutated cell must be able to divide to give rise to the cancer<sup>11,12</sup> then the considerations on cell survival and mutation induction can be combined and an equation for cancer induction can be derived from equations (2) and (3) to give:

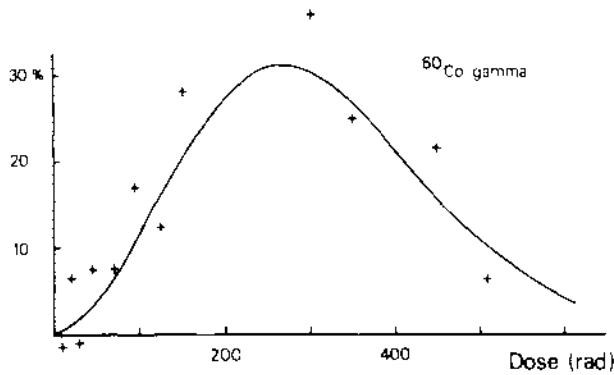
$$C = (1 - e^{-m(\alpha D + \beta D^2)}) e^{-p(\alpha D + \beta D^2)} \quad (6)$$

$p\beta$  coefficient ( $10^{-6}$ ) Variation of  $p\beta$  coefficient for animal and cell survival with dose rate

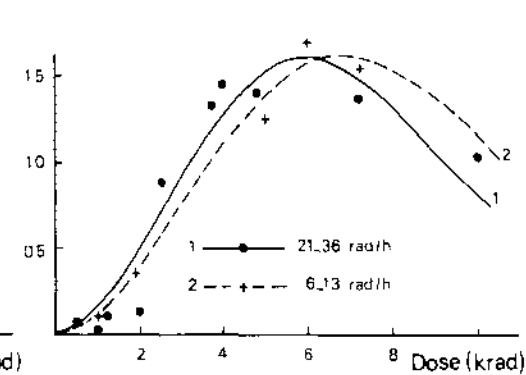


6. Comparison of the variation in the  $p\beta$  coefficients derived from animal and cell survival studies with irradiation dose rate (see ref. 7).

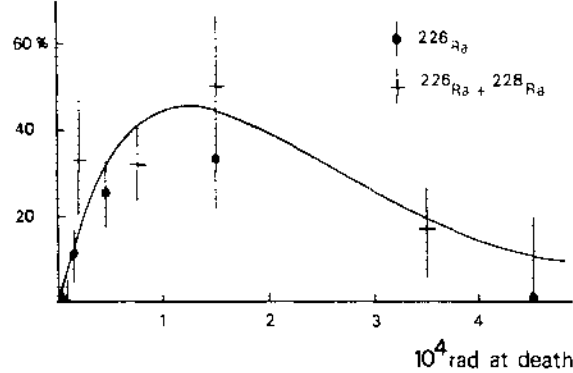
Myeloid leukemia incidence in mice



Incidence skin tumours in rat



Incidence Bone Sarcomas in Man



7. The fitting of the equation  $C = k(1 - e^{-m(\alpha D + \beta D^2)})e^{-p(\alpha D + \beta D^2)}$  to the incidence of myeloid leukemia in male mice (a) (ref. 13), skin tumours in rats (b) (ref. 14) and bone sarcomas in man (c) (ref. 15).  $k$  is a normalisation constant

	$k$	$m\alpha$	$m\beta$	$p\alpha$	$p\beta$
a	1.25	$2.0 \times 10^{-4}$	$9.0 \times 10^{-6}$	$2 \times 10^{-4}$	$9.0 \times 10^{-6}$
b 1	0.6	$7.2 \times 10^{-6}$	$1.8 \times 10^{-8}$	$8 \times 10^{-6}$	$2.0 \times 10^{-8}$
b 2	0.6	$7.2 \times 10^{-6}$	$1.35 \times 10^{-8}$	$8 \times 10^{-6}$	$1.5 \times 10^{-8}$
c	3.5	$3.0 \times 10^{-5}$	0	$7 \times 10^{-5}$	0

where  $m$  is the probability that a double strand break leads to a specific cancer mutation. Figure 7 shows the fitting of this equation to various data on cancer incidence. Figure 7a shows the equation fitted to the incidence of myeloid leukemia in male mice following  $^{60}\text{Co}$  gamma irradiation<sup>13</sup>. In figure 7b the effect of dose rate is demonstrated for the incidence of skin tumours in rats<sup>14</sup>, the two curves being fitted by varying the  $\beta$  coefficient only. In figure 7c an example of the effect of densely ionizing radiation on the incidence of bone sarcomas in man<sup>15</sup> is given using equation (6) with the  $\beta$  coefficient held at zero.

At low doses and low dose rates equation (6) reduces to

$$c = (1 - e^{-m\alpha D}) e^{-\beta\alpha D} \quad (6a)$$

This expression is linear from the origin, saturates to a maximum and then decreases. The results of the Oxford Survey provide a dose relationship for childhood cancer following obstetric radiography<sup>10</sup> at very low doses of radiation which can be compared with equation (6a). In figure 8 two examples of a possible fitting of equation (6a) to the dose response are presented. Curve number 1 is based on the assumption that the cell sensitivity of the foetus is similar to mammalian cells and gives a more or less linear fitting. Curve number 2 is based on the assumption that the foetus is more sensitive than normal cells and a clear curvature<sup>16</sup> is obtained. It is important to realise that the comparison presented in Figure 8 has certain short-comings the most important of which is that all cancers are considered together whereas the equation should be applied to specific cancers. The two curves in Figure 8 should not be considered as the only possibilities or even the extreme possibilities but only as a demonstration of the agreement between experimental results and the theory.

Some important comments can be made on the basis of equations (6) and (6a). Even at low doses there will be no threshold, and for sparsely ionising radiations a reduction in dose rate will lead to a sparing effect. At high LET, of course, equation (6a) applies for all dose rates and no sparing effect will be observed. Furthermore a specific mathematical feature of equation (6) is that the peak height is independent of the coefficients  $\alpha$  and  $\beta$  and only depends on the relationship between  $p$  and  $m$ ; this is demonstrated in figure 7b. The consequence of this is that for a specific cell type the maximum incidence is independent of irradiation type and conditions although the dose at which the peak occurs is dependent on these factors.

### Genetic Effects

No attempt will be made in this section to consider the multitude of problems involved in the estimation of genetic risk, but an attempt will be made to demonstrate that in two different stages of the same cell, the *Drosophila* oocyte, where the radiation induction of mortality has apparently different dose kinetics<sup>17,18</sup>, the same process of DNA double strand breakage could be involved.

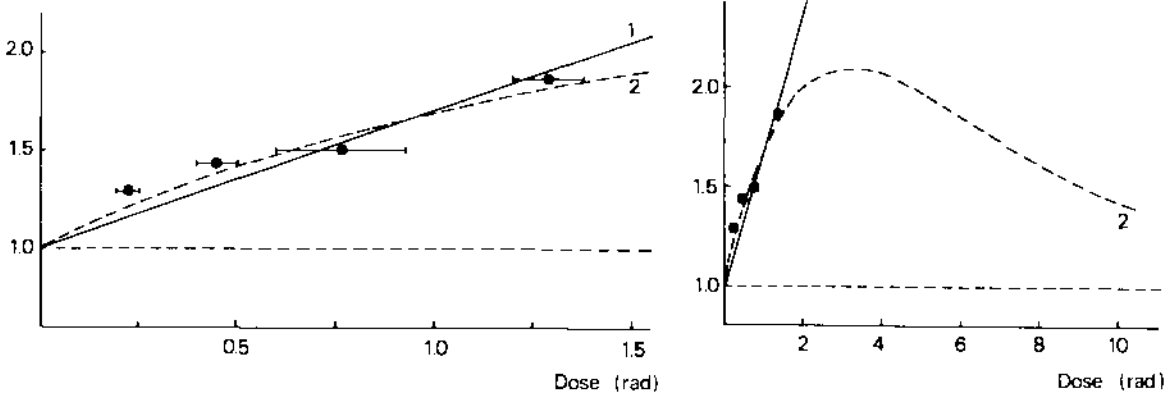
Figure 9 shows the mortality in *Drosophila* oocytes irradiated in stage 7 and stage 14. The curve for stage 14 oocytes has understandably been interpreted as an exponential curve; however, if the curve is analysed using the model and the equation

$$s = e^{-p(\alpha D + \beta D^2)},$$

then the best fit does lead to a positive  $\beta$  coefficient. This indicates that in this stage of the cell the induction of 'double strand breaks in one radiation event' is a dominant process but that the same mechanism of DNA double strand breakage could be involved in the induction of mortality in both stages of the *Drosophila* oocyte.

The induction of DNA double strand breaks is a process which can occur in all eukaryotic cells and it should therefore be possible to describe radiation effects in the reproductive cells of other insects and animals by the same dose

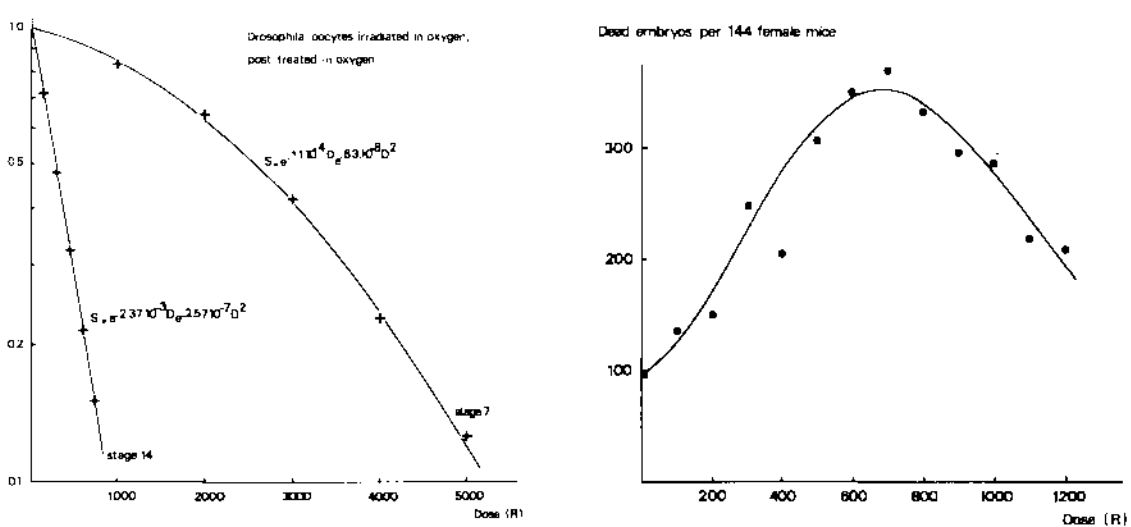
Relative incidence of Childhood cancers



8. The fitting of the equation  $C = k(1 - e^{-m\alpha D}) e^{-p\alpha D}$  to the relative incidence of all cancers in children following obstetric radiography (ref. 10).  $k$  is a normalisation constant.

	$k$	$m\alpha$	$p\alpha$
Curve No. 1	320	$2.2 \times 10^{-3}$	$2.2 \times 10^{-3}$
Curve No. 2	4.38	$2.2 \times 10^{-1}$	$2.2 \times 10^{-1}$

Mortality



9. The mortality of *Drosophila* oocytes, stage 7 and stage 14, fitted with the equation  $S = e^{-p(\alpha D + \beta D^2)}$ : (ref. 17, 18)

	$p\alpha$	$p\beta$
Stage 7	$1.06 \times 10^{-4}$	$6.3 \times 10^{-8}$
Stage 14	$2.37 \times 10^{-3}$	$2.57 \times 10^{-7}$

10. The relationship between a measure of the dominant lethality in mice and radiation dose (ref. 19) fitted by the equation

$$DE. = K (B + 1 - e^{-q(\alpha D + \beta D^2)}) e^{-p(\alpha D + \beta D^2)}$$

$K$	$B$	$q\alpha$	$q\beta$
1029	97/1029	$2 \times 10^{-4}$	$1.25 \times 10^{-6}$

$p\alpha$	$p\beta$
$1.6 \times 10^{-4}$	$1 \times 10^{-6}$

kinetics. An attempt to demonstrate this has been made in figure 10 which presents the relationship between a measure of the dominant lethality in mice and the radiation dose<sup>19</sup>. The experimental points represent the number of dead embryos per 144 female mice found following mating with male mice which had received a local testicular irradiation. The equation which has been fitted is

$$DE = K (B + 1 - e^{-q(\alpha D + \beta D^2)}) e^{-p(\alpha D + \beta D^2)}$$

where K is the number of implantations, and B represents the dead embryos found in the unirradiated control. The spermatozoa responsible for these dead embryos can also be 'inactivated' by the radiation.

This figure shows that the same dose kinetics are also found in the mouse cells as well as in the Drosophila cells. These kinetics do contain a dose rate effect which is in accordance with the findings quoted in Unsclear<sup>20</sup> that the induction of genetic effects are lower following chronic irradiation.

### Discussion

In this paper the development of a theoretical model to describe the effect of radiation on a series of biological end points has been briefly sketched.

The model is based on one critical radiation induced lesion which is known to occur in cells following radiation, the DNA double strand break. Correlations have been presented between the induction of DNA double strand breaks in vitro and cell survival; between cell survival and mutation induction; between cell survival and animal survival; between mutation induction and cell survival and cancer induction; and it has been shown that the same dose kinetics are involved in the induction of genetic effects.

We would like to emphasize the aspects of the model which we consider to be most important. The fit of the various equations to the experimental results is satisfying but is in itself not as important as the fact that the proposed molecular mechanism, which will be caused in all eukaryotic cells by radiation, forms a common thread which links the different biological effects. The dose kinetics and LET and dose rate effect arising through the mechanism are all reflected in the dose kinetics, LET and dose rate effects of the various biological end points. The second important aspect of the model is that it offers the possibility of interpreting multicellular and cellular biological effects on the basis of a common radiation induced molecular lesion.

The dose kinetics, the explanation of the dose rate effect and the LET effect developed in this paper are strongly reminiscent of the classical theory of chromosome exchange aberrations. It is important, however, to realize that the theory presented here is essentially different from the classical theory both in the starting assumptions and in the interpretation. The theory presented here starts from the assumption that the critical radiation lesion occurs at the molecular level and not the chromosome level, and the interpretation of the variations in the radiobiological effects is based on the biochemistry and metabolism of the DNA in the cell. The evidence in support of the assumption that the DNA double strand break is the critical lesion is the close agreement found between the coefficients determined for cell survival and the induction of DNA double strand breaks in vitro, and the fact that the analysis of the survival of synchronized cells at different stages of the cell cycle gives a variation in the  $\alpha$  coefficient which is in accordance with the partial separation of the DNA strands at the replication forks during the synthesis phase<sup>1</sup>.

It is clear that the development of the model from the molecular mechanism to the cellular and animal effects is theoretical and although it is based on assumptions which have been suggested previously, such as the somatic mutation theory of cancer, it is also somewhat speculative. The compatibility of the equations with both the experimental results and the proposed mechanism plus the coherence of the interpretation which can be obtained via the model are promising and it should not be too difficult to design experiments to test the

model more directly.

If it is assumed that the model presented here is valid the following implications of relevance to radiological protection may be inferred: The equations derived in the model are strictly non-linear but would provide an analysis of the results of high dose, high dose-rate radiobiological experiments to obtain values of coefficients which are applicable at low dose and low dose rate and which are of direct relevance to radiological protection. The equations would indicate that there is no threshold in the radiation effect and that at low doses a 'linear model' can be considered as applicable. The two parameters which would be of most importance for radiological protection are the absolute values of the ' $\alpha$ ' coefficients and the values of the limiting relative biological efficiency. The ' $\alpha$ ' coefficients would vary from cell to cell, from cell phase to cell phase and would be dependent to some extent on the environmental circumstances during and after the irradiation. The limiting relative biological efficiency would be dependent on the standard radiation, the cell type and the environmental circumstances but it would be somewhat higher than the RBE normally derived from radiobiological experiments. It would, in any case, be a constant value independent of dose and dose rate under standard conditions. A consequence of the model and the common mechanism would be that the limiting relative biological efficiency ( $RBE_0$ ) would be the same for the different biological end-points in the same cell irradiated under the same circumstances.

The model would indicate that considerable care should be taken in the choice of examples which are used to estimate risks. Although the theory would provide equations to describe biological end-points which could be used to extrapolate to low dose, low dose-rate conditions the equations would only apply to a homogeneous population irradiated under controlled conditions of uniform dose rate and radiation quality. These are conditions which are not usually satisfied in the cases used for the estimation of risks, at least somatic risks.

The molecular mechanism involved in the model is assumed to be common to all eukaryotic cells and as has been shown here can be related to many biological end-points. There would thus be a common basis for the extrapolation of results found in insects and animals to man. This extrapolation would of course have its restrictions and difficulties but at least the same fundamental process would be involved.

According to the model the molecular mechanism of DNA double strand breakage could form a common link between the biochemistry of DNA, and the radiation effect; the cell metabolism and how it affects the radiation effect via repair processes; the cell cycle and DNA configuration in the cell and the radiation effect; and it may provide a means of interpreting the results of radiobiological and radiation genetics experiments for extrapolation to radiological protection.

It is interesting to speculate on one other point which would arise if the model is valid and that is, that the molecular process, which is assumed in the model to lead to the radiation effect, is a process which is not unique to ionizing radiation. The same process of DNA double strand breakage can be caused by viruses, chemical agents, ultrasonic radiation and ultra-violet light. Thus, the model would suggest that the radiation hazard is not unique or especially different than the other everyday hazards we face.

#### Acknowledgements

This publication is contribution number 946 of the Euratom Biology Division. The work is supported by the Euratom contract 094-BIAN and by the Dutch Ministry of Agriculture and Fisheries.



## References

1. Chadwick, K.H. and Leenhouts, H.P., Phys. Med. Biol. 18, 78-87 (1973).
2. Freifelder, D. and Trumbo, B., Biopolymers 7, 681-693 (1969).
3. Hagen, U., Biochim. Biophys. Acta (Amst). 134, 45-58 (1967).
4. Chu, E.H.Y., Mutation Res. 11, 23-34 (1971).
5. Broerse, J.J., Int. J. Radiat. Biol. 15, 115-124 (1969).
6. Bond, V.P., Friedner, T.M., and Archambeau, J.O., Mammalian Radiation Lethality (Academic Press, N.Y.) (1965).
7. Puro, E.A. and Clark, G.M., Radiat. Res. 52, 115-129 (1972).
8. Burch, P.R.J., Radiation-Induced Cancer (IAEA, Vienna) 29-44 (1969).
9. Curtis, H.J., Radiation-Induced Cancer (IAEA, Vienna) 45-55 (1969).
10. Stewart, A., Health Phys. 24, 223-240 (1973).
11. Gray, L.H., Cellular Radiation Biology (The Williams and Wilkins Co. Baltimore) 7-25 (1965).
12. Mole, R.H. 8th Annual Meeting European Soc. for Radiation Biology, Basko Polje (1971).
13. Upton, A.C., Jenkins, V.K. and Conklin, J.W., Ann. N.Y. Acad. Sci. 114, 189-202 (1964).
14. Albert, R.E., Newman, W. and Altshuler, B., Radiat. Res. 15, 410-430 (1961).
15. Mays, C.W., Taylor, G.N., Jee, W.S.S. and Dougherty, T.F., Health Phys. 19, 601-610 (1970).
16. Stewart, A. and Kneale, G.W., Health Phys. 24, 359 (1973).
17. Sankaranarayanan, K., Mutation Res. 7, 357-368 (1969).
18. Sankaranarayanan, K., Mutation Res. 7, 369-383 (1969).
19. Leonard, A., Mutation Res. 3, 73-78 (1966).
20. United Nations Scientific Committee on the Effects of Atomic Radiation, Ionizing Radiation: Levels and Effects (1972).
21. Fox, M. and Nias, A.W., Current Topics in Radiation Research Quarterly. 7, 71-103 (1970).
22. Schneider, D.O. and Whitmore, G.F., Radiat. Res. 18, 286-306 (1963).

RECENT TRENDS TOWARDS THE DOSE HAZARD REFERENCE IN STANDARDS  
FOR DIFFERENT NUCLEAR ACTIVITIES

Lidia Failla - "Chairman of the Sub Committee 4 Radioisotopes" of the  
UNI (Italian Unification Organization) joined with ISO  
Rome, Italy

ABSTRACT

Dose hazards using radionuclides groupings are considered, with a view to the importance of including the nuclides utilization modes in their classification.

The problem of giving quantitative values to parameters for the evaluation of dosage risk is analyzed, changing from the concept of curies to the concept of rems, giving IAEA regulations for transportation of radioactive materials as an example.

Value of the reference dose is examined, mentioning the priority meter used in Italy in surveillance activities on instruments containing isotopes.

---

Radioprotection rules, prepared by international organizations as recommendations, regulations and directives, are, in most cases, completed by a radionuclides grouping called 'radiotoxicity grouping'. It considers the various degrees of hazard, mostly on the basis of metabolism and damages which can consequently be caused to man.

These groupings are very important, since they provide the basis for any further consideration on the risk in the different uses of radionuclides. It is not correct, however, to associate radiotoxicity with risk. In fact, radiotoxicity represents only one, even if important, of the elements which characterize risk.

That is to say that the said groupings represent a fundamental theoretical basis, but are not sufficient to calculate dose risks, since they do not consider the modes of nuclide utilization.

For example, with respect to hazard, it is not sufficient to place  $^{241}\text{Am}$  in the first radiotoxicity group and  $^{131}\text{I}$  in the second group, without considering the modes of utilization. It is clear that a sealed source of  $^{241}\text{Am}$  is much less hazardous than a source of  $^{131}\text{I}$ , of the same activity, used in therapy.

On the contrary, this distinction is of fundamental importance from the point of view of the different radiotoxicity and it constitutes the basis for a further formulation which considers the other parameters already mentioned.

Euratom radioprotection directives determine certain statements or authorization obligations for definite radioactivity levels, based on radiotoxicity groupings. For the group with the highest radiotoxicity, such level is fixed at 0.1  $\mu\text{Ci}$ ; for the next group in 1  $\mu\text{Ci}$  and then in 10 and 100  $\mu\text{Ci}$  for the other two groups. These levels do not take into consideration the modes of utilization.

With the experience of decades, we cannot forget that the activity level of the particular nuclide utilized cannot be the only determining criterion of the hazard.

The possibility is foreseen, then, to reach classifications which, besides radiotoxicity, take also into account the modalities of usage.

Consequently, two main problems arise.

The first one concerns the characterization and definition of parameters which, for the different modes of usage, permit the evaluation of dosage risk.

When possible, it would be opportune and, from the scientific point of view, certainly more suitable to give quantitative values to these parameters. For some of them, it is rather easy. If we consider, for example, a  $^{192}\text{Ir}$  source, we may think, even being conservative, to give zero value to this source for the dispersion parameter, considering the metal shell (layer) and the lack of brittleness of the source. We cannot do the same with  $^{60}\text{Co}$ , for example, notwithstanding its metallic state, but because of its brittleness, when used in conditions where this brittleness can be involved.

The radiation parameter of a pure alpha particles emitting source is certainly different and lower than of a beta or gamma emitter, on the basis of the scheme contained in the instrument used.

The problem is not easy, since it is necessary to examine each radionuclide and for each of them, give different quantitative values to the usage conditions parameters, and to their radiotoxicity with reference to the dose absorption risk. On the contrary, para-

meters choice and definition could be the same for all radionuclides and for all uses.

The above statement, besides being substantial from the practical point of view, presents a formulation of principle which is very different from the one used in radioprotection by some international organizations and, consequently, by some national rules.

The new radioprotection formulation is a change from the concept of curies to the concept of rems. This means to substitute the capability of the dose risk pertaining to the radionuclide and to the different characteristics of modes of usage to the discriminative and determinant capability of the radioactivity value connected to the radiotoxicity of the single nuclides.

IAEA regulations for transportation of radioactive materials<sup>1</sup> are based substantially on the above concept. In those regulations, the maximum activity allowed in "type A" packages is fixed on the basis of a radionuclides radiotoxicity classification which is connected, however, to their hazards in the particular field of transportation, with consideration to the maximum permissible dose in case of accident, assuming certain dispersion and possible absorption conditions. In the last regulations, the radionuclides classification was abolished, because of the opportunity to consider the chemical and physical characteristics of each of them.<sup>2</sup>

This means that IAEA always considered nuclides in connection to the type of utilization (transportation), with reference to the dose hazard they may have in case of accident, on the basis of the above mentioned parameters.

The second problem which presents itself in view of this new optics of radioprotection is connected with the value of the reference dose.

In the mentioned regulations, IAEA always assumed the value of 3 rems.

It is clear that this problem, even if of great importance, is not difficult to solve, since, assuming a value and calculating parameters on its basis, it is easy to calculate its multiple and submultiples.

For example, IAEA fixed the maximum activities which can be contained in "type A" packages, by the utilization of certain parameters with a dose hazard of 3 rems. If utilization conditions

and the matter would require a thousand times greater dose, it would be sufficient to multiply by a thousand the maximum values of the permissible activities in the mentioned packages.

In order to illustrate the instance above, we believe in the opportunity to mention a method for establishing a priority range in the surveillance activities on the different instruments containing isotopes for industrial applications that we used in Italy. Such formulation is based upon the different hazard which the instruments present on the basis of their particular type, the radionuclide and the number of instruments present at the same time in the same room.

The problem was limited to the instruments which are of wider use in Italy, as:

- gammaradiography apparatus, containing  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{192}\text{Ir}$ ,
- irradiators, containing  $^{60}\text{Co}$ ,
- analytical balances, containing  $^{204}\text{Tl}$  and  $^{90}\text{Sr}$ ,
- light indicators, containing  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{226}\text{Ra}$ ,  $^{241}\text{Am}$  and  $^{90}\text{Sr}$ ,
- gas chromatography apparatus, containing  $^{63}\text{Ni}$  and  $^3\text{H}$ ,
- level indicators, containing  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{226}\text{Ra}$ ,  $^{241}\text{Am}$  and  $^{90}\text{Sr}$ ,
- thickness meters, containing  $^{204}\text{Tl}$ ,  $^{137}\text{Cs}$ ,  $^{226}\text{Ra}$ ,  $^{90}\text{Sr}$ ,  $^{85}\text{Kr}$ ,  $^{147}\text{Pm}$  and  $^{144}\text{Ce}$ ,
- lightning rods and fire monitors, containing  $^{241}\text{Am}$  and  $^{226}\text{Ra}$ ,
- electrostatic discharge apparatus, containing  $^{210}\text{Po}$  and  $^{226}\text{Ra}$ ,
- sulforimeters, containing  $^{147}\text{Pm}$  and  $^3\text{H}$ ,
- calibration sources, containing  $^{226}\text{Ra}$ ,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,
- moisture gauges, containing  $^{241}\text{Am}$ ,
- weight and density meters, containing  $^{85}\text{Kr}$ ,  $^{204}\text{Tl}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$  and  $^{137}\text{Cs}$ ,
- fluorescence analyzers, containing  $^{238}\text{Pu}$ ,
- radiation monitors, containing  $^{85}\text{Kr}$
- powder analyzers, containing  $^{85}\text{Kr}$ ,
- production control instruments, containing  $^{241}\text{Am}$ .

In order to define the priority criteria in surveillance, we considered dose hazards with respect to the instrument used, to

the conditions of use and the contained radioisotope. The reference dose was the same adopted by the IAEA for transportation, that is 3 rems, but it was considered as a dose hazard absorbable in one year, while IAEA consider it as a dose which may be absorbed only once in case of an accident involving "type A" packages. Parameters were the following:

- activity of the radionuclide contained in each instrument,
- M.P.C. in air ( $\mu\text{Ci}/\text{cm}^3$ )  $\cdot 10^9 \text{ cm}^3$ , which given an approximate dose of 3 rem,<sup>3</sup>
- number of instruments in each plant.

On the basis of the mentioned criteria, it was possible to list 160 plants, for a total of 1636 instruments for surveillance purposes. It can be noted that the radiation dose was not taken into consideration, since the gammarradiography apparatus and the irradiators were automatically included in the greater hazard class. Because of the radionuclides used and their activity, the other instruments present pre-eminent risk, because of contamination hazard rather than radiation hazard.

On the other hand, we are only making an approximation of the problem, since our only scope is to indicate a priority in the surveillance actions and not a scientific study, as it would be desirable if the new radioprotection formulation were accepted.

The plant classification, with respect to the dose hazard expressed in rem/year, is the following:

$$\begin{aligned}
 & A \leq 3 \\
 & 3 < B \leq 3 \cdot 10^2 \\
 & 3 \cdot 10^2 < C \leq 3 \cdot 10^3 \\
 & 3 \cdot 10^3 < D \leq 3 \cdot 10^4 \\
 & E > 3 \cdot 10^4
 \end{aligned}$$

In the case of Italy, the following applies:

Number plant	Hazard level
2	A
7	B
11	C
15	D
125	E
Total	160

In conclusion, we may try to summarize this complex matter as follows.

Radionuclide groupings on the basis of their radiotoxicity are certainly useful, but are not determinant in individuating hazard levels. For this purpose, it is necessary to complete these groupings with the modes of utilization of radioisotopes, determining a series of quantitative parameters. On the basis of the mentioned criteria, we will be able to classify them with consideration to dose risk, for which the radionuclide activity and its radiotoxicity shall have become only two of the parameters for the purpose of hazard specification.

This means to start anew and integrate what IAEA already made in the field of transportation, with a change of hazard evaluation from nuclide activity in curies to dose hazard in rems.

Rom, 28 May 1973

#### BIBLIOGRAPHY

- 1) IAEA - "Regulations for the Safe Transport of Radioactive Materials" - 1967 - Safety Series No 6.
- 2) IAEA - GOV/1529 - 3 Août 1972.
- 3) ICRP - Report of Committee II on Permissible Dose for Internal Radiation - Publication 2, 1959

Опыт использования клинических данных для обоснования предельно допустимых уровней профессионального облучения.

А.К.Гуськова, Е.А.Денисова, Э.Н.Львовская, Г.И.Кирсанова, И.А.Грибова.  
Институт Гигиены труда и профзаболеваний АМН СССР. г.Москва.

#### SUMMARY

The results of observation of workers dealing with high energy accelerators are discussed. Unacceptable doses accumulated during 10-15 years resulted in 10-100 rad. Radiation damages have not been observed. Clinical data have proved the conservatism of the present established maximum permissible dose of professional irradiation for these groups of workers.

В настоящем сообщении рассматривается конкретный вопрос об использовании опыта динамических целенаправленных наблюдений для клинического обоснования принятых предельно допустимых уровней некоторых видов профессионального облучения.

В соответствии с международными рекомендациями /1/ и принятыми в СССР в 1969г нормами по радиационной безопасности /2/ клиническое понятие предельно допустимой дозы трактуется как предельная величина профессионального облучения, которое даже будучи продолжено в этом максимальном значении в течение всей трудовой деятельности человека, не вызовет у него отклонений в состоянии здоровья, нарушающих состояние основных органов и систем и как-либо ограничивающих его трудоспособность.

Для обоснования этой величины, помимо экспериментальных данных, должны привлекаться материалы ряда клинических наблюдений, но при строгом соблюдении следующих условий:

- наличие уверенности в относительной точности дозиметрической характеристики воздействия радиации, с восстановлением динамики формирования суммарной дозы за длительный период наблюдения - не менее 10-15 лет;

- Возможность сопоставления результатов исследования в динамике групп лиц, отличающихся по уровню доз в границах исследуемого диапазона, а также подвергающихся воздействию заведомо более высоких и особенно более низких доз облучения;



- одновременное параллельное обследование адекватной контрольной группы достаточной численности с использованием тех же методов исследования и выбором строгих критериев существенности отличий как по результатам статистической обработки, так и по возможной клинико-физиологической значимости выявленных сдвигов;

- целенаправленное углубленное по сравнению с обычной схемой медицинское обследование, имея в виду попытку выявить наиболее ранние отклонения в состоянии органов и систем, критических для данного типа радиационного воздействия.

С учетом указанных предпосылок нами были проанализированы данные 850 человек, работающих в течение 10-15 лет на ускорителях заряженных частиц Объединенного Института Ядерных Исследований в г. Дубне в сопоставлении с результатами обследования 200 лиц адекватной контрольной группы.

Дозиметрические исследования /3-5/ показали, что основной вклад в суммарную дозу излучения вносит гамма излучение от наведенной радиоактивности и нейтроны широкого спектра. При этом вклад в общую дозу от сопутствующих компонентов /бета излучение, заряженные частицы и т.п./ не превышает 10-20%. Дозы облучения у подавляющего большинства обследованных не превышали предельно допустимой величины /5 бэр в год/. Такая характеристика условий облучения типична и для других ускорителей /6-10/.

Дополнительные сопоставления проводились с данными наблюдения за персоналом исследовательских реакторов /свыше 1000 чел./, обследованных по тем же принципам, дозы облучения которых за весь период работы не превышали 1/3 предельно допустимой величины в год, суммарно порядка 10-25 бэр. Для сравнения с эффектами более высоких в прошлом уровней облучения привлекались данные на медицинских рентгенологов /2500 чел./ и промышленных радиотрафистов /400ч./ /11-14/. У части лиц этих групп, начавших работать до 1960 г, когда принятая величина ПДД составляла 15 бэр/год, экспозиционные дозы гамма и рентгеновского излучения могли достигать 70-400 бэр с неравномерным ее распределением за весь период трудовой деятельности. Контрольные группы к этим контингентам подбирались дополнительно, общая численность их составила около 500 чел.

Углубление традиционного объема медицинских исследований исходило из стремления: а/ выявить возможные, в связи с общим облучением организма, реакции на воздействие радиации как на раздражитель; б/ уточнить состояние критических в данной ситуации органов и структур /клеточные органы, хрусталик, хромосомный аппарат

лимфоцитов периферической крови и миеокариоцитов костного мозга/. В соответствии с этим был расширен объем методических приемов, уточняющих состояние анализаторов, регуляторных механизмов /особенно регуляции общей и регионарной гемодинамики в покое и при предъявлении адекватных нагрузок/. Оценивалась по некоторым тестам умственная работоспособность. Исследовалась периферическая кровь, у части лиц анализировались пунктаты костного мозга, определялась частота и характер aberrаций в культуре лимфоцитов периферической крови. Помимо обычного офтальмологического осмотра проводилась биомикроскопия хрусталика с помощью щелевой лампы /15-19/.

В качестве основной категории определения состояния здоровья использовались следующие основные понятия или синдромы:

- Здоров - отсутствие жалоб и объективных отклонений от принятых за норму пределов колебаний показателей при специальных клинико-физиологических исследованиях;

- Практически здоров - наличие отклонений, не сказывающихся на самочувствии и работоспособности, без признаков клинически отчетливой функциональной недостаточности какой-либо системы;

- Неспецифический синдром функциональной неустойчивости или повышенной лабильности регуляции в ряде сопряженных систем, адаптирующих организм к внешней среде;

- Общее соматоневрологическое заболевание, выявленное в момент обследования;

- Профессиональное лучевое заболевание. Следует упомянуть, что ни у одного человека из числа обследованных нами в диапазоне доз близких к ПДД, не было выявлено профессионального заболевания, соответствующего нашему представлению о возможном для данного типа радиационного воздействия первом варианте хронической лучевой болезни по классификации А.И. Гуськовой и Г.Д. Байсоголова /20/.

Анализ полученных результатов позволяет дать следующую общую оценку состояния здоровья лиц, работающих в течение 10-15 лет в условиях облучения, близких к ПДД общего воздействия проникающей радиации. Состояние здоровья большинства обследованных в течение всего периода наблюдения остается вполне удовлетворительным. Существенных различий между основной и контрольной группами на конечный срок наблюдения нет. Среди обследованных обеих групп при последних осмотрах несколько увеличилась частота заболеваний общесоматического характера и клинические проявления их стали более яркими. Это, по видимому, связано с увеличением возраста обследованных на 10-15 лет и сопутствующими изменениями образа жизни, пита-

ния, физическо<sup>ю</sup> нагрузки и т.п. Частота, характер и структура заболеваемости не отличались достоверно в основной и контрольной группах. При использовании специальных методических приемов у лиц основной группы, особенно закономерно в диапазоне доз свыше 1/3 ЦД, наблюдается статистически существенное учащение комплекса отклонений, касающихся: а/расширения объема физиологических функциональных сдвигов на нагрузки по типу синдрома повышенной лабильности нервно-сосудистой регуляции с формированием в части случаев синдрома нейрокругляторно<sup>ю</sup> дистонии гипотонического типа /рис. 1/. Частота синдрома функциональной неустойчивости увеличивалась по сравнению с исходно<sup>ю</sup> и данными контрольной группы при достижении суммарной величины дозы облучения порядка 25-35 бэр, а в дальнейшем уменьшалась более медленно у лиц с больше<sup>ю</sup> дозо<sup>ю</sup> облучения. Эти сдвиги могут рассматриваться как реакция адаптации к изменившимся условиям внешней среды.

- б/некоторого ускорения и учащения признаков возрастной инволюции структуры хрусталика в отдельных возрастных группах /рис. 2/.

- в/признаки активации эритроидного ростка костного мозга. У большинства лиц, работающих при дозовых нагрузках от 1/3 до 2-5 ЦД, в первые годы происходило расширение диапазона колебаний гематологических показателей, а в дальнейшем показатели стабилизировались на нормальном уровне /рис. 3/. Колебания средних величин показателей крови в основной и контрольной группах в динамике были идентичны, что представлено на примере числа лейкоцитов и тромбоцитов /рис. 4/.

Клиническая значимость выявленных сдвигов повышенно<sup>ю</sup> лабильности регуляции на данный момент должна быть оценена как адаптационная реакция ко всему комплексу влияний окружающей среды. Регуляторный сдвиг является полиэтиологическим по своему происхождению. Об этом свидетельствует наличие его у части работающих в близком диапазоне доз, регресс частоты сдвигов при возрастании сроков наблюдения и суммарно<sup>ю</sup> дозы, а также доказанное влияние ряда других неблагоприятных факторов режима жизни, злоупотребления курением и др.

Во всем изученном диапазоне доз не выявлено очерченных клинических синдромов профессионального лучевого заболевания. Перевод другой группы работающих /рентгенологи/ в условия, соответствующие ныне принятым ЦД внешнего облучения из ранее неблагоприятных, при суммарных дозах 100-150 р, сопровождался развитием отчетливых восстановительных сдвигов в кроветворении /13, 14/. Частота различных общесоматических заболеваний и структура заболеваемости и у пер-

сонала реакторов, соответствовала таковой в контрольной группе, а течение их не обнаруживало каких-либо очевидных особенностей. Сопоставление с экспериментальными данными может также подтвердить сохранность в данном диапазоне доз основных физиологических функций и отсутствие существенных сдвигов в состоянии здоровья./21/.

Все указанное позволяет считать, что принятые ПДД внешнего облучения являются достаточно обоснованными по критерию непосредственных клинико-физиологических реакций, устанавливаемых в процессе 10-15-летнего динамического наблюдения в диапазоне суммарных доз до 100 бэр. Целесообразно продолжение наблюдения для получения информации о поздних эффектах с использованием программ обследования, подобных приведенной выше.

#### Л И Т Е Р А Т У Р А

1. Рекомендации по защите от излучений МКРЗ /1962-1971/.
2. Нормы радиационной безопасности, НРБ-69. Атомиздат, М./1970/
3. Комочков М.М. и др. - Оценка уровней ионизирующего излучения вблизи синхротрона на Ю Гэв. Дубна /1965/, Р-2231.
4. Лебедев В.Н. и др. -ж. Гигиена труда и профзаболевания, 4 /1966/, 9.
5. Зайцев Л.Н. и др. - Основы защиты ускорителей. Атомиздат, /1971/.
6. The effects of routine occupational radiation exposure in workers at the Lawrence Livermore Laboratory, Berkeley, Ca. /1967/.
7. Radiation hazard near high-energy accelerators and occupational exposures. Conf. Geneva, November 5-7 /1969/.
8. European organization for nuclear research. Health Physics /1972/
9. Population doses from occupational exposure no. A/AC/02/R/255 Un (Add. II) /1970/
10. A publication of the Department of National Health and Welfare Radiation Protection Division Ottawa, Canada. /1969/.
11. Гуськова А.К. и др. -ж. Гигиена и санитария, /1967/, 9, 39-45.
12. Денисова Е.А. и др. - в кн. Вопросы эксперимент. и клин. радиологии, Изд. Здоровье, Киев /1968/, 140.
13. Солдатова В.А. и др. -ж. Медицинская радиология, /1967/, 3, 73.
14. Летавет А.А. и др. - Основные итоги и задачи мед. наблюдения за лицами, работающими с источниками ионизирующего излучения в СССР на современном этапе. - докл. на IV конфер. по мирному использованию атомной энергии. Женева /1971/, A/Conf. 49/P/439.
15. Гуськова А.К. и др. - в кн. Биологическое действие протонов высоких энергий. Атомиздат. /1967/, 373-400.
16. Гуськова А.К. и др. -ж. Гигиена труда и профзаболевания, 6/1972/17.
17. Гуськова А.К. и др. -ж. Гигиена труда и профзаболевания, 8/1972/18.
18. Соколов В.В. и др. -ж. Советская медицина, 8. /1970-, 145.
19. Соколов В.В., Грибова И.А. - Гематологические показатели здорового человека. Медицина. М. /1972/.
20. Гуськова А.К., Байсоголов Г.Д. - Лучевая болезнь человека, М/1970/.
21. Григорьев Ю.Г. - Тез. докл. Всесоюзного симпозиума "Хрон. действие внешнего гамма облучения на организм собак". М. /1972/.

Подписи к рисункам к докладу А.К.Гуськовой и др. — "Опыт использования клинических данных для обоснования ЦДУ профессионального облучения".

Рис.1 — Частота синдрома функциональной неустойчивости у лиц основной группы с различной дозой облучения в сравнении с контрольной группой в динамике.

Рис.2 — Частота изменений в хрусталике при исследовании в проходящем свете с помощью электроофтальмоскопии с линзой + 15д у лиц основной и контрольной групп в динамике.

Рис.3 — Результаты динамического изучения числа лейкоцитов и тромбоцитов в основной и контрольной группах.

Рис.4 — Результаты динамического изучения количества лейкоцитов и тромбоцитов у обследуемого А.

*Гуськова*  
*Д. М. Гуськов*

EARLY HAEMATOLOGICAL DETECTION OF THE EFFECTS OF LOW DOSES OF  
IONIZING RADIATION IN PROFESSIONALLY OR MEDICINALLY EXPOSED  
PERSONS

T. Predmerszky, Olga Geszti, I. Előd and I. Loványi  
"Frédéric Joliot-Curie" National Research Institute f. Radiobiology  
and Radiohygiene  
Budapest, Hungary

Abstract

For early detection of the effects of low doses of ionizing radiation in professionally or medically exposed persons, we elaborated the method called by us X-ray resistance of the erythrocytes. We found the haemoglobin level of blood plasma elevated following professional exposure as well as after diagnostic X-ray procedures, therapeutic X-irradiation and internally applied therapeutic doses of radioactive isotopes. We attributed this elevation to increased erythrocyte membrane permeability and proved it by in vitro irradiation of the blood of persons exposed in vivo to low X-ray doses. The decreased X-ray resistance of erythrocytes was found to be dose-dependent. Our method is apt to demonstrate the immediate effects of very low doses on the human blood. Thus, it may be found valuable in radiation health control.

Early detection of eventual effects of low-level ionizing radiation in professionally, medically or accidentally exposed persons is a problem of outstanding importance in radiation health control. The changes induced by these effects might be functional or structural, or possibly both, and may be detected by various techniques of chiefly biochemical, cytogenetical, cytological and haematological character. The alterations demonstrated by these methods are the so-called biological indicators of radiation damage.

The great majority of the haematological tests are based upon quantitative and qualitative changes of the leukocytes. We, on our part, supposed that the erythrocytes exposed to ionizing radiation in the peripheral blood may also undergo certain changes. Owing to the large mass of erythrocytes these changes should be detected by relative ease.

We studied the membrane permeability changes of the erythrocytes, which could be easily followed by examining the actual plasma haemoglobin concentration. Successive determinations, e.g. before and after exposure, may supply quantitative data concerning the changes of erythrocyte membrane permeability, i.e. radioreistance of the erythrocytes.

## Materials and Methods

The examinations were carried out in the following groups:

### I. in vivo exposure

1. Seventy persons, professionally exposed to chronic doses of ionizing radiation far below the maximum permissible level, working either in the X-ray machine industry, or in the X-ray department of some medical institution for several years. The examinations were performed in course of routine health control.

2. Fifty persons, treated with therapeutic X-ray doses for various diseases, once or repeatedly.

3. Ten persons treated with therapeutic doses of I-131 isotope for malignant goiter, once or repeatedly.

4/a. Hundred persons, in whom diagnostic X-ray examination of the stomach was performed;

4/b. Thirty-two persons, in whom irrigoscopy was performed.

5. Unexposed, healthy control group.

### II. in vitro exposure

Ten ml human blood samples were irradiated in Petri dishes with 25, 50 and 100 R doses, resp., (measured in air) by a therapeutic X-ray machine.

In each case heparinized blood was utilized. In Group 2, the samples were drawn immediately after exposure; in Group 3, both before and 3 to 4 days after I-131 administration; in Groups 4 a. and b. before and after X-ray examination. Plasma haemoglobin determination was performed photometrically.

The in vitro irradiated blood samples derived from the unexposed control group. In vitro irradiation was performed also in the Groups 3. and 4 b.

The results were evaluated by statistical analysis (Student's "t" test) wherever a satisfactory number of data rendered such an analysis possible (See Table I.).

Summarized results of radioresistance in exposed groups

Type of exposure	No. of persons	Elevation of plasma Hb mg %	P
Professional external	70	2.41	< 0.001
Therapeutic X-ray	50	3.09	< 0.001
I-131-therapy, internal	10	0.71 - 1.06	-
Diagnostic gastric X-ray	100	1.16	< 0.01
Irrigoscopy	32	statistically insignificant	

In Groups 1. and 2. the results were compared with those of the unexposed controls, whereas in Groups 3. and 4. the difference between plasma haemoglobin level before and after exposure has been calculated.

## Results

Group I/1: in professionally exposed persons a mean plasma Hb concentration of  $3.02 \pm 0.39$  mg% was found, against the value  $0.608 \pm 0.01$  mg%, measured in unexposed controls. The difference is highly significant ( $p < 0.001$ ).

Group I/2: in therapeutically X-irradiated patients the mean plasma Hb concentration was  $3.7 \pm 0.68$  mg%. The difference against the unexposed control group is highly significant ( $p < 0.001$ ); whereas there is no significant difference when compared with the results of the professionally exposed persons.

Group I/3: slight elevation of the plasma Hb level was found on day 4. after I-131 (mean:  $+ 0.71$  mg%). The elevation was somewhat more marked in those cases where the therapeutic doses of I-131 were applied repeatedly (mean:  $+ 1.06$  mg%). The difference became more pronounced when irradiating the blood samples in vitro with 50 and 100 R X-ray doses (25 R: no effect). Particularly high was the elevation following in vitro 100 R X-irradiation, in cases of repeated isotope administration.

Group I/4 a.: the mean difference between plasma Hb values in samples taken before and after gastric X-ray examination was  $+1.16 \pm 0.036$  mg%, highly significant ( $p < 0.01$ ). The difference was also highly significant between the mean results of X-ray examinations when reducing the intensity of electric current from 4 mA to 1-2 mA by the use of an amplifier and TV set (mean:  $+1.35 \pm 0.14$  mg% against  $+0.69 \pm 0.083$  mg%).

In those cases where the plasma Hb determination was repeated 3 days after the gastric examination, we found values almost on the level of the basic (pre-exam.) ones. Thus, the decrease of erythrocyte radioresistance, induced by gastric fluoroscopy, seems to be reversible<sup>1</sup>.

Group I/4 b.: irrigoscopy exerted no demonstrable effect on erythrocyte membrane permeability. Similarly, no differences were found after in vitro X-irradiation as to the samples from the unexposed control group.

Groups I/5 and II.: the plasma Hb values of the control group were already cited in comparison with the groups of exposed individuals. The plasma Hb elevation, induced by in vitro 25, 50 and 100 R X-irradiation, resp., are represented by Fig. 1.

## Discussion

The level of plasma Hb in the peripheral blood is fairly constant; apart from the haemolytic diseases only severe physical stress is known to induce its significant elevation. Its increase is generally attributed to partial haemolysis, induced by increased erythrocyte membrane permeability, that may be caused also by high doses of in vivo or in vitro irradiation<sup>2,3,4,5,6</sup>. The results of our examinations seemed rather surprising, since these were obtained in the majority of cases after exposure to low-level doses.

When analysing plasma Hb elevation found in the various groups, exposed under a variety of circumstances, it seems rather obvious that the changes were developing during exposure to ionizing radiation exerting a direct effect on the erythrocytes in the circulation. Particularly convincing seems the comparison of the effects of the two diagnostic procedures (Groups I/4 a. and b.). During gastric X-ray examination large vessels (e.g. abdominal aorta, v.cava, v.portae) and organs of considerable blood content are included in the observation field that are passed several times by



RADIORESISTANCE OF ERYTHROCYTES IN UNEXPOSED CONTROL GROUP.

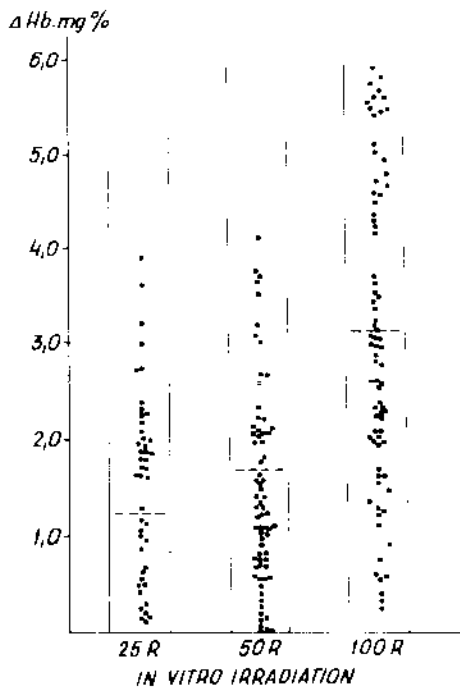


Fig. 1

in the blood, the organ of highest water and oxygen content.

Until now, we failed to establish any correlation between the dose of radiation, from external or internal source, or the duration of exposure in professionally involved persons and plasma Hb elevation. The dose-dependence, which was clearly manifested in the *in vitro* irradiated blood samples, justifies the hope that the method elaborated by us may be further developed, and will become a useful biological indicator, in a true quantitative sense, of radiation effect due to low-level ionizing radiation. Presently, the method is already apt to demonstrate the immediate as well as the chronic effects of very low doses on the human blood. Thus, it may be found valuable in radiation health control.

References

1. Geszti, Olga, Előd, I., Bojtor, I., Predmerszky, T. and Loványi, I.: *Strahlentherapie*, 142:213, 1971.
2. Holthusen, H.: *Strahlentherapie*, 14:56, 1951.
3. Ting, T.P. and Zirkle, R.E.: *J. Cell. comp. Physiol.* 16:1975, 1940.
4. Shapiro, B., Kollmann, G. and Asnen, J.: *Radiat. Res.* 27:139, 1966.
5. Lindemann, B.: *Strahlentherapie*, 101:1, 1956.
6. Árky, I., Szász, I., Gárdos, G., Szélényi, J.G., Breuer, J.H., Várterész, V. and Hollán, S.R.: *Haematologia*, 3:51, 1968
7. Gerhardt, P.: *Fortschr. Roentgenstr.* 107:529, 1967
8. Dormandy, T.L.: *Brit. J. Haematol.* 20:457, 1971.

the blood stream during the time of exposure. Here a significant rise in plasma Hb values was found. In course of irrigoscopy, however, only the a. iliaca and eventually a small section of the abdominal aorta may be exposed. Here the plasma Hb values remained unaltered.

We attribute also the elevation of plasma Hb concentration, induced by low-level chronic, or acute, *in vivo* and *in vitro* exposure to ionizing radiation from external or internal sources, to the increase of erythrocyte membrane permeability, in other words, to the decrease of erythrocyte radioresistance. This alteration may be due to changes of intracellular enzymatic activity<sup>7</sup>, or to red cell lipid autooxidation. According to Dormandy<sup>8</sup>, the susceptibility of erythrocytes, even to atmospheric oxygen, can be considerably increased in this respect by their preliminary exposure to ionizing radiation, producing free radicals, particularly

ФИЗИОЛОГИЧЕСКИЕ ИССЛЕДОВАНИЯ ЦЕНТРАЛЬНОЙ НЕРВНОЙ  
И СЕРДЕЧНО-СОСУДИСТОЙ СИСТЕМЫ ЛИЦ, ПОДВЕРГАВШИХСЯ  
МНОГОЛЕТНЕМУ ПРОФЕССИОНАЛЬНОМУ ОБЛУЧЕНИЮ

В.К.Сельцер, В.В.Гампер, Г.М.Кацнельсон,  
Н.В.Лосева, А.И.Степанов  
Институт радиационной гигиены, Ленинград СССР

ABSTRACT

The group of 625 men and women subjected to mild doses of occupational radiation (less than 0.3 MPD) for a period over 10 years were under observation. Modern methods of physiological reactions and mathematical analyse were applied. No marked functional changes in central nervous and cardiovascular systems were revealed as compared with control group. No latent insufficiency of the functions was also found in both groups after functional loads.

Были проведены 10-ти летние наблюдения (1962-1971 гг.) за смешанной группой в 625 человек, подвергавшихся профессиональному действию ионизирующего излучения.

В исследуемую группу входили как сотрудники ряда научно-исследовательских институтов и лабораторий, так и рабочие и инженерно-технический персонал промышленных предприятий. 335 человек, работающих на тех же объектах в аналогичных условиях, но не подвергавшихся лучевому воздействию, составили контрольную группу.

Главным фактором профессиональной вредности являлось внешнее облучение:  $\gamma$ , жесткое- $\beta$  и рентгеновское. Производилась оценка радиационно-гигиенической обстановки, контролировались уровни гамма-фона в производственных помещениях, а на ряде объектов автоматической установкой непрерывного контроля регистрировалась радиоактивность воздуха. Дозиметрический контроль осуществлялся целым рядом систем наблюдения с помощью стационарной, переносной и индивидуальной дозиметрической аппаратуры. В ряде случаев основой контроля являлось измерение радиоактивности в биопробах (в моче).

Надёжная и безаварийная работа, систематический дозиметрический контроль, чёткая организация производственных операций в радиационно опасных зонах обеспечили то, что величина индивидуальных доз облучения в течение всего срока наблюдения была значительно ниже предельно допустимой для лиц, профессионально связанных с источниками ионизирующего излучения.

Как видно из таблицы 1, средние годовые дозы лучевого воздействия на протяжении 10 лет формируются довольно равномерно, составляя в среднем 1,02 р в год. В последние годы эта величина неуклонно снижается. Средняя суммарная доза, накопленная работающими за 10 лет, составила 10,21 р т.е. около 0,2 предельно допустимой дозы (ПДД). У половины обследованных суммарная доза за весь период не достигала 0,1 ПДД; у 1/3-она составляла 0,1-0,3 предельно допустимой дозы у 1/6-была выше 0,3 ПДД, но не превышала предельно

допустимый уровень облучения

Таблица I

Формирование дозовых нагрузок за 1962-71 гг./М ±m/

Г о д ы	Д о з а в р.	% годовой ПДД
1962	0,95 ± 0,08	19,0
1963	1,44 ± 0,06	28,8
1964	1,30 ± 0,07	26,0
1965	0,90 ± 0,06	18,0
1966	1,26 ± 0,08	25,2
1967	1,63 ± 0,06	32,6
1968	0,65 ± 0,03	13,0
1969	0,64 ± 0,04	12,8
1970	0,76 ± 0,05	15,2
1971	0,68 ± 0,04	13,6

Гамма-спектрография, проведенная на большом жидкостном стинциляционном счетчике человека показала, что практически нет разницы в уровне содержания радиоактивных изотопов в организме ряда профессионалов (где существовала потенциальная возможность инкорпорации) и у лиц контрольной группы. Наблюдениями установлена значительная неравномерность накопления дозы во времени и прерывистость облучения. Так, по данным еженедельного контроля около 46% рабочих недель проходит при лучевом воздействии, которое находится ниже уровня чувствительности ИФК; 9% - на уровне чувствительности, т.е. едва достигает величины 10 мр. на человека в неделю; 42% - в пределах от 10 до 100 мр. на человека в неделю и лишь 3% проходит в условиях превышения недельной дозы. Однако, и в пределах каждой недели имелась существенная неравномерность в накоплении дозы. Тщательный хронометраж производственных операций показал, что реальная длительность лучевого воздействия у абсолютного большинства лиц, колеблется в пределах 150-300 рабочих часов в год, т.е. составляет 10-20% годового рабочего времени. Исследование двух важных интегральных физиологических систем центральной нервной и сердечно-сосудистой производились ежедневно и были направлены на поиск возможных изменений уровня их функционирования.

Средний возраст лиц, составивших профессиональную группу, в начале исследования равнялся 30±1,2 года. Лиц, составивших контрольную группу, 31±1,7. Половой состав обеих групп был представлен приблизительно равным количеством мужчин и женщин.

Изучение центральной нервной системы проводилось обычными клиническими и клинико-лабораторными методами. При этом не было найдено существенных различий между группами. При изучении биоэлектрической активности записывалась фоновая электроэнцефалограмма, обработка которой осуществлялась графически с последующим анализом на ЭВМ. Применялись разнообразные функциональные нагрузки в том числе 3-х минутная дозированная гипервентиляция, вызывавшая рост амплитуды основного ритма (таблица 2).

Как следует из данных таблицы, за весь период наблюдений, у испытуемых обеих групп не отмечается сдвигов частоты основного ритма. В фоновой ЭЭГ не отмечено также каких-либо изменений выраженности общего количества основной активности (альфа индекс), а также пространственного распространения  $\alpha$ -активности в лобные отделы (затылочно-лобный градиент). Из данных таблицы также следует, что более тонкий показатель - коэффициент автокорреляции фаз фоновой ЭЭГ не изменяется на протяжении ряда лет. Несколько возрастает лишь показатель реакции основного ритма на гипервенти-

Таблица 2

Данные электроэнцефалографических наблюдений

Показатели	Время	Начало на блюдений	Через 5 л.	Через 10 л.
	группа			
Частота $\alpha$ -ритма/в гц/	контр.	10,3 $\pm$ 0,02	9,0 $\pm$ 0,03	10,1 $\pm$ 0,02
	проф.	9,5 $\pm$ 0,01	9,9 $\pm$ 0,03	10,5 $\pm$ 0,02
$\alpha$ -индекс	контр.	82,0 $\pm$ 4,2	78,0 $\pm$ 3,9	77,6 $\pm$ 6,0
	проф.	81,0 $\pm$ 1,5	73,4 $\pm$ 6,9	75,1 $\pm$ 4,0
Затылочно-лобный градиент	контр.	1,8 $\pm$ 0,01	1,7 $\pm$ 0,05	2,0 $\pm$ 0,04
	проф.	1,9 $\pm$ 0,07	2,2 $\pm$ 0,02	2,1 $\pm$ 0,03
Коэффициент автокор- реляционной функции	контр.	0,6 $\pm$ 0,01	0,5 $\pm$ 0,02	0,6 $\pm$ 0,01
	проф.	0,6 $\pm$ 0,01	0,5 $\pm$ 0,01	0,6 $\pm$ 0,02
Реакция на дозирован- ную гипервентиляцию	контр.	1,3 $\pm$ 0,01	1,3 $\pm$ 0,02	1,2 $\pm$ 0,01
	проф.	1,4 $\pm$ 0,01	1,3 $\pm$ 0,01	1,2 $\pm$ 0,02

ляционную нагрузку, что связано, по-видимому, с возрастным уменьшением реактивности мозга на развивающуюся при гипервентиляции гипоканию. Эти изменения наблюдаются также в обеих группах.

Наблюдалась также небольшая группа лиц, которые в начале трудовой деятельности в 40-ых и первой половине 50-ых годов подвергались переоблучению. Реконструктивно восстановленные суммарные дозы, накопленные за первые 3-5 лет работы с источниками излучения у этих лиц составляли 150-200 рентген. У некоторых из них в тот период диагностировались лучевые реакции.

При первых обследованиях в 1961-62 годах у этих лиц наблюдались изменения биоэлектрической активности, повышенный уровень синхронизации биопотенциалов мозга. Реакция на гипервентиляционную нагрузку была незначительной; порог возникновения медленной активности при этом понижен. У этих же лиц обнаруживались симптомы вегетативно-сосудистой дистонии: лабильность пульса, непостоянство артериального давления со склонностью к гипотонии. Однонаправленность изменений гемодинамики и биоэлектрической активности мозга находит объяснение в работах Mounier M. 1963 г.<sup>1</sup> и других, установивших тесную функционально-морфологическую связь центров сосудистой регуляции с образованиями, ответственными за синхронизирующий эффект, расположенными в области гипоталамуса и ретикулярной формации ствола мозга. Полученные данные совпадают с наблюдениями Ливанова М. Н. (1962 г.).<sup>2</sup>

В последних исследованиях в 1970-71 годах наблюдавшиеся у упомянутых лиц особенности биоэлектрической активности мозга и гемодинамики сгладились, и функциональные показатели обеих систем уже ничем не отличались от таковых у лиц основной группы.

Данные исследования состояния сердечно-сосудистой системы по основным гемодинамическим показателям представлены в таблице 3. Знакомство с содержанием таблицы показывает, что ни по группе общих гемодинамических характеристик (показатели 1-5), ни по характеристикам тонуса магистральных сосудов (6-7), ни по показателям структуры сердечного сокращения и систолы левого желудочка (8-12), ни по объёмно-энергетическим параметрам сердца (13-17) не определяется сколь-либо значительных различий по группам. Средние показатели покоя соответствуют нормальному уровню этих характеристик присущих людям данного возраста. Материалы таблицы отражают также устойчивость важнейших функциональных показателей сердечно-сосудистой системы у профессионально облучавшихся лиц на

Таблица 3

## Функциональные характеристики кровообращения х)

Показатель	Начало наблюдений		Через 5 лет		Через 10 лет	
	контр.	профес.	контр.	профес.	контр.	профес.
1. ЧСС 1'	70±0,6	72±0,3	67±0,5	70±0,2	72±0,5	72±0,4
2. АДс мм.рт.ст.	116±1,2	115±0,9	118±0,8	119±0,6	129±1,1	127±0,7
3. АДд мм.рт.ст.	75±0,4	73±0,3	77±0,5	78±0,3	81±0,6	80±0,6
4. МОК л.	3,9±0,1	3,8±0,1	3,6±0,1	3,5±0,1	3,9±0,1	3,8±0,1
5. ПС дин.	1990±40	2010±70	2170±70	2180±40	2190±70	2220±60
6. СРПВэ см/сек	585±16	610±9	620±11	635±14	655±12	660±16
7. СРПВм см/сек	640±11	670±8	690±14	720±12	740±16	730±15
8. СП	0,39±0,002	0,39±0,002	0,38±0,002	0,39±0,002	0,40±0,002	0,41±0,001
9. АС сек.	0,06±0,001	0,06±0,001	0,06±0,001	0,06±0,001	0,06±0,001	0,06±0,001
10. ИС сек.	0,04±0,001	0,05±0,001	0,05±0,001	0,04±0,001	0,04±0,001	0,04±0,001
11. Н сек.	0,10±0,002	0,11±0,001	0,11±0,002	0,10±0,002	0,10±0,001	0,10±0,001
12. Е сек.	0,27±0,002	0,26±0,001	0,27±0,003	0,26±0,002	0,26±0,002	0,26±0,001
13. НДИС мм.рт.ст./сек	2155±60	1930±40	1920±70	2050±50	2270±60	2290±50
14. ОСВ мл/сек	189±3	196±4	190±4	194±2	196±4	188±4
15. ЕМОК сек.	16,4±0,2	16,3±0,1	16,3±0,2	16,5±0,2	16,6±0,2	16,5±0,1
16. ЭСС джоули	0,58±0,006	0,59±0,004	0,59±0,005	0,60±0,002	0,61±0,002	0,61±0,005
17. МЛЖ ватт	2,26±0,02	2,29±0,02	2,31±0,02	2,27±0,03	2,33±0,04	2,30±0,04

х) ЧСС-частота сердечных сокращений; АДс-артериальное давление систолическое; АДд-артериальное давление диастолическое; МОК-минутный объем крови; ПС-периферическое сопротивление; СРПВ-скорость распространения пульсовой волны по сосудам эластического (э), мышечного (м) типа; СП-систолическое отношение Фогельсона-Черногорова; АС-фаза асинхронного сокращения; ИС-фаза изометрического сокращения; Н-период напряжения; Е-период изгнания; НДИС- нарастание давления в изометрическую фазу; ОСВ-объемная скорость выброса; ЕМОК-время изгнания минутного объема крови; ЭСС-энергия сердечного сокращения; МЛЖ-мощность левого желудочка.

протяжении 10 лет исследования.

В таблице 4 представлены данные об изменении основных гемодинамических показателей, возникающих сразу после выполнения стандартной физической нагрузки. Как видно из этой таблицы, у лиц основной группы имелась адекватная реакция сердечно-сосудистой системы, не отличающаяся от таковой у лиц контрольной группы. Применение разнообразных функциональных проб (орто-и клиностатической, Ашнера-Данини, холодной и др.) также не выявляет различий в реакциях системы кровообращения в обеих группах. Это говорит об отсутствии скрытой недостаточности функции у лиц профессионально связанных с излучением.

Таблица 4  
Изменения гемодинамических показателей после дозированной нагрузки (20 приседаний за 30 сек) в % к исходной величине

Показатель	Начало наблюдений		Через 5 лет		Через 10 лет	
	контр.	профес.	контр.	профес.	контр.	профес.
1. ЧСС	+40±2,0	+37±2,0	+43±3,0	+41±2,0	+44±2,0	+43±1,0
2. АДс	+10±1,0	+11±1,0	+9±1,0	+8±1,0	+9±0,5	+10±0,4
3. АДд	-5±0,3	-4±0,3	-6±0,2	-6±0,3	-4±0,2	-5±0,2
4. МОК	+72±4,0	+69±2,0	+76±3,0	+74±4,0	+66±4,0	+75±3,0
5. ПС	-39±1,0	-37±1,0	-42±2,0	-41±2,0	-37±1,0	-44±2,0

Как следует из таблицы 5 около 90% лиц, работавших с ионизирующим излучением и столько же лиц контрольной группы имели нормальные электрокардиографические кривые на протяжении всего обследования.

Таблица 5  
Электрокардиографические характеристики (% от наблюдений)

Показатель	время группа	Начало наблюдений		Через 10 лет		
		контр.	профес.	контр.	профес.	
Электрическая ось сердца	нормограмма	26,9	31,8	25,9	26,4	
	правограмма	37,7	33,8	28,8	30,2	
	левограмма	26,2	27,6	33,2	32,8	
Нормальные	Правильный синусовый ритм	53,9	44,9	52,9	45,0	
	Синусовая брадикардия	умеренная	24,8	29,5	24,4	26,9
		выраженная	3,8	6,3	4,2	5,4
	Синусовая тахикардия	умеренная	0,6	0,9	0,7	1,4
Синусовая аритмия	умеренная	4,6	3,9	3,0	3,9	
	выраженная	3,1	7,7	2,7	6,8	
Частичная блокада правой ножки пучка Гиса		24,4	28,0	26,8	27,1	
Патологические	Недостаточность коронарного кровообращения	2,4	1,9	3,7	3,2	
	Диффузные мышечные изменения	5,4	3,6	5,8	4,7	
	Желудочковая экстрасистолия	0,8	0,7	1,6	1,8	
	Блокада ножки пучка Гиса (правой или левой)	0,6	0,6	1,0	0,9	

Это несколько превышает процент нормальных ЭКГ-кривых, выявляемых при массовых обследованиях соответствующих возрастным групп населения, что можно объяснить имевшим место медицинским отбором при поступлении на работу. Выявленные патологические изменения, как правило, носят невыраженный характер: это признаки недостаточности кровообращения миокарда, нарушение возбудимости с переходящим появлением дополнительных очагов водителя ритма, непостоянная брадиаритмия и др.. Некоторое нарастание патологически измененных электрокардиограмм у лиц обеих групп, по всей вероятности, связано с увеличением возраста наблюдаемых.

Таким образом, многолетнее изучение функционального состояния сердечно-сосудистой системы по основным гемодинамическим показателям в покое не выявило каких-либо специфических изменений, а также динамики, связанной с увеличением стажа. Адекватная реакция в ответ на применение физической нагрузки свидетельствует об отсутствии скрытой недостаточности функции.

Итоги 10-ти летних наблюдений позволяют утверждать, что реальные дозы профессионального лучевого воздействия в различных областях народного хозяйства, как правило, не превышают 0,3 ПДД в год. Временные особенности накопления доз в огромном большинстве таковы, что длительность лучевого воздействия составляет лишь незначительную часть биологического времени. Все это делает мало вероятной возможность соматических проявлений профессионального облучения.

#### ЛИТЕРАТУРА

1. MOUNIER M. Physiologie und Pathophysiologie des vegetativen Nervensystems. Stuttgart Hippocrates Verl., 1963.
2. ЛИВАНОВ М.Н. Некоторые проблемы действия ионизирующей радиации на нервную систему. М., 1962.

# TRIAL EVALUATION OF THE LONG-TERM SURVEY OF DEATH CAUSES IN THE POPULATION LIVING NEAR RADIATION RICH HOT SPRING

Kazuho Maeda

Faculty of Medicine, University of Tokyo, Tokyo, Japan

Shiro Abo

National Institute of Radiological Sciences, Chiba, Japan

Yoshiyasu Kurokawa

Power Reactor & Nuclear Fuel Development Corp., Tokyo, Japan

Yoji Minagawa

Nippon Telephone & Telegram Public Corp., Tokyo, Japan

## Abstract

An attempt was made to distinguish the effect of long-term exposure with very low dose rate ionizing radiation applying a type of retrospective cohort analysis on the population living adjacent to a hot spring rich in radium and radon emanation. Randomly sampled households were interviewed and inquired causes of death of the family members, over three generation. Death cases collected were tabulated according to the List B by sex, and compared the rank of these B causes tabulated from the sample with that of control.

Noticeable difference could not be found between the sample and control in this investigation. The detailed explanation of the cohort type analysis which was used in this study — Regional Cohort Analysis — are presented, and a feasibility of its application for the evaluation of the effect of long term and low dose exposure of agents such as ionizing radiation are discussed.

## Introduction

Many epidemiological studies have been done to investigate the effect of long-term exposure with low dose rate ionizing radiation for man. Most of these studies had concern with some incidences which are believed to be induced by radiation such as leukemia. They did not always succeed in showing the significant findings. In general, the incidences induced by radiation are rare phenomena, so it is difficult to plan a epidemiological study aimed at such diseases.

The authors have planned a trial epidemiological study stressed on the continuation of characteristics of one's environment. Radium rich hot spring, Misasa hot spring located in the northwest part of Japan, has been selected as our field of survey. Sample population has stayed adjacent to the hot spring and passed their lives in their home local area. Randomly selected households were interviewed and inquired the causes of death cases occurred in their family, which extended over three generations.

Control households were also staying near the hot spring and passed their lives under the exclusively same social and cultural conditions as that of sample, however there are not hot spring in their area.

## Method of Survey

The authors believe that in a kind of cohort analysis, the subjects should be ensured in their uniformity on the matters refers to. Therefore, ordinal cohort analysis should be done with a population born in a same year. Usually it does not add new cases to a cohort during its observation, except in a special project like as the follow up study on the A-bomb survivors.

In this study, we adopted a cohort type analysis which deals with regional oriented cohort. The cases are put sequentially into the cohort and are defined



as following.

A man who was born in his home local area, grew and lived through their whole life at the same place, and finally died there is the basic constituent of this cohort irrespective to the year of his birth. The individual cases, although they might have passed under much different conditions, should be under the same long lasting natural environment such as the back ground radiation. So, the authors assured that the uniformity of subjects did not disturb owing to the sequential addition of individual cases to the starting group.

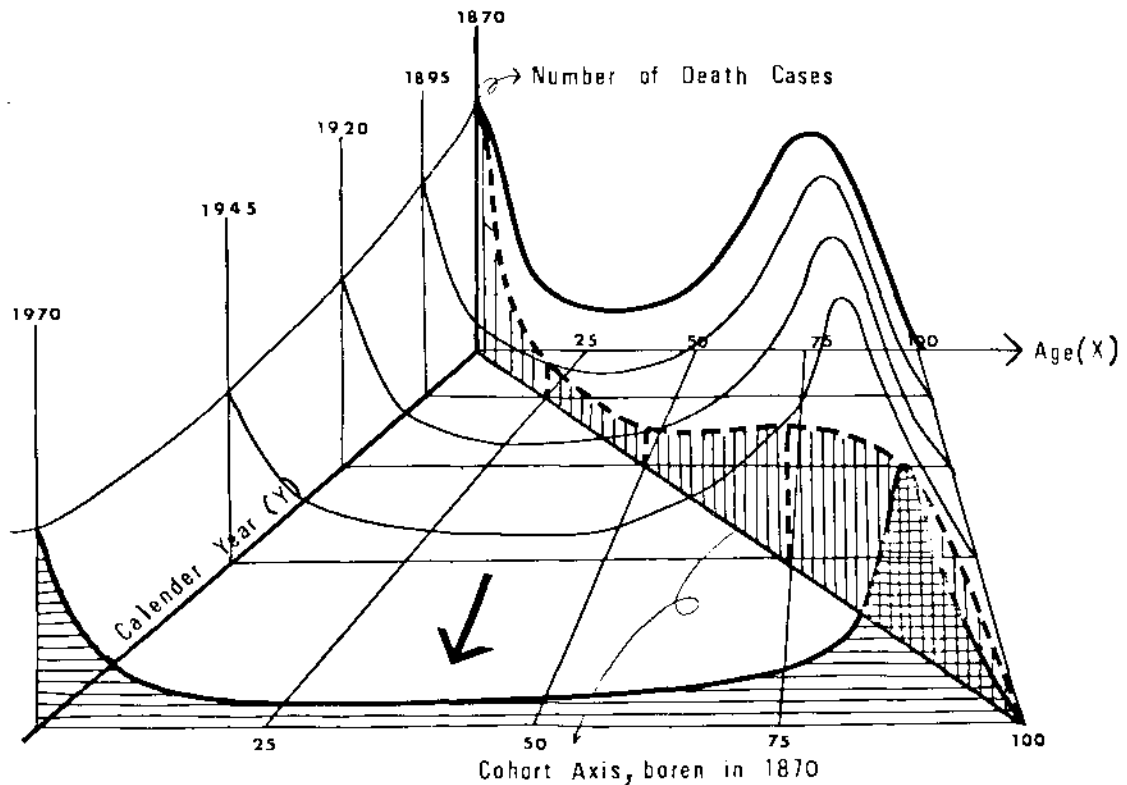


Fig. 1 SCHEMATIC ILLUSTRATION OF REGIONAL COHORT OF DEATH CASES

Fig. 1 shows the scheme of our regional cohort. The curved surface standing on the XY plane expresses the all death cases occurred in an area from the year 1870 to 1970. The vertical plane obliquely crossed the surface means a cluster of death cases recorded in the cohort born in the year 1870. These consist of the subject group with which performs the ordinal cohort analysis. The subject of our regional cohort is included in the part of curved surface bounded by the vertical oblique plane and downward along the arrow to the lower end of the surface.

In the illustration of the figure, it may be clear that in the regional cohort results to include relatively much cases which died in younger ages. It might be rather desirable to collect many younger death cases in promoting some specific survey project. To evaluate the effects of low level radiation, it should be mind to identify the disorders that might be occurred due to genetic influences of radiation.

Although unlikely, in the retrospective study, it does seem very difficult to identify details for each death cases. Unfortunately in our trial survey, there were not any additional informations related to the collected death cases. Therefore, it precludes making the most of the merit of the regional cohort

mentioned above in present analysis. For that reason, the all death cases inquired from the sample households and the control have been used as the subject of our investigation. It can be schematically expressed that the subject of this study distributed over the all surface of curved surface in Fig. 1.

A household who moved into the area, where the sample households and the control were selected, was excluded as the sample subject. However, a case of woman who had married with a man in the area, and had been a resident of the region until the end of her life was adopted as the subject of analysis.

Twentyone households were interviewed and collected 165 death cases, and from the 24 households 183 cases were available as the control.

Indoor air gamma ray dose rate of the all households was measured using a survey meter with a NaI (Tl) scintillator. The average dose rate of the households in hot spring area was  $13.1 \pm 1.9$  (st. dev.)  $\mu\text{R/hr}$ , and that of in the control area was  $11.8 \pm 2.0$   $\mu\text{R/hr}$ . Both averages are higher than a Japanese average (8  $\mu\text{R/hr}$ ). The difference of the air gamma ray dose rate of both areas is not significant. However, people who lived in this sample area had taken a bath every day in the natural hot spring, and had habitually drunk the water of those springs. It was because, that the rustic Japanese people believed, not only taking a bath in hot spring but drinking the spring water was good for their health.

It can be suggested that people who had stayed in the hot spring area would take a considerable amount of Ra and Rn into their body, and would be exposed by internal deposited radioisotopes. In the present study, we did not measure the contents of radioisotopes in the spring water and in the water of wells which are daily used by the residents. So, we can't propose any statements suggesting the difference in the level of internal exposure between the sample subjects and the control.

### Results and Discussion

Collected death cases were tabulated according to the List B by sex. The oldest death cases certified in the year 1871 and the most recent one occurred in 1972. The tabulation of the List B shows in Table 1 (sample groups) and 2 (control). They are in line in the order of number of cases belongs to each B causes. There are not any noticeable difference of the order of death causes in all tabulations between the sample groups and control. If we would have found some difference in the order of death causes, we could test the significance using the rank correlation test. In the male tabulation, there appears, in rather high order, the injury resulting from operation of war (BE50c) in both groups. It occurred exclusively in the World War II. Based on the results now mentioned, we did not get significant findings on the effect of long-term exposure with very low dose rate of ionizing radiation in this investigation. However, provided that there were some other informations concerning to the causes of collected death cases, we would have gotten some interesting results associated with the purpose of our project.

The definition of a individual available to join the group of regional cohort analysis is so clear and easy that this analytical method can be applied even in an area living a large population. We hope to continue the project using our survey method applying to large population than reported in this presentation.

When some significant result is obtained through an investigation aimed at the effect of low dose rate radiation, it should be examined elaborately from various points of view. It is, of course, necessary to investigate not only on the causes of death but on many other factors associated with the mortality in the local population. Although unlikely, after repeated careful investigation using regional cohort analysis, it can be expected to suggest a basis to be able to comment on the present principles of risk evaluation on low level exposure of radiation — linear to the extent zero exposure.

TABLE 1. LIST OF MAIN DEATH CAUSES OF THE SAMPLE POPULATION  
(AREA ADJACENT TO THE HOT SPRING)

Male			Female			Both Sexes		
1. B45a	Senility	20	1. B45a	Senility	14	1. B45a	Senility	34
2. B30	CVD*	13	2. B30	CVD	13	2. B30	CVD	26
3. B5	Pulmon. tbc.	11	3. B19	Malig. neoplasms	7	3. B5	Pulmon. tbc.	13
4. BE50c	Injury resulting from war	11	4. B29	Other diseases of heart	3	4. B19	Malig. neoplasms	12
5. BE48	Accidents	6	5. B41	Complication of pregnancy	3	5. BE50c	Injury resulting from war	11
6. B19	Malig. neoplasms	5	6. B5	Pulmon. tbc.	2	6. BE48	Accidents	7
7. B32	Pneumonia	4	7. B34	Peptic ulcer	2	7. B34	Peptic ulcer	6
8. B34	Peptic ulcer	4				8. B32	Pneumonia	5
	All other cause groups and unknown causes	26		All other cause groups and unknown causes	26		All other cause groups and unknown causes	51
	Total	100		Total	65		Total	165

\*CVD : Cerebrovascular disease

TABLE 2. LIST OF MAIN DEATH CAUSES OF THE SAMPLE POPULATION  
(CONTROL AREA)

Male			Female			Both Sexes		
1. B45a	Senility	27	1. B45a	Senility	28	1. B45a	Senility	55
2. B30	CVD	15	2. B30	CVD	12	2. B30	CVD	27
3. BE50c	Injury resulting from war	9	3. B19	Malig. neoplasms	9	3. B19	Malig. neoplasms	17
4. B19	Malig. neoplasms	8	4. B29	Other diseases of heart	5	4. BE50c	Injury resulting from war	9
5. BE48	Accidents	7	5. B28	Ischaemic heart disease	3	5. BE48	Accidents	8
6. B2	Typhoid fever	4	6. B32	Pneumonia	3	6. B29	Other disease of heart	7
7. B32	Pneumonia	4				7. B32	Pneumonia	7
	All other cause groups and unknown causes	30		All other cause groups and unknown causes	19		All other cause groups and unknown causes	53
	Total	104		Total	79		Total	183

References

1. Hiroyuki Toyokawa, Personal communication
2. Virginia A. Clark, Carl E. Hopkins, J. Chron. Dis. 20, 565-569, 1967

Acknowledgements : This study was sponsored by the Nuclear Safety Research Association Japan.

X-RAY EFFECTS ON THE EMBRYO AND FETUS:  
A REVIEW OF EXPERIMENTAL FINDINGS

Roberts Rugh and William Leach  
Division of Biological Effects, Bureau of Radiological  
Health, FDA, USPHS, Rockville, Maryland 20852 U.S.A.

ABSTRACT

Concerns for protection of the fetus from possible effects of ionizing radiation can be justified on the basis of results from abundant mammalian experiments which deal with radiation teratogenesis during embryonic and fetal pre-natal development. Experimental results relative to teratogenesis indicate that early stages of development may be more sensitive to radiation than later stages of development. Animal data on radiation teratogenesis thus supports concerns for radiation protection during early pregnancy. Doses of 25 rads to the fetus during organogenesis are associated with teratogenesis. During early embryonic development teratogenesis has been observed after doses of 10 rad or less to the fetus. Some human diagnostic x-ray procedures exist which deliver doses of radiation associated with teratogenesis, when such doses are administered to fetal animals at appropriate periods of development.

. . . . .

Probably the most radiosensitive biological entity is the cell of the embryo or fetus during its differentiation into its final form. Irradiation of embryonic cells may lead to effects which include immediate cell death, nuclear pycnosis after a few hours, congenital anomalies seen at birth, or malignancies which may appear after a variably long period of latency. There are at least two factors associated with the high radiosensitivity of the embryonic and fetal mammal. First, each division subsequent to fertilization of an ovum gives rise to new clones of cells and proliferation is rapid. Second, if the progenitor of a clone of cells has its chromosomes altered by radiation and survives to produce a clone of cells, all descendant cells of that clone will be similarly altered. The consequence of this radiation induced change may include the development of a birth anomaly. The earlier in development an embryo is traumatized, the more far-reaching would be any effects that could be produced. Cells irradiated at later stages of development will be progenitors of still future clones, however, with more limited involvement for the fetus as a whole. Once the mosaic of cells of a fully formed organism is derived, damage to a single cell may be relatively inconsequential to the whole organism in terms of teratogenic risk. Experiments in animals have demonstrated that radiation doses of 10 rads or less administered to the mammalian embryo during early developmental phases can be teratogenic or even lethal.<sup>1-6</sup> It has been shown that 25 roentgens to the developing rodent can destroy neuroblasts. These important precursors of neural tissue are present in the fetus throughout its developmental period, and persist into early postnatal life.<sup>5,6</sup> Thus, our first proposition is that irradiation of the mammalian embryo and fetus is associated with a risk. The risk is greater during early embryonic and fetal development, and may be expressed in a number of dose dependent phenomena including teratogenesis and fetal death. A series of charts showing x-ray anomalies in animals are presented on succeeding pages. The most common anomalies attributed to radiation relate to the central nervous system. This may be due to the embryological fact that this system not only makes its appearance very early in development, but it continues to differentiate through gestation and into the neonatal period. Thus, while the first trimester is the most radiosensitive, the entire

gestation period must be considered as radiosensitive, particularly for the developing nervous system.<sup>7</sup>

Our second proposition is that the fetus of a woman whose pregnancy is unrecognized could be exposed to diagnostic radiations. It has been reported<sup>8</sup> that 23 percent of pregnant women in this country in 1963 were exposed to ionizing radiations, 21 percent of these were exposed during their first and most critical trimester, and 13 percent of these will be abdominal exposures. In a recent year when there were 3.5 million live births, this would have meant about 805,000 pregnant women were exposed to diagnostic radiations, and 22,000 embryos or fetuses could have been x-irradiated during their most critical stages of development.<sup>8</sup> The importance of radiation protection of fetuses from possible anomaly production is emphasized when one recognizes that approximately 7 percent of children are born each year with congenital anomalies, and possibly a million pregnancies never achieve birth.<sup>8</sup> Although the proportion of these anomalies and fetal deaths which may be due to radiation is not known, the elimination of avoidable fetal irradiation during pregnancy should result in a reduction of defects which may be related to radiation exposure.

Our third proposition is that dose relations cannot be extrapolated directly from mouse to man. The mouse is 15 times more radiosensitive than *Drosophila* to radiation-induced mutations.<sup>10,11</sup> The human embryo or fetus could be either more or less radiosensitive than either *Drosophila* or the mouse. However, fundamental biological reactions to radiation exposure in man and animals are qualitatively the same, though dosimetric and time relations may vary. Epidemiologic data suggests that low levels of radiation in humans during the third trimester of pregnancy may be associated with a small but significantly increased risk of childhood leukemia, though considerable controversy exists in this area.<sup>1,2</sup>

"CONGENITAL ANOMALIES REPORTED FOR THE MAMMAL AS DUE TO EMBRYONIC OR FETAL X RADIATION"\*\*\*  
ROBERTS RUGH - DREW, PH.D., F.D.A., BRH, DBE

ANOMALY	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	
ABDOMINAL LESION																							
ABORTION, INDUCED																							
ACRANIA																							
AGNATHIA																							
ALBINISM																							
AMAUROSIS																							
AMELIOGENESIS IMPERFECTA																							
ANAL ATRESIA																							
ANEMIA																							
ARACHNOIDAL																							
ANKYLOBLASTIC																							
AROPHTHALMIA																							
ANASTOMIA																							
ANGLIA																							
APROSOPIA																							
AQUEDUCT, NARROW																							
ARRHINOCEPHALIA																							
ATLAS, ABNORMAL																							
BLINDNESS																							
BLISTERS, CRANIAL																							
BLOOD VESSELS																							
BUSH MARK																							
BONE & JOINT ANOMALIES																							
BRACHYDACTYLY																							

These anomalies have been reported in the literature from experiments and observations on a variety of mammals, principally rodents (mouse, rat, and hamster), but also dogs and humans. Below the table are listed the comparable developmental ages for the human based upon the differentiation of specific organ systems. No data could be included as to the dose in rads which produced the above results because such data would differ somewhat with species. This is therefore a table of radiosusceptible stages of gestation as derived from rodent studies with possible extrapolation to the human. Italics is included as an anomaly since it may well result from anomalous development.

NOTE: The extrapolated gestation days for the human represent the earliest comparable date, but usually an exact comparable date for the human would span a period of several days to weeks, especially as development progresses. (Continued)

Our fourth proposition is that of the gamut of diagnostic procedures, of which there are 66 that present some possible risk to the human embryo or fetus  
 10 procedures could deliver as much as 4 rads to the gravid uterus,  
 and three of these could deliver up to 20 rads:

- Abdominal aortography - 20 rads (with fluoroscopy and/or ciné)
- Barium enema - 6 rads
- Carcinoma of the cervix - 6 rads (with fluoroscopy)
- Celiac angiography - 20 rads (with fluoroscopy and/or ciné)
- G.I. series, lower tract - 6 rads
- Hysterosalpingography - 6 rads
- Pelvimetry - 4 rads
- Placentography - 4 rads
- Renal arteriography - 7.5 rads
- Urethrocytography - 20 rads (with fluoroscopy and ciné)

The physician must therefore consider and balance the benefit to the mother against the risk to the fetus. Since it is generally accepted that the effects of radiation may be cumulative, the aggregate radiation exposure of several procedures, if multiple diagnostic procedures are performed, may also need to be considered in weighing benefit vs. risk.

Our fifth proposition is that certain diagnostic radiation procedures can deliver doses to the mammalian embryo which carry a high risk of anomaly production based on experimental evidence. It has been demonstrated that 10 rads can kill the early mouse embryo<sup>2</sup> and 25 rads can destroy neuroblasts<sup>3,7</sup> during early gestation.

ANOMALY	GESTATION DAYS (Mouse)	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	
BRAIN, APLASIA																								
BRAIN STEM ANOMALY																								
BEHAVIOR																								
CALCANEAL TUBEROSITY																								
CARDIAC INVERSION																								
CARDIO VASCULAR ANOMALIES																								
CARTILAGE, MALFORMED																								
CATARACTS																								
CAUDATE NUCLEUS																								
CENTRAL ANKYLOSIS OF																								
CENTRAL NERVOUS SYSTEM																								
CEREBELLUM, CEREBELLAR FOLIA																								
CEREBRAL ATROPHY																								
CEREBRAL VESICLES MALFORMED																								
CERVICAL VERTEBRAE DISLOCATED																								
CHEST, FUNNEL																								
CHONDRODYSPLASIA																								
CHORIO-RETINITIS																								
CHROMOID PLEXUS, ATROPHIC																								
CHROMOSOME ABERRATIONS																								
CIRCULATORY SYSTEM MALFORMED																								
CLAW, PERSISTENT																								
COLOBOMA																								
CORPUS CALLOSUM DEFORMED OR ABSENT																								
CRANIAL BLISTER																								
CRANIAL NERVES ALTERED																								
CRANIOSCHISIS																								
CRANIUM, VAULTED																								
CYCLOPIA																								
DEGENERATION, GENERAL																								
DIGESTIVE SYSTEM ANOMALIES																								
DIGIT, ANNULAR GROOVE IN 5th																								
DIGITAL ANOMALIES																								
DIGITAL REDUCTIONS																								
DUCTUS ARTERIOSUS, PERSISTENT																								
DYSURCHIA, ABDOMINAL																								
EAR, DEFORMED																								
ECTRODACTYLIA																								

Our sixth proposition is that a period of radiation safety, i.e., no possibility of fetal radiation, for the above listed diagnostic procedures exists during the early part of the female estrus cycle. It is known that a woman with a steady 28-day menstrual cycle probably ovulates between 10 and 17 days, most likely on day 14. If a woman menstruates normally, it can be assumed that she cannot conceive for 10 days after its onset (although it has been stated that conception has on rare occasions occurred on the seventh day of the cycle). This fact has given rise to the so-called 10-day rule, which attempts to minimize fetal radiation risks by performing elective diagnostic procedures during the period of the estrus cycle when possibility of pregnancy is minimal.

ANOMALY	GESTATION DAYS (Mouse)																					
	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21
EDEMA																						
ENCEPHALITIS																						
ENDOCARDIAL CUSHION ANOMALIES																						
EPILEPSY																						
ENCEPHALOCYCLE																						
ENCEPHALOPATHY																						
ENDOCRINE DISTURBANCES																						
ENZYMES, MALFUNCTIONS OF																						
EXENCEPHALY																						
EXOPHTHALMOS																						
EXOSTOSIS																						
EXTREMITIES, DISTURBANCES OF																						
EYE DEFECTS																						
EYELIDS, OPEN AT BIRTH																						
EXTROPHY OF THE BLADDER																						
FEET, CLUB																						
FETAL DEATH																						
FOOT, VOLAR FLEXION OF																						
CANGLION, BASAL ANOMALIES OF																						
GASTROSCHISIS																						
GENITO-URINARY ANOMALIES																						
GERM CELL DEPLETION																						
GIRDLE, PELVIC MALFORMATIONS OF																						
GLAUCOMA, CONGENITAL																						
GONAD, DEGENERATE OR MISSING																						
GROWTH RETARDATION																						
HEMANGIOMA																						
HARELIP & CLEFT PALATE																						
HEAD NARROW																						
HEAD, DEFECTS OF																						
HEARTING ANOMALIES																						
HEART ANOMALIES																						
HETEROTROPIA																						
HIP, CONGENITAL DISLOCATION OF																						
HIPPOCAMPUS, DEFICIENT																						
HYDROCEPHALUS																						
HYDROCELE																						
HYDROMYELY																						
HYDRONEPHROSIS																						
HYDROURETER																						
HYPERMETROPIA																						
	GESTATION DAYS (Human Extrapolated)																					
	2	3	4	5	9	14	20	25	27	30	36	38	45	55	80	(Cont'd)						





ANOMALY	GESTATION DAYS (MOUSE)																					
	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21
SNOUT, SHORT OR ANOMALOUS																						
SPINA BIFIDA																						
SPINAL CORD ANOMALIES																						
STERILITY																						
STERNAL ELEMENTS' LOSS																						
STRABISMUS																						
STUNTING																						
SYNASTOSIS																						
SYNOCTYLIA																						
TAIL ANOMALIES																						
TESTES DEGENERATE OR ABSENT																						
THORACO-ABDOMINAL HERNIA																						
THORACIC & CERVICAL SEGMENTAL JUMBLING																						
TUMOR, LATE DEVELOPMENT																						
URINOGENITAL SYSTEM																						
VASCULAR ANOMALIES																						
VENTRICLES, DILATION OF I & III																						
VERTEBRAL COLUMN																						
VISCERAL HERNIA																						
WEBBED FINGERS AND/OR TOES																						
WEIGHT REDUCTION																						

GESTATION DAYS (Human Extrapolated)

2 3 4 5 9 14 20 25 27 30 36 38 43 55 80

EXTRAPOLATION TABLE FOR MOUSE AND HUMAN DEVELOPMENT (DAYS)

MOUSE	HUMAN	MOUSE	HUMAN
0	1	9	25-1/2
1	2	9-1/2	26
2	3	10	27
4	4	10-1/2	28-1/2
5	5-6	11	30-3/4
5-1/2	7-8	11-1/2	33-1/2
6	9-10	12	36
6-1/2	11-13	12-1/3	36-1/2
8	14-17	13	38
8-1/3	18-20	14-1/2	47
8-1/2	20-1/2	15-1/2	65+
8-2/3	21	16-1/2	84+
8-4/5	24-1/3		

Extrapolations based entirely upon comparable development of specific organ systems. It is clear that after mouse day 13 the extrapolations are not reliable since development from then to birth is more concentrated for the mouse than for the human.

Anomalies listed above 226.

REFERENCES

- Rugh, R., 1965. "Definition and limits of the concept of 'small dose' of ionizing radiations in mammals." Excerpta Medica Int. Congr. Proc. XIth Int. Congr. Radiol., Rome.
- Rugh, R., M. Wohlfromm, A. Varma, 1969. "Low dose x ray effects on the pre-cleavage mammalian zygote." Rad. Res. 37:401-414.
- Rugh, R., 1971. "X-ray teratogenesis in the mouse and its possible significance to man." Radiology 99:433-443.
- Jacobsen, L., 1968. "Low dose x irradiation and teratogenesis." Acta. Path. Scand. (Suppl. 193).
- Hicks, S.P. 1954. "Mechanism of radiation anencephaly, anophthalmia, and pituitary anomalies: Repair in mammalian embryo." Arch. Path. 57:363-378.
- Hicks, S.P., C.J. D'Amato, M.J. Lowe, 1959. "The development of the mammalian nervous system. I. Malformations of the brain, especially the cerebral cortex, induced in rats by radiation. II. Some mechanisms of the malformations of the cortex." Jour. Comp. Neurol. 113:435-469.
- Rugh, R., 1959. "Ionizing radiations: Their possible relation to the etiology of some congenital anomalies and human disorders." Mil. Med. 124:401-416.
- Brown, M.S., P.L. Roncy, J.N. Gitlin, R.T. Moore, 1967. "X-ray exposure during pregnancy." J.A.M.A. 199:309-314, and "Medical x-ray visits and examinations during pregnancy, United States 1963." DHEW, National Center for Health Statistics, Series 22, No. 5, 1968.
- Apgar, V., G. Stickles, 1968. "Birth Defects: Their significance as a public health problem." J.A.M.A. 204:79-82.
- Russell, W.L., 1955. "Genetic effects of radiation in mice and their bearing on the estimation of human hazards." Proc. Int. Cong. Peaceful Use of Atomic Energy, Geneva, 11:382-383 and 401-402.
- Russell, W.L., 1956. "Comparison of x-ray induced mutation rates in Drosophila and mice." Am. Nat. 90:69-80.
- Stewart, A., 1973. "An epidemiologist takes a look at radiation risks." DHEW Publication (FDA) 73-8024, BRH/DBE 73-2.

STUDY OF THE ADRENAL FUNCTION IN SUBJECTS WITH LONG-TERM  
LOW LEVEL OCCUPATIONAL EXPOSURE TO IONIZING RADIATIONS  
H.I.Popescu and Ioana Lancranjan  
Institute of Hygiene and Public Health, Bucharest, Romania

Abstract

A study of 24 hr urine elimination of 17-hydroxycorticosteroids (17 OH) by Poster-Silber's method was carried out in 115 men (mean age 40 years) with long-term low level occupational exposure to penetrating rays (mean duration 11 years). The exposure during the last 5 years did not exceed the maximum allowed doses.

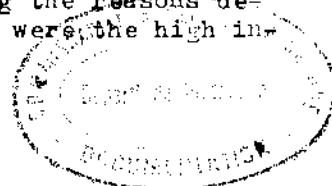
A statistical significant decrease of 17 OH eliminations suggesting a mild adrenal insufficiency was revealed in the subjects exposed as compared with the controls. In 10 subjects whose 17 OH were low, the ACTH Gel test was normal suggesting that this mild adrenal insufficiency is due to an insufficient pituitary secretion. The determination of 17 OH after more than 3 months interruption of exposure in a few subjects showed increased values in all. In subjects in whom the determination of 17 OH elimination was repeated after a lapse of 2 years of continuous occupational exposure, we noticed a decrease of values as compared with the previous levels.

The results suggest that the follow-up of 17 OH elimination in subjects occupationally exposed to ionizing radiations may be an objective criterion both for the appraisal of an asthenic syndrome induced by occupational exposure to ionizing radiations and for the assessment of the opportunity of temporary interruption of exposure.

Introduction

As it is well known, the blood cell count does not represent a satisfactory biological indicator for exposure to ionizing radiations to doses not significantly exceeding the maximum allowed ones.<sup>1,2</sup> Owing to this fact, a great number of biological parameters were assessed in order to detect earlier the alterations due to occupational exposure to ionizing radiations and for a more precise characterization of the radiation disease. We mention among these the increase of the binucleated lymphocytes in the peripheral blood<sup>3</sup>, the chromosomal alterations revealed by cultures of the lymphocytes<sup>4</sup>, the assessment of a number of metabolites eliminated in urine<sup>5,6</sup>, etc.

In order to reveal precocious biological alterations, we have investigated in the last years the hormonal modifications which appear during protracted exposure to ionizing radiations. We are presenting in this study the results of the investigation of the adrenal glands function in long-term occupational exposure to ionizing radiations. Among the reasons determining us to investigate the adrenal glands function were, the high in-



cidence of hypotension<sup>5</sup> as well as of the asthenic syndrome in subjects long-term occupationally exposed to ionizing radiations and the radiation disease too.

#### Material and Methods

We have studied in a number of 115 men, the 24 hr eliminations of 17- $\beta$ -hydroxycorticosteroids (17 OH) in urine by the Porter-Silber's method. In 10 subjects whose levels of 17 OH eliminations were low, the ACTH Gel test was carried out.

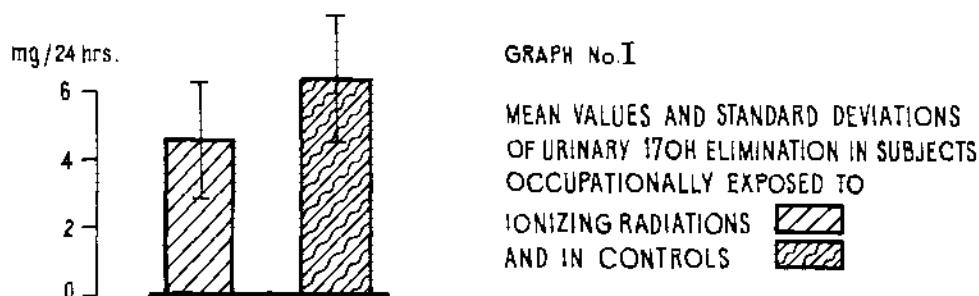
The investigated men were radiologists physicians (40 subjects), technicians exposed to penetrating X or gamma rays during industrial radiographic check-up (45 subjects), dial painters with radioactive paints (10 subjects) as well as uranium miners (20 subjects). The mean age of all men was 40 (20 - 60) years and the mean duration in occupation was 11 (1 - 32) years.

The external doses received by these subjects during the last 5 years did not exceed the maximum allowed ones for occupational exposure. The yearly external doses ranged between less than 480 mrad to maximum 2,900 mrad for the first three subgroups. The uranium miners' external total accumulated doses were comprised between 2 and 10 rads. We have had no information about the total body burden in the dial painters or working level months in the uranium miners either, because we performed no whole body counter for the dial painters and there were no radon daughters' measurements in the uranium mine from which the miners were coming.

Although the doses received during the last years were less than the allowable ones, in some cases, especially in radiologists physicians, the maximum allowable doses were exceeded in the past. As a consequence 6 of the physicians showed peripheral blood cell and bone marrow alterations and another 6 showed mild radiodermatitis of the hands. One of the dial painters was suffering from chronic radiation disease (bone marrow hypoplasia and alterations of the bones' normal structure, especially of the upper and lower limb girdle). The selected men had never had Tb histories including pleuritis, recent febrile or consumptive diseases, liver failure or other causes which could induce adrenal gland insufficiency or disturbances in the metabolism of steroid hormones. At the time of investigation they showed normal peripheral blood cell counts. Twenty-five healthy men, unexposed to radiation, of a similar mean age (39.5 years) were controls.

#### Results

The results of 17 OH eliminations/24 hrs both in exposed subjects and in controls are shown in graph No. I. The results were processed by t Student test. The mean value of 17 OH elimination in exposed subjects was  $4.64 \pm 1.75$  mg/24 hrs. The controls' values were  $6.40 \pm 1.85$  mg/24 hrs.



The difference between these values was highly significant ( $t = 4.545$ ,  $p < 0.001$ ).

Though we did not succeed in finding significant differences between the subgroups of subjects investigated, in relation with the amount of the external doses received, the 17 OH elimination values inside each group were the more decreased the longer the duration in occupation: significant differences were noticed between the physicians having a duration of less than 20 years in occupation ( $5.65 \pm 1.16$  mg/24 hrs) as compared with those having more than 20 years ( $4.84 \pm 1.46$  mg/24 hrs). This difference is significant at a 5 % threshold.

There is a similar situation inside the subgroup of industrial technicians exposed to penetrating X or gamma rays: the number of subjects showing eliminations of 17 OH less than 5 mg/24 hrs is higher in those having over 5 years of occupation as compared with those having less than 5 years.

In 3 subjects in whom the 17 OH determination was repeated after a lapse of two years of continuous occupational exposure, we noticed a decrease of values as compared with their previous levels. In another 3 subjects we repeated the 17 OH urinary determination after a lapse of 3 - 8 months of interruption of exposure to ionizing radiations (table I). In these subjects we noticed an increase of the value of 17 OH eliminated as compared with the levels found before interruption. All these subjects who also complained of asthenia accentuated as the day progressed and dizziness, symptoms suggesting the adrenal gland participation in the picture of the asthenic syndrome, reported they decreased or disappeared after the interruption of exposure.

Table I  
The 17 OH elimination (mg/24 hrs) in three subjects long-term exposed to ionizing radiations after the interruption of exposure

Subject No.	Age (years)	Duration in occupation (years)	Date	17 OH (mg/24 hrs)
1	38	7	Nov. 1971	4.34
			Febr. 1972	4.66
2	45	14	Jan. 1972	4.81
			Mar. 1972	5.08
3	40	9	Apr. 1972	3.44
			Dec. 1972	4.50

In all subjects long-term occupationally exposed to ionizing radiations the percentage of those showing decreased eliminations of 17 OH (less than 5 mg/24 hrs) was 62.2. The safety interval of this percentage ranged between 60 and 78.7 at a 1 % threshold.

In 10 subjects whose eliminations were low (mean value 4.23 mg/24 hrs) we carried out the ACTH Gel test for two days. The individual eliminations of 17 OH after exogeneous stimulation were more than twice higher as compared with their basal levels in each subject, always exceeding 10 mg/24 hrs (mean value 21.02 mg/24 hrs).

#### Discussion

Our results are in agreement with the experimental studies which demonstrated alterations of the adrenals after acute irradiations 9-14.

In subjects long-term exposed to irradiation, the decreased eliminations of 17 OH point to adrenal hyosecretion. This may be a primeval one, due to adrenal lesions induced by ionizing radiations, as the experimental studies revealed, or may be secondary to a hypothalamo-pituitary insufficiency. The stimulation test we carried out indicates that the decreased eliminations of 17 OH may be attributed - in subjects investigated - to an insufficient ACTH secretion. This hypothesis is also indirectly supported by our previous results<sup>15</sup> which showed a decrease of the eliminations of total urinary gonadotrophins in a significant number of subjects long-term exposed to ionizing radiations.

Our results suggest that the follow-up of the 17 OH elimination in subjects occupationally exposed to ionizing radiations may be an objective criterion both for the appraisal of an asthenic syndrome induced by occupational exposure and for the assesement of the opportunity of temporary interruption of exposure.

#### References

1. Williams, E.K. The White Cell Count in Relation to Occupational Radiation Damage. *Acta Radiol.* 41, 1, 21-9 (1954).
2. Chanteur, J., Pellerin, P. Haematological Control Under Normal Conditions, In: Manual on Radiation Haematology. Techn.Rep.Ser.No.123, 369-72, Vienna (1971).
3. Ingram, M., Barnes, S.W. Observation on the Blood of Cyclotron Workers. *Phys. Rev.* 75, 1765 (1969).
4. x<sup>x</sup> Radiation Induced Chromosome Aberrations in Human Cells. In: Report of the United Nations Scientific Committee of the Effects of Atomic Radiation. United Nations suppl.13, annex C (A/7613), N.Y. (1969).
5. Smith, H., Bates, B.T. An Assesement of Those Metabolites Considered to Be of Value in the Diagnosis of Exposure to Radiation. STI/PUB, 199-215, Vienna (1965).
6. Jammet, H.P. Valeur des indicateurs biochimiques. In: Biochemical Indicators of Radiation Injury in Man. I.A.E.A., 233-58, Vienna(1971).
7. Martland, S.H. Occupational Poisoning in Manufacture of Luminous Watch Dials. *J.A.M.A.* 92, 6, 466-473 (1929).
8. Jayle, M.P. Analyse des stéroïdes hormonaux, Vol.1-3, Masson, Paris (1961, 1962).
9. Betz, E.H. Contribution à l'étude du syndrome endocrinien provoqué par l'irradiation totale de l'organisme. Masson, Paris (1956).
10. Yago, N., Omata, S., Kobayashi, S., Ichii, S. Effects of ACTH and Whole Body X-Irradiation on the Concentrations of Enzymes, Nicotinamide, Nucleotides and Cytochromes in Rat Adrenal. *J.Biochem.(Tokyo)* 62/3, 339-344 (1967).
11. Taceva, J., Pospisil, M. Urinary Ketosteroid Excretion as an Index of Adrenal Activity After Whole Body Irradiation of Rats and Correlation with Survival. *Ind.J.radiat.Biol.* 13/3, 289-291 (1967).
12. Ferro, G., Calzavara, F., Bernardi, F. Ricerca comparativa sulle modificazioni dell'eliminazione dei 17 chetosteroidi in ratti maschi castrati chirurgicamente et attinicamente e studio istologico dei testicoli irradiati con dosi scalari. *Quad.Radiol.* 33/2, 139-150 (1968).

13. Mitkiewski, K., Brzezinski, R. Some Histochemical Reactions in the Rat Adrenal Cortex Changed by a Single X-Ray Dose. *Folia Histochem. Cytochem.(Krakow)*, 6/2, 195-202 (1968).
14. Surowiak, J. Quantitative Changes of Acid Phosphatase in the Hypothalamus, Pituitary, Adrenals and Thyroid in Mice (*Mus musculus L*) Exposed to Large, Single Doses of U.V. or X-Rays, Taking into Account the Circadian Rhythm. *Folia Biol.(Krakow)* 7/2, 105-140(1969).
15. Popescu, H.I., Klepsch, J., Lancranjan, I. The Urinary Total Gonadotrophins Eliminations in Subjects Long-Term Occupationally Exposed to Ionizing Radiations. Paper presented to the IX-th Ann. Meet. of the Europ.Soc.for Radiat.Biol., Rome, Sept.26-th - 28-th, (1972).

## PERSONNEL DOSIMETRY INCLUDING TLD

### OCCUPATIONAL EXPOSURE TO X - RAYS IN POLAND - PROBABILISTIC EXTRAPOLATION TO LIFE-TIME DOSES.

J.Jankowski, J.Liniecki, B.Krych  
Institute of Occupational Medicine  
Lodz, Poland

#### Abstract

The individual occupational exposure to X-rays in Poland has been monitored since 1966, embracing ab.15 thousand persons. The percentage of those monitored who did not reveal exposures above 0,4 R per annum has been declining steadily over the last years, starting from 5,2 per cent in 1966 and reaching the value of 1,5 per cent in 1972. Extrapolating from situation over the last 6 years a percentage of the employed was calculated who are likely to exceed 20 or 50 R over a period of 40 years. For the former value this probability varies between 0,7 and 4,0 per cent for total population. Probability to exceed 50 R of life-time exposure for total population varies between 0 and 1,5 per cent.

#### Introduction

Monitoring of X-ray exposed personnel /above 15.000 persons/ is being carried out in Poland since 1966. The annual distribution of individual exposures for the period 1966-1972 is given in table 1! From this table it follows that percentage of persons in the lowest class /  $\leq 0,4$  R/y/ was increasing up to 1969, getting stabilized thereafter at the level of ab.98 per cent.

Similar situation can be noted in regard to yearly numbers of quarterly exposures in excess of 3 and 1,3 R for male and female personnel, respectively, as well as for the annual exposures above 5 R /table 2/.

In most cases these "excessive" exposures resulted from single incidents and not from cumulation of higher values over several

months in a quarter or callendar year. In other words some "steady-state" in regard to magnitude of exposure has been reached.

T a b l e 1

Distribution of exposure values among X-ray workers in Poland from 1966 through 1972.

Year	Percentage of monitored persons falling in following exposure ranges: /R/						
	≤ 0,4	0,4-1,2	1,2-3	3-5	5-12	12-25	> 25
1966	94,80	3,26	1,09	0,17	0,68	-	-
1967	96,52	2,22	0,65	0,25	0,24	0,12	-
1968	97,38	1,93	0,45	0,08	0,08	0,07	0,01
1969	97,63	1,73	0,46	0,08	0,06	0,03	0,01
1970	98,30	1,33	0,27	0,05	0,04	-	0,01
1971	98,50	1,04	0,30	0,08	0,06	0,01	0,01
1972	98,50	1,13	0,20	0,07	0,06	0,04	-

T a b l e 2

Data on quarterly individual exposures above 1,3 and 3 R for female and male workers, respectively, and on yearly exposures above 5 R.

Year	"Excessive" quarterly exposures		"Excessive" yearly exposures	
	Number of persons	in per cent of the total monitored population	Number of persons	in per cent of the total monitored population
1966	156	1,26	85	0,69
1967	95	0,76	45	0,36
1968	59	0,43	25	0,16
1969	52	0,36	15	0,10
1970	30	0,22	9	0,06
1971	33	0,22	11	0,07
1972	34	0,22	16	0,10

However, from all these data it does not follow that by a simple multiplication of exposures over a single year it is possible to obtain a distribution of life-time values. Even if an average percentage of exposures above some pre-set limit is from year to year



constant for the whole surveyed population, the persons monitored do not fall into the same exposure-category every year, and therefore, the probability to exceed for instance an occupational life-time exposures of 20 or 200 R might differ from that for 0,5 or 5 R in any given calendar year.

This study was undertaken at suggestion of the I.C.R.P., with an attempt to arrive at some estimate of a probability to exceed - at conditions currently prevailing - the life-time limits of exposure of 200, 50 and 20 R. Relation of these exposure values to absorbed doses will also be discussed.

#### Methods of analysis

This preliminary analysis was made on results of routine monitoring of X-ray personnel in Poland over the period from 1966 through 1971. That results obtained by means of the film-badge service yield data reflecting real situation were demonstrated independently by one of us<sup>2</sup>.

From 15.388 persons whose names are enrolled in the files only those who have been working over the whole period in question, were submitted to analysis /7010 subjects/. For each person exposure values cumulated after 1, 2, ..., 6 years were extracted from the files. Similar procedure was made for several occupational subgroups /health service personnel in general, radiologists and other physicians, industrial radiographers/. The mean weighted exposure of the whole population and of the subgroups was then calculated.

#### Results and discussion

The distribution of individual exposure values summed up after 1, 2, ..., 6 years is presented in fig. 1. It should be mentioned that median value /0,25 R/ of exposure category 0 - 0,5 R which entered the calculation is - particularly in the first year - a serious overestimate of the real value, because a vast majority of the recorded results were blank readings / < 120 mR/y/. For purposes of simplicity this exposure range 0 - 0,5 R was not further subdivided, but this reservation should be kept in mind when considering the mean weighted cumulative exposure after 1, 2, ..., 6 years as given in table 3.

An average yearly increase of the mean weighted accumulated exposure amounts to ab. 0,08 R, and if this rate of increase were main-

tained, mean accumulated value after 40 years would reach ab.3-4 R. This reasoning remains valid only for accepted assumptions that: 1/ a steady-state in exposure conditions was prevailing over the last 6 years, what cannot be perfectly true, and 2/ that no change in X-ray technology will occur in future.

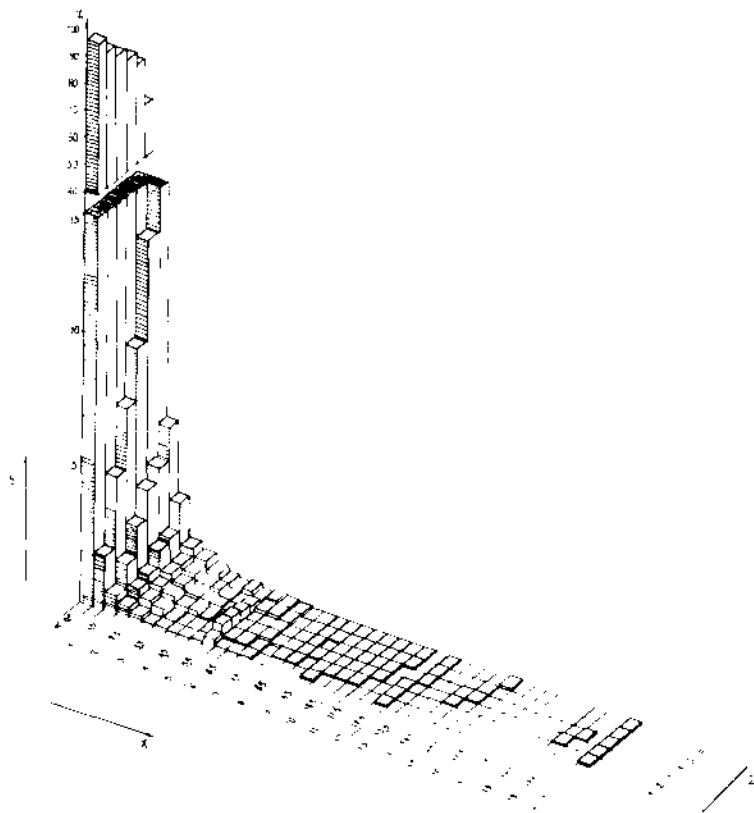


Fig.1

Distribution of individual cumulative exposures after 1,2.....6 years /1966 - 1972/. /Poland,X-ray workers/.

x - axis - ranges of cumulative exposure,

y - axis - percentage of exposed workers followed over the period 1966 - 1971,

z - axis - consecutive years / $n_i = 1,2.....6$ /.

The latter presumption is almost certainly non-realistic, because wide introduction of X-ray machines with image intensifiers may be expected and this should reduce exposure of the personnel considerably. Thus mean accumulated exposure of 3-4 R for people working over forthcoming 34 years may be taken as an upper probable limit of the real value.

T a b l e 3  
 Mean weighted exposure /R/ acquired by the  
 monitored population and some subgroups.

Number of years at exposure /n <sub>i</sub> /	Total	Health service as a whole	Physicians	Industrial radio- graphers
1	0,32	0,32	0,31	0,34
2	0,43	0,40	0,36	0,50
3	0,46	0,47	0,43	0,57
4	0,55	0,53	0,49	0,65
5	0,60	0,58	0,55	0,69
6	0,73	0,72	0,69	0,81
Mean yearly increase of exposure	0,082	0,080	0,076	0,094
Probable mean accumula- ted exposure for the group at the end of 40 year - period	3,52	3,44	3,27	4,01

Furthermore, this value should not be identified with the absorbed dose to the whole body or the red bone-marrow. Our studies<sup>3</sup> as well as those made by other authors<sup>4</sup>, have shown that effective quanta energy of the scattered X-radiation to which personnel of medical X-ray laboratories is exposed /this occupational group dominates the whole/, is close to  $\sim 30$  keV. This is equivalent to reduction of the dose absorbed in bone-marrow against surface dose by a factor of 5-6. In other words, expected upper limit of the 40 years mean bone-marrow dose /dose equivalent/ for the whole group would amount to ab. 0,5 rad /rem/.

Considering now a probability to exceed preset-limits of life-time exposure of 200, 50 and 20 R, numbers of the monitored people were computed who in each of the consecutive six years have exceeded the values of  $n_i \cdot \frac{1}{40}$  of the preset limit, where  $n_i$  denotes the consecutive years starting with 1966. The procedure was started for the life-time limit of 20 R as it provided highest numbers for computations. Results of this calculation are presented in fig. 2.

A constant decrease of the number is clearly apparent. Some deviations from this general rule /chance fluctuations/ were observed in the first 2-3 years for industrial radiographers but the same

trends were prevailing over the last 3-4 years, and thus the group will be treated as a whole.

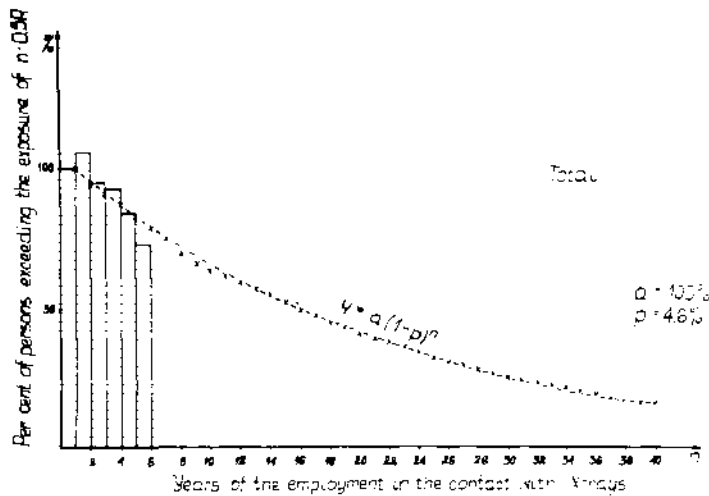


Fig.2

Percentage of 7010 subjects /ordinata/, submitted to the analysis, who exceeded exposure values =  $n_i \cdot 0,5 R$  over the consecutive years / $i = 1, 2, \dots, 6$ //abscissa/.

It was attempted to fit several statistical distributions to these data, however, the results of these trials applying  $\chi^2$  - test were negative. To arrive at some estimate of the probability to exceed the 20 R exposure over 40 years two basic approaches were accepted:

a/ protection against X-rays in the period 1966-1971 represented a steady-state situation which will not change in the future. The percentage who exceeded exposure  $6 \cdot 0,5 R$  resulted only from a stochastic character of repetition of doses above the preset value of  $0,5 R/y$  and for each 6 years period the probability is the same for every monitored person. This assumption would mean that a similar percentage will exceed 20 R exposure over 40 years /4, 12 for the whole group, 3, 27 for physicians, 5, 50 for industrial radiographers /table 4/. In view of the trends in fig.2 this estimate must be treated as an upper probable limit and it may be postulated this is most likely an exaggeration of the real situation,

T a b l e 4

Estimates of the probability to exceed occupational life-time exposure of 20 R.

Occupational group	Number of persons submitted to the analysis	Number and percentage of persons who exceeded 6·0,5 R	Value of p -%/year	Probable % of people who could exceed 20 R life-time exposure as based on extrapolation of the formula $y = a/1-p/n$
Total population	7010	288/4,12/	4,6	0,71
Health service as a whole	6345	255/4,03/	5,4	0,58
Physicians	2355	77/3,27/	5,7	0,34
Industrial radiographers	601	33/5,50/	6,0	0,67

b/ the fraction of population in question, exceeding the exposure =  $n_i \cdot 0,5 R$  will keep declining with increase of the number of years =  $n_i$ . This might occur due to the improved protection against X-rays, however this is not very likely at present technology in view of the data given in table 1 and 2. It might also be due to the fact that average period of repetition by the same persons of the experience of sporadic acquisition of larger doses, resulting in the transfer to the category of accumulated exposure  $>n_i \cdot 0,5 R$ , is much longer than 6 years. In any case, whatever the explanation is accepted it was assumed that decrease in the percentage  $/y_i/$  of persons exceeding the limit of  $n_i \cdot 0,5 R$  will follow the trend prevailing over the period of 1966-1971, according to a geometric progression of the form:

$$y_i = a / 1 - p/n_i \quad /1/$$

where:  $n_i$  - consecutive year of the employment in contact with X-rays,

a - percentage of people exceeding the limit  $n_i \cdot 0,5 R$  in the year when it reached a maximum /usually 1966 or 1967/,

p - average fractional yearly decrease of the percentage of the people exceeding cumulative exposure =  $n_i \cdot 0,5 R$  over the period of observation /from the

maximum value till 1972 /were a was accepted as 100 %/.

The value of  $p$  was calculated from available data /see table 4/ and an average trend for all workers is presented in fig.2. The extrapolated values of  $y_i$  for  $n = 40$  are given in table 4 and they are of the order of few tenths of a per cent. To what extent this extrapolation is sound must remain unknown for the time-being, and the estimate seems to represent a likely lower range of the probable value, because the average period of repetition of sporadic larger exposures may be shorter than 40 years. On the other hand any serious change in the present X-ray technology would be expected to reduce considerably the exposure, and therefore the calculated probability. To summarize, it may be postulated that percentage of X-ray workers, exposed above 20 R over 40 years /starting with 1966/ is likely to range between a fraction of / $\sim 0,7$ / and few / $\sim 4$ / per cent. Similar estimates for likelihood to exceed 50 R at assumption b/ yield negligibly small values, at assumption a/ the amount for all occupational categories to ab. 1,5%, ranging for various subgroups from 0,8 to 2,3%. At both assumptions the probability to exceed 200 R is practically nil.

The relationship between surface exposure and absorbed dose is difficult to assess in persons exposed to amounts of radiation /X-rays/ higher than the average. Were it qualitatively similar scattered radiation, as that encountered normally in medical X-ray laboratories / $E_{\text{eff}} \sim 30$  keV/, than absorbed dose /rads/ in the red bone-marrow would correspond numerically to  $1/6 - 1/5$  of exposure - roentgens measured at the surface<sup>5</sup>. In such an instance 20 and 50 R would correspond to some 3-4 and 8-10 rad, respectively, and estimated likelihood to exceed 20 and 50 R would relate to these bone-marrow doses. On the other hand if exposures from the right hand side of the distribution are due - at least in part - to the action of primary-beam radiation, the average absorbed doses among those who exceed the life-time limit of 20 or 50 Roentgens would be closer to similar values in rads.

#### References

1. Jankowski J., Krych B., Polski Przegląd Radiologii i Medycyny Nuklearnej 36, 637/1972/.
2. Jankowski J., Phys. in Med. Biol. 17, 814/1972/.

3. Jankowski J., Postępy Fizyki Medycznej /to be published/.
4. Maruyama T. and others, Health Physics 21, 563/1971/.
5. Jones A.R., Measurement of the dose absorbed in various organs as a function of the external gamma ray exposure, AECL 2240 Atomic Energy of Canada /1964/.

## PROGRESS IN PERSONNEL NEUTRON DOSIMETRY

Dale E. Hankins  
University of California  
Los Alamos Scientific Laboratory  
Los Alamos, New Mexico 87544

### Abstract

This paper reviews recent progress in personnel neutron dosimetry. A survey of the various albedo-neutron dosimetry systems that have been developed shows the advantages and disadvantages of each of the systems. The energy dependence of the albedo-neutron dosimeters is rather poor, and the effect of this on their accuracy and usefulness is discussed.

Fission-fragment damage to polycarbonates has been studied extensively, with most investigators using the spark-counter technique for evaluation. A survey is made of the systems currently in use, and the problems associated with their use are discussed, including sensitivity, evaluation time, and exposure of the wearer to the fissionable material.

Other systems under study but not yet in practical application are discussed briefly and include thermally stimulated exoelectron emission (TSEE), direct response of thermoluminescent-dosimeter (TLD) materials to fast neutrons, and the response of TLD materials to recoil protons.

### Introduction

The development of dosimeters to determine neutron exposure has not been as successful as the development of gamma-ray dosimeters. For many years, exposures to gamma and x rays were determined from film badges and, more recently, from glass dosimeters and TLD's. With these dosimeters, personnel exposure can be determined with acceptable accuracy, the largest uncertainty being the variation in readings caused by the orientation of the wearer to the source during exposure.

Neutron dosimeter advances have been much slower and less successful. For many years NTA film was the only fast-neutron dosimeter available, but now there are two additional practical dosimetry systems: albedo-neutron dosimeters and fission-fragment track registration. Albedo-neutron dosimeters have recently been placed into service by several organizations, but probably more widespread interest and study have been given to fission-fragment track registration. In this paper I will review and compare these three systems and briefly discuss other techniques which are being studied but which have not yet been adapted to personnel dosimetry.

### Film Badges

The earliest evaluations of personnel doses from neutrons were made with neutron film plates which were inconvenient to read, expensive, and easily broken. NTA film was being used as early as 1947<sup>1</sup> and was significantly more convenient than the film plates.



The major disadvantages of NTA film are the fading of tracks and the energy dependence. The fading problem was recognized early,<sup>2</sup> and has never been entirely solved. To overcome the fading, desiccation and sealing of the film was suggested as early as 1954.<sup>3</sup> Sealing of film has been tried by many investigators, with varying degrees of success. Several organizations that routinely seal NTA films in an envelope made of aluminum, ethylene, and paper report success in eliminating or slowing the fading process.<sup>4-7</sup>

The energy dependence of the nuclear track film was improved by using a complex packaging technique which gave a flatter energy response for neutron energies up to 14 MeV.<sup>3</sup> Unfortunately, this packaging procedure is no longer available commercially, and no improvements in neutron films have been reported since 1954.

The response of NTA film to 2-MeV neutrons is about twice as great as its response to 0.5-MeV neutrons,<sup>4,8</sup> thus limiting the usefulness of NTA film. NTA film has a low-energy cutoff of around 0.5 MeV, but in practice the effective cutoff may be as high as 1.0 MeV, depending on the ability of technicians to discern short tracks. For low-energy neutron spectra, such as the leakage spectra from large reactors, NTA film has proved to be largely unsatisfactory, but for high-energy neutrons from accelerators and neutron sources, such as PuBe and AmBe, the film has been satisfactory.

#### Albedo-Neutron Dosimeters

When the human body is exposed to neutrons, some of the incident neutrons are backscattered to create a flux of neutrons of various energies leaving the body. These neutrons are called albedo neutrons. A dosimeter placed on the body to measure this flux of backscattered neutrons is called an albedo-neutron dosimeter. Such dosimeters are usually designed to detect thermal neutrons. When cadmium is used to eliminate incident thermal neutrons, the dosimeter response is caused mainly by the thermal neutrons backscattered from the body.

The possibility of making a personnel dosimeter based on the detection of backscattered thermal neutrons is not new and dates back to the earliest days of the atomic-energy programs. Several types of thermal-neutron detectors were considered, including film. Studies usually showed that the response was not proportional to the dose for various neutron energies, i.e., the dosimeters were highly energy dependent. Most studies were discontinued at that point.

The introduction of <sup>6</sup>Li TLD's, which proved highly sensitive to thermal neutrons created a new interest in albedo-neutron dosimeters. It is doubtful that this interest was caused by any indication that such a dosimeter would be successful but probably because the dosimeters were easy to make and cost of the dosimeters and associated study was small. Several albedo-neutron dosimetry systems were developed and studied,<sup>9-15</sup> and differing conclusions were reached regarding their accuracy, sensitivity, and usefulness. The differences were caused primarily by variations in dosimeter design. Little consideration was given to what dimensions or material should be used to give the best dosimeter. To provide information necessary for a good albedo-neutron dosimeter design, an extensive study was performed at Los Alamos Scientific Laboratory (LASL).<sup>16,17</sup> The study included many variations in design, including dosimeter size and thickness, and different TLD locations in or on the dosimeter.

The study used dosimeters of 30-mil cadmium, 2 in. in diameter, and having 1/8-, 1/4-, 1/2-, and 3/4-in. thicknesses of polyethylene. Data were obtained at the nine TLD positions shown in Fig. 1. Other dosimeters of 15- and 30-mil cadmium (no polyethylene) with diameters of 3/8, 1/2, 5/8, and 7/8 in. were also used to determine the effect of diameter size and cadmium thickness on the dosimeter's response to neutrons.

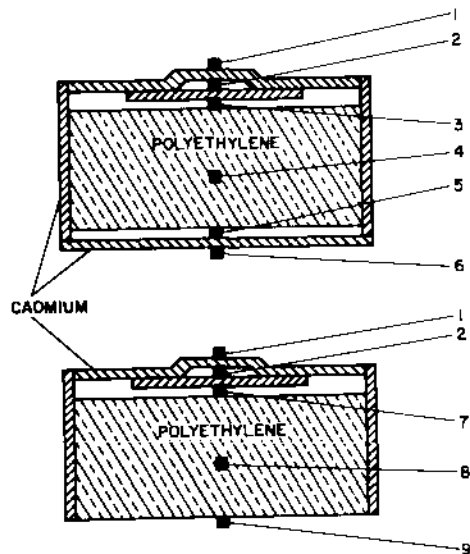


Fig. 1. Schematic of the dosimeters showing the nine TLD positions.

The study showed that albedo-neutron dosimeters are very energy dependent for fast-neutron energies (between 100 keV and 10 MeV). The fast-neutron energy could not be determined from a wide variety of such dosimeters or from a combination of dosimeters, consequently one could not correct for the energy dependence of the dosimeter.

A major problem is the thermal-neutron response of the dosimeter. Thermal-neutron leakage cannot be eliminated from dosimeters of reasonable size, and moving the dosimeter away from the body or wearing it backwards can cause large errors in dosimeter response, even for thermal-neutron dose rates as small as 1 to 2% of the fast-neutron dose (measured in rem).

The effect of the energy dependence of albedo-neutron dosimeters can be seen in Table I. The dosimeter response decreases rapidly as the fast-neutron energy

TABLE I

RELATIVE RESPONSE OF AN ALBEDO-NEUTRON DOSIMETER TO NEUTRON ENERGIES, SOURCES, OR EXPOSURE AREAS

Neutrons	Relative Response
~220 MeV	~0.003
~14 MeV	0.025
PuB	0.077
PuBe	0.080
PuF <sub>4</sub>	0.13
<sup>238</sup> Pu sphere	0.14
PuLi	0.30
<sup>239</sup> Pu metal ~3 kg	0.31
<sup>239</sup> Pu facility	~0.34
<sup>238</sup> Pu facility	~0.44
PuF <sub>4</sub> with 2-in.-polyethylene shielding	0.75
Reactor leakage	1.2

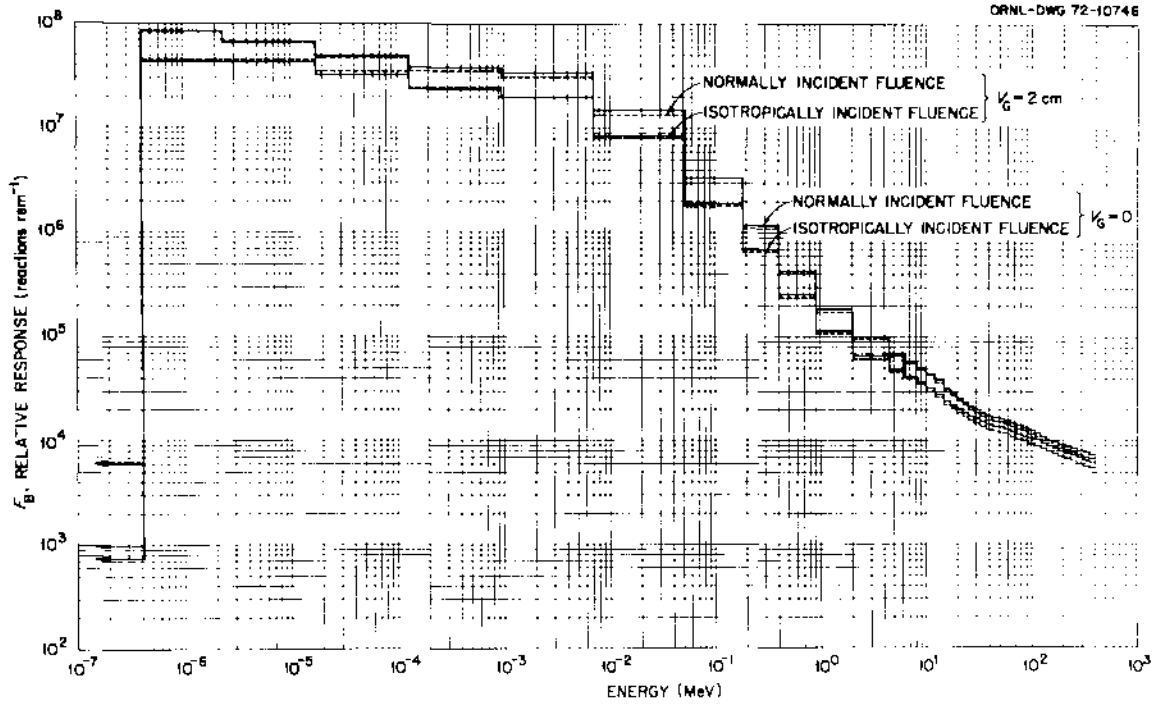


Fig. 2. Calculated response of the albedo-neutron dosimeter.

from several energy neutron sources is increased. These variations in observed response agree well with calculations made by Alsmiller and Barish<sup>18</sup> (Fig. 2).

Several albedo-neutron dosimeters and systems are in use. Figure 3 shows schematics of five dosimeters from the United States and one from England. The schematics have been drawn so that the bottom of the figure represents the side of the dosimeter that would be against the body.

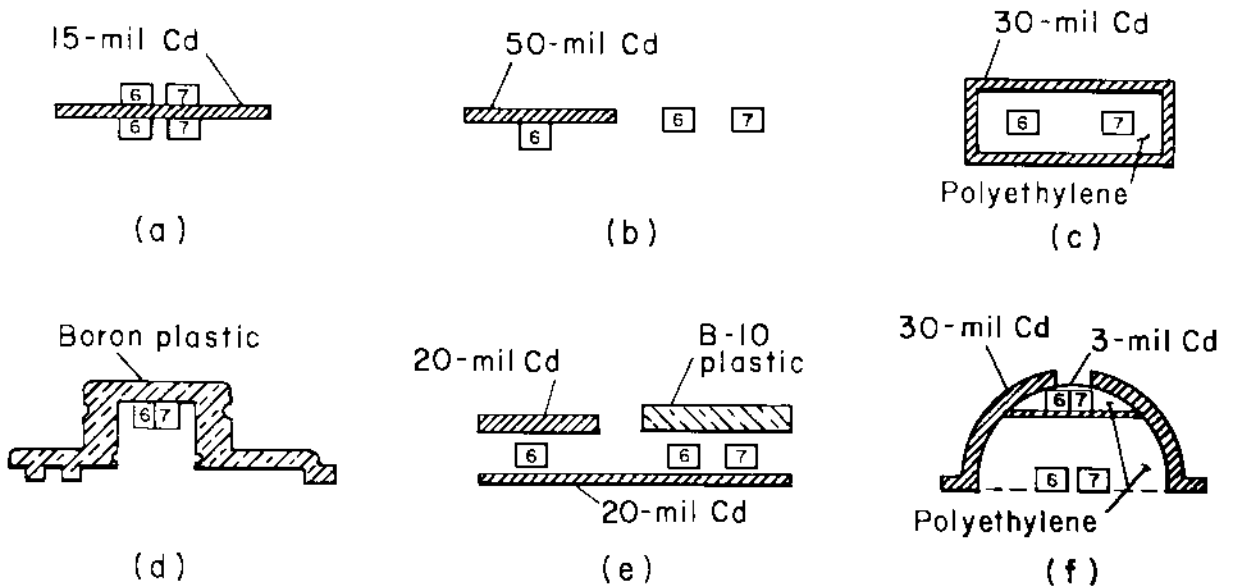


Fig. 3. Schematics of albedo-neutron dosimeters.

The dosimeter schematic in Fig. 3(a) is the simplest design and the dosimetry system most frequently considered. A dosimetry system of this type is in use at Rocky Flats.<sup>9,15</sup> This dosimeter uses 15-mil cadmium (other investigators have used different cadmium thicknesses). The response of the  $^6\text{Li}$  TLD's is corrected for gamma rays by using the  $^7\text{Li}$  TLD's. Because the cadmium is too thin and too small in diameter to eliminate all incident thermal neutrons, the thermal-neutron response of the  $^6\text{Li}$  TLD located above the cadmium is used to correct the response of the lower  $^6\text{Li}$  TLD for thermal-neutron leakage through and around the cadmium. The response of the dosimeter changes with distance from the body, and a large error can occur if it is worn backwards. To avoid this problem, the Rocky Flats dosimeter consists of TLD's mounted on a card which is slipped into a special pocket sewn on the chest area of the coveralls. A  $^7\text{Li}$  TLD located away from the cadmium (to avoid thermal-neutron capture gammas in cadmium) is used to measure the gamma-ray exposure.

To do away with large errors in dosimeter readings,<sup>16</sup> it should be stressed that dosimeters (where the TLD's are not completely surrounded with cadmium or  $^{10}\text{B}$ ) must be held against the body and not be worn backwards.

Most neutron exposure at Rocky Flats is to spontaneous fission energy neutrons and the energy dependence of the dosimeter does not significantly affect the dosimeter accuracy except when the work is located behind neutron shielding. When this occurs, a correction is applied to the reading of the dosimeter. The correction is based on the presence of more thermal neutrons behind the shielding, indicated by a high reading of the  $^6\text{Li}$  TLD located above the cadmium.

The dosimeter<sup>14,15</sup> in Fig. 3(b) is used at the Hanford facilities. The neutron component of the dosimeter consists of only three TLD's mounted in a card containing a total of five TLD's that is slipped into a holder similar to a film badge. The TLD's are read by placing the cards in an automatic reader thus making the dosimetry system practical for a large number of people. The incident thermal-neutron flux is determined by using the  $^6\text{Li}$  and  $^7\text{Li}$  TLD's that are not cadmium-covered. The size of the cadmium over the other  $^6\text{Li}$  TLD is too small to eliminate all incident thermal-neutron leakage, and a correction based on the reading of the bare  $^6\text{Li}$  TLD is applied. The reading of the  $^6\text{Li}$  TLD under the cadmium is corrected for gamma-ray response by using the  $^7\text{Li}$  TLD. All TLD's in the dosimeter have a shield which gives the same gamma-ray absorption as cadmium. However, incident thermal neutrons produce capture gamma rays in the cadmium, thereby increasing the reading of the  $^6\text{Li}$  TLD's located below the cadmium, and introducing a small error.

This dosimeter is worn loosely on the clothing, and thus may be at varying distances from the body. As the distance increases, fast-neutron response decreases and thermal-neutron response increases. An average body-to-dosimeter distance is assumed, and a calibration is performed at that distance. If the dosimeter is worn backwards, a large error in the reported dose can result.

The dosimeter<sup>16,19</sup> in Fig. 3(c) is in experimental use at LASL. It requires only one pair of TLD's completely surround with polyethylene and cadmium. The dosimeter was designed to permit correct dose interpretation even if worn backwards or at varying distances from the body. It is small (1-1/2 by 3/4 by 3/8 in.) and is worn by taping it to the bottom of a film badge. It could be designed into a film-badge size packet with the TLD's mounted on a card, thereby permitting an automated readout technique to make feasible the issuance of dosimeters to a large number of people.

This dosimeter was designed to have a thermal-neutron response equal to the response of the dosimeter to 0.1-MeV neutrons, making a separate measurement of the thermal dose unnecessary. The thermal- and fast-neutron dose determination requires only two TLD's, one  $^6\text{Li}$  and one  $^7\text{Li}$ . The sensitivity of this dosimeter

is lower than that of a dosimeter designed with cadmium or boron on one side only.

To measure the beta and gamma components of the dose, there must be additional TLD's separate from the dosimeter. If desired, the thermal-neutron dose could be determined by using an additional  $^6\text{Li}$  TLD. The thermal-neutron dose could be used to give information about shielding conditions in a way similar to the technique used at Rocky Flats.

The boron-loaded plastic dosimeter shown in Fig. 3(d) was developed in England.<sup>21,22</sup> It has a thermal-neutron response equal to the dosimeter's response to reactor-leakage intermediate-energy neutrons. If used for fast neutrons, the dosimeter's response is much lower than the thermal-neutron response, and a separate measurement of the thermal fluence would be required to correct for the thermal-neutron overresponse of the dosimeter. This dosimeter was designed for use at reactors, and the developers feel it is useful only for reactors.<sup>23</sup> The shape of the dosimeter reduces its response to incident thermal neutrons because the TLD's are "hidden" in a recess. The dosimeter's response to thermal neutrons also varies as a function of dosimeter distance from the body, and it is assumed that an average distance applies. It is attached to the clothing by a safety pin snapped to the dosimeter. Additional TLD's to measure incident beta and gamma radiations can be placed in a cup located on top of the dosimeter.

The dosimeter<sup>15,24</sup> shown in Fig. 3(e) was developed at Lawrence Livermore Laboratory (LLL). The dosimeter uses two  $^6\text{Li}$  TLD's, one under a shield of  $^{10}\text{B}$  and the other under cadmium. Because the cross sections of these materials vary at low-neutron energies, one can determine theoretically whether the dosimeter has been exposed to high- or intermediate-energy neutrons. However, in practice, the difference between the two  $^6\text{Li}$  readings is small for even the lowest energy neutrons, and the results obtained are not always meaningful. Also the gamma reading of the  $^7\text{Li}$  TLD's under the  $^{10}\text{B}$ , used to correct the  $^6\text{Li}$ -TLD readings for their gamma-ray exposure, is not always the same as it would be if the TLD's were placed under the cadmium. This is caused by the shielding of low-energy gammas (<100 keV) by the cadmium, which reduces the TLD reading, and the capture of thermal neutrons in cadmium, which increases the TLD reading under the cadmium.

The back of this dosimeter is cadmium-covered and could be worn backwards without causing a large error in its response. It also responds acceptably when worn at varying distances from the body. Readout of the dosimeter is automated. The dosimeter is a plastic disk  $\sim 1\text{-}3/8$  in. in diameter, similar to the LLL gamma TLD dosimeter<sup>25</sup> which can be used to determine the incident thermal-neutron and gamma-ray dose.

The dosimeter shown in Fig. 3(f) is the largest dosimeter shown -  $\sim 2$  in. in diameter.<sup>15,27</sup> Because of its weight, it is worn on a belt and is not likely to be worn backwards or at varying distances from the body.

The incident thermal-neutron dose is determined using the TLD's at the top of the dosimeter, and a correction for the incident thermal-neutron leakage around the dosimeter is made to the response of the TLD's next to the body. Of the dosimeters shown in Fig. 3, this dosimeter has the highest sensitivity because a substantial amount of polyethylene is used. It is commercially available, without TLD's or belt, for about \$16.00.

This Savannah River Plant dosimeter has the same fast-neutron energy dependence as other albedo-neutron dosimeters but its response to intermediate-energy neutrons is slightly lower because of its large size and polyethylene content.<sup>18</sup> Its decreased response may give it a slight advantage for reactor-leakage spectra.

There are several sources of error in albedo-neutron dosimeter readings which one should consider when selecting a dosimeter.

1. The "wearing" error, i.e., whether the dosimeter is being worn backwards or at varying distances from the body.

2. The error caused by the  $^7\text{Li}$  TLD's not being in the same location as the  $^6\text{Li}$  TLD's, i.e., not under cadmium or  $^{10}\text{B}$ .

3. The error in the correction applied for incident thermal-neutron leakage through or around the cadmium or  $^{10}\text{B}$ .

4. The error caused by uncertainties in each of several TLD readings.

5. The error caused by energy dependence. The neutron-energy dependence of albedo-neutron dosimeters is so large that these dosimeters are acceptable only if the neutron energy is known.<sup>12,16,19</sup> For example, if both PuBe- and PuLi-neutron sources are handled, readings from any of the dosimeters in Fig. 3 could not be correctly interpreted, since the response of the the dosimeter to PuLi neutrons is higher by a factor of  $\approx 3.8$  than the response to PuBe neutrons. For another example, the response to neutrons from a PuF<sub>4</sub>-neutron source is only one-third that of the response to neutrons from PuF<sub>4</sub> in the process line where PuF<sub>4</sub> is converted to plutonium metal.

The severe energy dependence of albedo-neutron dosimeters requires field calibrations which may be done by either of two techniques.<sup>19</sup> The first is by taping the dosimeter onto a suitable moderator such as a cylindrical 1-gal polyethylene jug filled with water. The front of the jug is positioned at a point where the neutron dose rate has been measured. The readings from the TLD's and the total dose are then used to determine the calibration factor. The second calibration procedure uses a 3-in.- and a 9-in.-sphere neutron instrument. The ratio of the count rates from these spheres is directly related to the sensitivity of the albedo-neutron dosimeter.<sup>19</sup> Although this technique was devised for use with the LASL dosimeter, it can be adapted to other dosimeters. With this technique, one can make a rapid survey of an area to determine if albedo-neutron dosimeters can be used and, if so, one can determine the appropriate calibration factor.

#### Fission-Fragment Track Registration

The fission-fragment damage track technique for neutron dosimetry is an interesting new approach to dosimetry. Its general interest is demonstrated by a survey conducted by Griffith.<sup>27</sup> In response to a questionnaire, he received replies from 118 laboratory groups in 20 countries indicating that they had made studies of track registration. Of these, 46 indicated an interest in neutron dosimetry. In spite of this widespread interest, relatively few organizations are using track registration for routine neutron dosimetry of personnel.

When an energetic charged particle passes through a good insulating solid substance, such as mica or a polycarbonate, electrons are scattered away, leaving a damaged region. If the material is subsequently etched, a cylindrical region along this track is preferentially etched and develops into a pit or track which can be seen through a microscope. The energetic particles can be any of several charged particles but, for personnel dosimetry, only fission fragments have proved useful.

To avoid the tedious procedure of counting tracks with a microscope and to improve sensitivity, a spark-counting technique was developed. A thin piece of polycarbonate foil is placed in contact with a fissile material. When the fissile material is exposed to neutrons of the proper energy, fission occurs. Many of the fission fragments have adequate energy to penetrate this foil. When the foil is etched, holes appear in the foil where the fission fragments penetrated. The foil is placed between two electrodes, one of metal and the other consisting

of Mylar foil onto which a thin layer of aluminum has been evaporated. When a high voltage is applied, a spark jumps between the electrodes through one of the holes in the polycarbonate foil. Heat from the spark evaporates the aluminum layer from the Mylar in a small area around the hole. This effectively removes one electrode, and no additional sparks will occur through that hole. Sparking occurs at other holes, and this process continues until a spark has occurred at each hole. The sparking rate is dependent on the capacitance built into the high-voltage line. By connecting the circuit to a scaler, the number of sparks that occurred can be determined. This is related to the number of fissions that occurred and to the flux of neutrons impinging on the fissionable material.

For a given flux of neutrons, the number of fissions that occur is proportional to the mass and the fission cross section of the fissile material. Figure 4 shows the cross sections of three fissile materials that have been considered for use as radiators.  $^{238}\text{U}$  is useful for short-term exposures, but for personnel neutron dosimeters it is generally considered unacceptable because of its high spontaneous fission rate. This causes a high number of background tracks over a period of a month and is therefore not useful for low exposure over long time periods.<sup>28,29</sup>  $^{235}\text{U}$ , not shown on Fig. 4, has also been considered for use. It has a high thermal- and intermediate-energy cross section and, although it has little value for fast-neutron dosimetry, it is being used as a monitor for intermediate-energy reactor neutrons.<sup>30</sup>

$^{232}\text{Th}$  and  $^{237}\text{Np}$  are the two fissile materials receiving the most consideration for personnel neutron dosimeters. Referring to Fig. 4, one apparent advantage of  $^{237}\text{Np}$  is that its cross section extends to lower neutron energies with an effective threshold energy of about 0.6 MeV, compared to 1.5 MeV for thorium. Another advantage is that the  $^{237}\text{Np}$  fission cross section is about a factor of 10 higher than the thorium cross section. Unfortunately, neptunium has a higher specific activity and creates a significantly higher external hazard from gamma-ray exposure to the wearer.<sup>28</sup> However, the energy of the neptunium gamma rays is lower than many of the gamma rays in the thorium chain. It is argued that neptunium gammas can be more easily shielded,<sup>29</sup> thereby reducing personnel exposure (but increasing the weight of the dosimeter). There is also the possibility that dosimeters containing fissionable material may be taken home, lost, stolen, or in other ways find their way to the public. (The possibility of a child cutting teeth on one of the badges is often mentioned.) Fixing the fissionable material into the badge in a nonremovable manner is one possible solution. Consideration has been

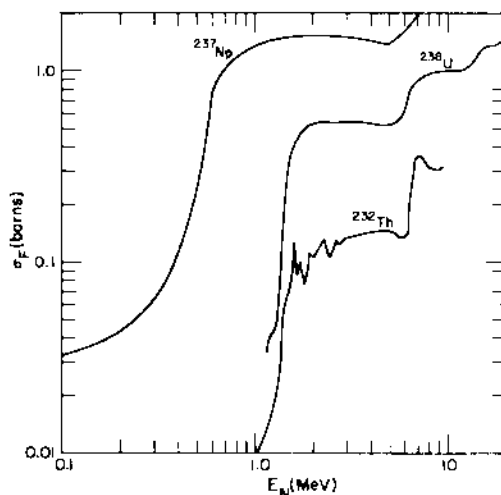


Fig. 4. Fission cross sections of  $^{237}\text{Np}$ ,  $^{238}\text{U}$ , and  $^{232}\text{Th}$  as a function of neutron energy.

given to a neptunium alloy of aluminum,<sup>15</sup> a sealing coat of gold,<sup>31</sup> or polycarbonate foil.<sup>29</sup> Unfortunately, sealing or packaging reduces the sensitivity of the dosimeter by reducing the number of fission fragments that may cause tracks.

Gamma exposure rates from fissile material in a dosimeter are kept low by using as little of the fissile material as possible (2 to 5 mg/cm<sup>2</sup>) and spreading it uniformly over an area of about 2 to 5 cm<sup>2</sup>. Since fission fragments are easily absorbed in the fissile material, the use of thicker foils does not appreciably increase the number of tracks. The sensitivity of a dosimeter using <sup>237</sup>Np as the fissile material is about six sparks for a fast-neutron dose of 10 mrem,<sup>29</sup> and for thorium reported values vary from 0.8<sup>29</sup> to ~3.0<sup>28</sup> sparks per 10 mrem. The time required to count the tracks in a mica or polycarbonate foil using a microscope is about the same as that needed to evaluate an NTA film. With some spark-counter techniques, the time required to evaluate a foil may be reduced, but with others the time may be greatly increased.

At present, only three U.S. laboratories are known to be routinely using fission-track dosimeters on personnel. Argonne National Laboratory is using a <sup>235</sup>U system with mica foils.<sup>30</sup> Brookhaven National Laboratory is using thorium and a polycarbonate foil, with spark counting for evaluation.<sup>28</sup> Oak Ridge National Laboratory (ORNL) is using a finger ring of thorium and polycarbonate foil, with spark counting for evaluation.<sup>29</sup>

Probably the most widely used fission-fragment dosimetry system is that of Prêtre<sup>32,33</sup> in Switzerland. It has been recently modified to use two fissile materials, one of thorium and the other of either <sup>235</sup>U or <sup>209</sup>Pu, depending on the application.<sup>34</sup> A thorium dosimetry system is in use at Julich, Germany,<sup>35</sup> a thorium finger system is used at Karlsruhe, Germany,<sup>36</sup> and a thorium dosimeter for the hands is in use at Geel, Belgium.<sup>37</sup> Other dosimetry systems are being studied and were reported at the IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.

#### Selecting a Dosimetry System

Three dosimetry systems are presently being used for fast-neutron personnel dosimetry: NTA film, albedo-neutron dosimeters, and fission-fragment track dosimeters. Each dosimetry system has its advantages and disadvantages. No one system is the ultimate dosimetry system for all applications.

In the selection of a dosimetry system, consideration must be given to the energy dependence of the dosimeters over the energy region encountered at a facility. For example, the three dosimetry systems discussed here have an energy dependence of about a factor of 10 for neutron energies between 0.5 and 14 MeV. Over this energy region, fission fragment dosimeters using <sup>237</sup>Np and NTA film have responses that increase, whereas the response of albedo-neutron dosimeters decreases. In addition to the energy dependence, the lower energy threshold must be considered. For example, fission-fragment damage dosimeters using <sup>237</sup>Np would be useful for neutrons of 1 MeV energy, but if <sup>232</sup>Th is used, the dosimeter would have no reading. Only albedo-neutron dosimeters and fission-fragment dosimeters using <sup>235</sup>U will respond to neutrons having energies of less than ~0.5 MeV, and for facilities that have these lower energy neutrons these are the only dosimeters that should be used. Of the three dosimetry systems discussed, the albedo-neutron dosimeter is the only one that respond to thermal-, intermediate-, and fast-neutron energies, but its energy dependence may make it difficult to interpret the response into dose.

Selection of a dosimetry system may depend on the location of the facility. In humid climates, NTA film fades rapidly, and if the films are not sealed, results can be of questionable value. In dry climates, where fading is minimal or when the film is sealed, NTA film can be used. Albedo-neutron and fission-fragment track dosimeters do not fade.



The time necessary to evaluate an exposed dosimeter may be an important consideration in selecting a dosimetry system, particularly if large numbers of dosimeters are issued. The time to process and read NTA film is similar to that required for etching and microscopic evaluation of fission-fragment damage dosimeters. Evaluating the foils by using the spark counter can reduce the time slightly if a simple (but less accurate) sparking procedure is adopted, but the complex procedures would require more time. The reading of TLD's in an albedo-neutron dosimeter requires varying amounts of time, depending on the number of TLD's in the dosimeter and whether the TLD's are individually handled or have an automatic readout capability.

Another consideration is the ability of the dosimeter to discriminate against gamma rays or to evaluate neutron exposure in the presence of gamma rays. Only the fission-fragment dosimeter can be used to measure neutron doses in the presence of significantly high gamma-ray doses. The ability of albedo-neutron dosimeters to discriminate against gamma rays varies, depending on the design of the dosimeter,<sup>16</sup> but is generally considered adequate for gamma-to-neutron ratios of 3-to-1. With NTA film, the associated gamma rays for most practical applications, i.e., doses less than 1 R, do not cause a problem in evaluating the film.

Other items to be considered include the realization that the TLD reading could be lost by a defective reader, the fission-fragment polycarbonate foil could be ruined by overetching or tearing of the foil, and the NTA film could be defective, lost, or ruined in the photographic processing. The reading of the TLD's in albedo-neutron dosimeters cannot be repeated, whereas the NTA film and spark-counter foils can be reread if necessary.

Fission-fragment track dosimeters require that a person carry a fissionable material which may expose him to radiation, and could pose a problem if lost. This has led many organizations to consider these dosimeters unacceptable.

The acceptance of the wearer should be given consideration. Some persons object to wearing an albedo-neutron dosimeter on a belt. Dosimeters placed in pockets sewn on coveralls are not practical in areas where coveralls are not required for other reasons. Generally, a film-badge size dosimeter is considered acceptable, but the clamp, pin, snap, etc., to attach it to the clothing has never been universally accepted by the wearer.

Several comparisons of these dosimetry systems have been made. One comparison of albedo-neutron dosimeters and film indicated that greater accuracy could be obtained with film,<sup>38</sup> and other studies indicate the albedo-neutron dosimeter is preferred.<sup>14,15,31</sup> One organization has considered using a combination albedo-neutron and fission-fragment track dosimeter,<sup>28</sup> another group is studying albedo-neutron dosimeters and NTA film.<sup>39</sup> These combinations are being used because they give two evaluations of the dose which, in combination, cover the intermediate- and high-energy regions.

#### Studies of Other Dosimetry Techniques

Other personnel neutron-dosimetry techniques are being studied. Most have inadequate sensitivity at the present time, but efforts are being made to increase their sensitivity.

One of the earlier studies investigated various TLD materials<sup>14</sup> to see if the fast-neutron response could be enhanced. TLD materials were prepared by The Harshaw Chemical Company and contained materials such as uranium, manganese, sodium, dysprosium, gadolinium, and other related compounds. They were distributed to investigators who found that the addition of these materials usually decreased total TLD sensitivity and, with the exception of the uranium loading, failed to increase the fast-neutron sensitivity. Uranium-loaded TLD materials showed a

slight increase in sensitivity, probably caused by the capture of thermal neutrons produced by the degrading of fast neutrons in the TLD materials and surroundings. To the author's knowledge, this work has been discontinued.

Thermally stimulated exoelectron emission (TSEE) has been considered as a possible personnel neutron dosimeter. Many of the reproducibility problems reported in earlier work have been solved.<sup>14,15,29,31</sup> Unfortunately, the dosimeters require ~4 rad of fast-neutron dose to give the same reading as 1 R of gamma rays.<sup>15</sup> When the neutron dose is expressed in rem, ~30 to 40 rem of neutron dose is required to give the same reading as 1 R of gamma rays. For the dosimetry system to be useful, the gamma component of the dose must be small compared to the neutron component. These dosimeters are useful around some critical assemblies, but for routine personnel neutron exposure, the gamma-ray response of TSEE materials is so large that the neutron response cannot be determined. Development work on TSEE is still continuing.<sup>31</sup>

Mixing of TLD materials with liquid or solid organic recoil proton radiators to increase the response of the TLD materials to neutrons has been reported.<sup>31</sup> In most cases, liquid or solid organic proton radiators have been used and this technique requires the separation of the constituents prior to evaluation. A recent study at ORNL<sup>31</sup> has placed TLD material in a homogeneous matrix which can be heated repeatedly without decomposing. The response to mixed radiation fields from ORNL's Health Physics Research Reactor is three times higher than for Teflon-embedded phosphors having the same gamma radiation sensitivity, the enhanced response being due to the fission neutrons. Although there is an enhanced response, the ratio of neutron-to-gamma sensitivity is still too small to be useful for most applications. This work is continuing.

Thick-film emulsions<sup>40</sup> are of interest to high-energy accelerator health-physics personnel. The response of these films to high-energy neutrons may be useful in evaluation of personnel exposure, and there is a proposal for additional research.

Track registration of recoil protons and other nuclei, mainly carbon and some alpha tracks from (n, $\alpha$ ) reactions at higher neutron energies, has been reported.<sup>27</sup> Track registration of protons or nuclei would avoid the disadvantage of having the person carry a fissionable material. The disadvantage is that the recoil tracks are small, more difficult to count visually than fission-fragment tracks, and do not create holes in the thin foil to make spark counting possible. A method proposed by Tommasino<sup>41</sup> for amplification of the tracks has been used with reasonable success.<sup>29</sup> However, because of the time, procedures, and equipment required, it is questionable whether such a system would be useful for routine application. If necessary, this technique can be used to reevaluate an exposed NTA film, after the photographic emulsion has been removed, to determine the neutron dose with a relatively high sensitivity.<sup>29</sup>

#### Discussion and Summary

Many of the studies in this paper have been discussed at the four meetings of the USAEC Workshop on Personnel Neutron Dosimetry.<sup>14,15,31,42</sup> This group consists of people who are actively working on personnel neutron dosimetry development and application. The exchange of ideas, developments, and methods has eliminated much of the duplication of effort that existed before these meetings and has been of value in guiding research efforts of the participants. These meetings have provided an opportunity to discuss openly some of the problems with various dosimetry systems, for example, the hazards of fissile material in dosimeters, the energy dependence of albedo-neutron dosimeters, and the special problems associated with high-energy accelerator health physics. The meetings also serve to keep the participants informed of recent developments in personnel neutron dosimetry.

There are now three dosimetry systems available for consideration by users: NTA film, albedo-neutron dosimeters, and fission-fragment track dosimeters. None of these systems is the ultimate dosimeter. Each has been extensively studied and its limitations and advantages are known. Selection of a dosimetry system will depend on a number of factors, and a dosimeter useful at one facility may not be considered useful at another.

Interesting dosimetry techniques are being studied and may have future application for personnel neutron dosimetry, but at present most lack sensitivity or the ability to separate adequately the gamma and neutron components.

#### References

- [1] J. S. CHEKA, Phys. Rev. 71, 836 (1947).
- [2] J. S. CHEKA, Phys. Rev. 74, 127 (1948).
- [3] J. S. CHEKA, Nucleonics, Vol. 12, 6, 40 (1954).
- [4] P. N. KRISHNAMOORTHY, G. VENKATAKAMAN, D. SINGE, and DAYASHANKAR, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.
- [5] J. JASIAK and T. MUSEALOWICS, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.
- [6] H. J. DUNSTER, National Radiological Protection Board, England, personal communication (1972).
- [7] W. T. THORNTON, D. M. DAVIS, E. D. GUPTON, Oak Ridge National Laboratory report ORNL-3126 (1961).
- [8] C. H. DISTENFELD and J. R. KLEMISH, JR., Brookhaven National Laboratory report BNL-17452 (1972).
- [9] R. B. FALK, Dow Chemical Company report RFP-1581 (1971).
- [10] H. E. PRESTON, United Kingdom Atomic Energy Authority Reactor Group report AEEW-M801 (1968).
- [11] J. B. C. BROWN, F. M. DAYTON, J. A. HALL, J. R. HARVEY, and A. M. WEBB, Berkeley Nuclear Laboratories Research and Development Department report RD/B/R828 (1967).
- [12] D. E. HANKINS, Los Alamos Scientific Laboratory report LA-4341 (1970).
- [13] A. KORBA and J. E. HOY, Health Phys. 18, 581 (1970).
- [14] C. N. UNRUH, Ed., Battelle Northwest Laboratory report BNWL-SA-2978 (1969).
- [15] C. N. UNRUH, Ed., Battelle Northwest Laboratory report BNWL-1616 (1971).
- [16] D. E. HANKINS, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.
- [17] D. E. HANKINS, Los Alamos Scientific Laboratory report LA-4832 (1972).
- [18] R. G. ALSMILLER, JR., and J. BARISH, Oak Ridge National Laboratory report ORNL-TM-3984 (1972).
- [19] D. E. HANKINS, Los Alamos Scientific Laboratory report LA-5261 (1973).
- [20] G. W. R. ENDRES, Battelle Northwest Laboratory report BNWL-SA-3955 (1971).
- [21] J. R. HARVEY, W. H. R. HUDD, and S. TOWNSEND, Berkeley Nuclear Laboratories Research and Development Department report RD/3/N1547 (1969).
- [22] J. R. HARVEY, W. H. R. HUDD, and S. TOWNSEND, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.
- [23] J. R. HARVEY, Berkeley Nuclear Laboratories Research and Development Department, personal communication (1972).
- [24] R. V. GRIFFITH, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.
- [25] D. E. JONES, K. F. PETROCK, B. G. SAMARDZICH, and E. G. SHAPIRO, Lawrence Livermore Laboratory report UCRL-73084 (1971).
- [26] J. E. HOY, Savannah River Laboratory report DP-1277 (1972).
- [27] R. V. GRIFFITH, Lawrence Livermore Laboratory report UCRL-51362 (1973).
- [28] C. H. DISTENFELD and J. R. KLEMISH, JR., Brookhaven National Laboratory report BNL-17452 (1972).
- [29] K. BECKER, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.

- [30] T. J. YULE, R. J. ARMANI, and R. GOLD, Argonne National Laboratory report ZPR-TM-123 (1972).
- [31] C. N. UNRUH, Ed., Battelle Northwest Laboratory report BNWL-1777 (1973).
- [32] S. PRÉTRE and K. HEUSI, Eidg. Institut für Reaktorforschung, Würenlingen, Switzerland report TM-SU-149 (1972).
- [33] S. PRÉTRE, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.
- [34] S. PRÉTRE, Eidg. Institut für Reaktorforschung, Würenlingen, Switzerland, personal communication (1973).
- [35] M. HEINZELMANN and H. SCHÜREN, Kernforschungsanlage, Jülich, Germany (no date).
- [36] K. BUIJS, J. P. VAANE, B. BURGKIARDT, and E. PIESCH, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.
- [37] B. J. MIJNHEER, Bureau Central de Mesures Nucleaires, Geel, Belgium, personal communication (1973).
- [38] A. KNIGHT, T. O. MARSHALL, C. L. HARVEY, and J. HARVEY, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.
- [39] E. D. GUPTON, Oak Ridge National Laboratory, personal communication (1973).
- [40] Health Physics Group, Lawrence Berkeley Laboratory internal report 160 (1973).
- [41] L. TOMMASINO, Proceedings of Seventh International Colloquium on Corpuscular Photography and Solid-State Detectors, Barcelona, 1970.
- [42] D. E. HANKINS and E. J. VALLARIO, IAEA Symposium on Neutron Monitoring for Radiation Protection Purposes, Vienna, 1972.

## IONIZING RADIATION RECORDS FOR INDIVIDUALS

C. Sharp Cook  
Department of Physics  
University of Texas at El Paso  
El Paso, Texas 79968

### Abstract

Even though many people have expressed alarm over possible effects of the ever-increasing amounts of ionizing radiations in our environment, no records exist from which the cumulative dose for any individual adult in the United States can be tabulated. Rather than stop the construction of new nuclear power stations and limit the use of other radiation sources, such as for medical purposes, simply because we are not sure of the effects, it is time that we determine the nature and magnitude of the effects in such a way that they can be correlated with accurate measurements of radiation dose. Suggestions are made as to how a record-keeping system can be established, without undue involvement of the general populace and with a minimum of monetary cost.

### Ionizing Radiation and Public Concern

During the 20th century the magnitude of our ionizing radiation environment has increased slowly but steadily. The primary factors contributing to this increase have been commercial use of radionuclides, use of nuclear fission for nuclear weapons and for nuclear power, and an increased use of medical and dental radiology<sup>1</sup>.

Gross effects of ionizing radiations are well known and, based on these gross effects, standards have been developed limiting the dose an individual should receive<sup>2</sup>. However, some people have concluded that adverse effects may also ensue from very small doses<sup>3</sup>. For example, Stewart has found<sup>4</sup> that relatively small doses delivered to the fetus may induce an excess cancer risk during the first ten years of life. Gauden<sup>5</sup> has pointed out that chromosomal changes, especially in the fetus, may be induced by small amounts of ionizing radiation. A positive correlation has been found by Mole<sup>6</sup> between incidence of cancer in uranium miners and relatively small doses of ionizing radiation. These specific studies cover a very limited portion of the general population. Other studies, which appear to be based on insufficient data, have created controversy over the relevance of their findings<sup>7,8</sup>. Determination of the cause of selected biological effects and whether ionizing radiations have a role in the production of these effects is difficult because of the large number of variables that must be considered.

As long as controversy exists, public concern is justified. This concern has appeared for example as opposition to nuclear power plants, a sometimes emotional issue that leads to extensive hearings, delays and conflict, such as those associated with the application of the Long Island Lighting Company to build a nuclear power station at Shoreham, New York<sup>9</sup>. Concern over fallout radiation and its effects was a primary factor that led to a moratorium on

atmospheric testing of nuclear weapons, no less than a concern about the tremendous destructive power or the havoc these weapons would cause if used in a combat situation.

#### Record Keeping As It Exists Today

Despite public concern, no adult in the United States really knows the magnitude of the total dose he has accumulated from exposure to ionizing radiation. Part of the reason for this lack of data is the complexity of the situation. However, another relevant factor is an apathy toward proper record keeping and sometimes outright opposition. Many reasons are cited for this inaction but the end result is always an unknown factor that allows scientists to argue indefinitely over the effects of ionizing radiations. The same or similar problems exist in other parts of the world. However, since the information I am using comes primarily from U.S. sources, I shall limit my suggestions to those applicable to the United States. I believe many other parts of the world can use them with little or no modification.

Some ionizing radiation exposures are actually excluded from record-keeping requirements. For example, although regulations of the USAEC mandate record keeping for all persons who receive, possess, use or transfer by-product material, source material or special nuclear material and establish standards for protection against radiation hazards arising from activities under licenses issued by the USAEC<sup>10</sup>, these same regulations specifically state that nothing in them "...shall be interpreted as limiting the intentional exposure of patients to radiation for the purpose of medical diagnostics or medical therapy." Although it does not really so state, both the medical profession and record keepers have generally interpreted the quoted statement as meaning that permanent record keeping of ionizing radiation doses received for medical purposes is not necessary. Without this information, complete and permanent records of individual doses is impossible.

Many people apparently do not realize the magnitude of medically produced radiations, largely because of the lack of individual records. However, studies by the Atomic Bomb Casualty Commission<sup>11</sup> of people exposed to radiations from the nuclear detonations at Hiroshima and Nagasaki show that they were further exposed between 1946 and 1963 to a cumulative bone-marrow integral dose from medical X-rays comparable to or greater than the dose they received because of radiation from the respective nuclear detonations. These people are among the few and may be the only group of people in the world for whom accurate records of all radiation dose has been kept over a realistically long period of time.

#### Suggested Procedures

On numerous occasions suggestions have been made that records be kept of all individual radiation doses. Eason and Brooks recently reviewed<sup>12</sup> the history of some suggestions to maintain permanent records of cumulative radiation dose to individuals. They also discussed selected court and review board cases in which compensation was awarded for supposed exposure to ionizing radiation, even though there was no way of knowing the true radiation history of the concerned individuals. Cameron<sup>13</sup> has even proposed a unit for measurement of medical radiation exposure.

One reason often cited for not requiring permanent record keeping of medical radiations is the psychological effects on the individual if he knows that records are being kept. Another argument has been that the general public will not wear film badges or other radiation measuring devices. In my opinion both arguments evade the basic issues. The wearing of film badges or other personal dosimeters is not required, except for those working in an industry subject to USAEC regulation<sup>10</sup>, since almost all exposures of significant magnitude can be

recorded from other available information, maybe even more reliably than from personal dosimetry readings. After discussions with many individuals who can be classified as part of the general public, I have come to the conclusion that record keeping of ionizing radiation doses might produce a greater psychological effect on members of the medical profession than on the general public.

If records are to be kept, a system must be established that allows reasonably good controls and a minimum of bookkeeping and interference with the lifestyle of the individuals involved. To accomplish these goals, ionizing radiations and the methods of measurement need to be divided into three categories, general-area radiations, industrial radiations and medical radiations (including dental).

Background radiations form one type of general-area radiations. Although the magnitudes of these radiations vary from locality to locality, depending both on altitude and the amount of natural radioactivity in the vicinity, they should not fluctuate in any one area, except for effects caused by solar storms on cosmic-ray intensities<sup>14,15</sup>. Other general-area radiation sources are the airborne radioactivity that has been produced by atmospheric nuclear weapons tests and airborne releases of radioactive material from nuclear power plants. The radionuclides from these and possibly other man-made sources are distributed over the entire earth by atmospheric movements. The radionuclide  $^{85}\text{Kr}$  is of especial significance in the gaseous releases of nuclear power plants, since its intensity is expected to build up over the years<sup>16</sup>, as more nuclear plants go into operation. Regional records of these radiations can be maintained without requiring the wearing of dosimeters by individuals if central monitoring stations are placed in selected locations, provided appropriate surveys are made of nearby areas, to map expected variations in radiation exposure levels relative to those at the central station. Readings from these monitoring stations would become part of the record for individuals living in the area.

Industrial radiation records need to be kept in much the same manner as they are now. However, they must be maintained throughout the lifetime of each individual, as part of his permanent record. All too often records have not followed individuals when they change employment, making compilation of cumulative dose records impossible. I can cite several specific instances where records have been lost or destroyed.

Record keeping of medical radiology presents a problem of its own. Generally, doses are not uniformly distributed over the body. Also radiation energies vary considerably among different diagnostic and therapeutic sources. For this reason some basic records indicating the size and shape of the radiation field and any variations in radiation energies over this field must be kept for each treatment. Also, the dose to selected regions of the body must be kept as part of the record for each individual. Only in this way can a true correlation ever be made between cumulative effects of ionizing radiations and any subsequent observation of diseases that may be linked to radiation exposure, of the type cited by Eason and Brooks<sup>12</sup> as having been considered in selected court cases. Such records of dose to individual organs of the body should not present as much of a problem as it may at first appear. Doses for radiation therapy are now carefully calculated, generally by computer, to be able to place the maximum dose in the appropriate region of the body. There is no reason why such computerized calculations cannot be made for diagnostic work as well, so that the dose to specific body organs can be recorded for each radiation<sup>17</sup>.

Since we currently have no record of cumulative dose for any adult in the United States, the best way to establish a valid record-keeping system is to begin with children who have a known past history of medical radiation, most probably none. The system therefore need not be an elaborate full-scale system.

in its initial stages, but can be built up over the years as more and more children's records are added. The system need not necessarily be established everywhere at once. A pilot system could be established in an area covering up to a few states. Such a system could provide enough information within about 10 years or so to allow us to determine its feasibility and to decide how far and how fast it should be expanded. Unless we start somewhere we will continue to argue about the merits of any system and, because of the size of the project, probably never do anything.

#### References

1. A. W. Klement, C. R. Miller, R. P. Minx and B. Shlcien, "Estimates of Ionizing Radiation Doses in the United States 1960-2000", Report ORP/CSD 72-1. U. S. Environmental Protection Agency, August 1972.
2. J. A. Lieberman, Physics Today, Vol. 24, No. 11, pp. 32-38 (1971).
3. "The Effects on Populations of Exposure to Low Levels of Ionizing Radiation", Division of Medical Sciences, National Academy of Sciences, November 1972.
4. A. Stewart, Health Phys., Vol. 24, pp. 223-240 (1973).
5. M. E. Gauden, "Genetic Effects of Radiation", in Medical Radiation Biology, G. V. Dalrymple et al. (editors). W. B. Saunders Co., Philadelphia, 1973. pp. 52-83.
6. R. H. Mole, Health Phys., Vol. 20, pp. 485-490 (1971).
7. P. J. Lindop and J. Rotblat, Bulletin of the Atomic Scientists, Vol. 27, No. 7, pp. 17-24 (1971).
8. P. M. Boffey, Science, Vol. 171, pp. 780-783 (1971).
9. For a discussion of the Shoreham power station controversy see Bulletin of the Atomic Scientists, Vol. 28, No. 6, pp. 24-36 (1972).
10. Code of Federal Regulations, USAEC Rules and Regulations, Title 10, Part 20 (10CFR20), "Standards for Protection Against Radiation", pp. 63-76a.
11. S. Antoku et al, Health Phys., Vol. 23, pp. 291-299 (1972).
12. C. F. Eason and B. G. Brooks, Health Phys., Vol. 23, pp. 431-432 (1972) and private communication.
13. J. R. Cameron, Health Phys., Vol. 21, pp. 879-880 (1971). See also T. E. Burlin and B. M. Wheatley, Health Phys., Vol. 24, p. 113 (1973).
14. K. O'Brien and J. E. McLaughlin, Health Phys., Vol. 22, pp. 225-232 (1972); R. E. Gold and D. S. Peacock, J. Geophys. Res., Vol. 78, pp. 577-587 (1973)
15. C. L. Lindcken, D. E. Jones and R. E. McMillen, Health Phys., Vol. 24, pp. 81-86 (1973).
16. Committee on Power Plant Siting, "Engineering for Resolution of the Energy-Environment Dilemma", National Academy of Engineering, Washington, D. C., 1972, pp. 180-191.
17. T. D. Jones, J. A. Auxier, W. S. Snyder and G. G. Warner, Health Phys., Vol. 24, pp. 241-255 (1973).



THE RAPID DETERMINATION OF THE TRANSURANIUM ELEMENTS BY  
EXTRACTION CHROMATOGRAPHY IN URINES CONTAINING DTPA

Delle Site A., Santori G., Testa C.,  
CSN-Casaccia, CNEN,  
Rome, Italy.

Abstract

Microporous polyethylene (Microthene-710) supporting tri-n-octylphosphine oxide (TOPO) and di(2-ethylhexyl) phosphoric acid (HDEHP) has been used successfully to extract respectively Th, Pa, U, Np, Pu, Am and Cm from the urine. As the extraction takes place in an  $\text{HNO}_3$  medium, DTPA up to 2 g/l does not interfere. A particular investigation was carried out for Am in order to find out the best extraction pH. Finally it has been demonstrated that great losses of actinides occur when a coprecipitation with Ca and Mg phosphates are carried out in the presence of DTPA.

Introduction

During ten year experience in our laboratory several methods have been prepared for the determination of some actinide elements in the urine by means of extraction chromatography.

"Extraction chromatography" or "reversed phase partition chromatography" is a very useful tool to transform a liquid-liquid extraction into a chromatographic procedure by supporting some organic extractants on an inert microporous support<sup>1-3</sup>. By using Microthene-710 (microporous polyethylene) as the inert support and tri-n-octylphosphine oxide (TOPO) and di(2-ethylhexyl) phosphoric acid (HDEHP) as the extractants, many radiotoxicological determinations were performed; the urine was stirred with a slurry of Microthene-TOPO for Th, Pa, U, Np and Pu<sup>4-9</sup> and with Microthene-HDEHP for Am and Cm<sup>10</sup>; after this batch extraction, the slurry is transferred into a chromatographic column from which the radionuclide is eluted selectively with a suitable solution. Table I summarizes the experimental conditions and the relevant literature<sup>4-10</sup>.

As it is well known, di-ethylentriaminopentacetic acid is widely used in case of acute contaminations with plutonium and americium. On the other hand, this complexing agent can seriously interfere with conventional radiotoxicological analyses: some authors recommend the complete destruction of DTPA by a wet ashing<sup>11-12</sup> and others claim a wet mineralization with  $\text{HNO}_3$  and  $\text{H}_2\text{O}_2$  would be sufficient before the coprecipitation of plutonium with Ca and Mg phosphates<sup>13</sup>.

The aim of the present paper is to demonstrate that the techniques based on the extraction chromatography can be used successfully, and without any change, with urine containing large quantities of DTPA.

### Experimental

#### Reagents and apparatus

TOPO was supplied by Eastman Organic Chemicals (USA) and HDEHP by K & K (USA). Microthene-710, 50-100 mesh, was obtained by the Columbia Organic Chemicals (USA). DTPA was supplied by Fluka (Switzerland). Pa-233, U-233, Np-237, Pu-239, Am-241 and Cm-242 came from the Amersham Radiochemical Centre (England). Th-234 ( $\text{UX}_1$ ) was prepared in our laboratory<sup>6</sup>.

The other chemical reagents were of analytical grade.

An Intertechnique liquid scintillation apparatus connected to a Laben 400 channel analyzer was used for alpha counting after the batch extractions.

For the counting of Th-234 and Pa-233 a low background beta detector was employed.

The final recoveries of the alpha emitters were calculated by alpha counting with an Ortec solid state detector after electroplating the radionuclide.

The other apparatus (pHmeter, magnetic stirrer, chromatographic columns, etc.) were of conventional type.

#### Experimental results

To 500 ml of urine containing 6,000 dpm of Pu-239 (~44 ng), different quantities of DTPA (10 mg, 100 mg and 1000 mg) were added and the conventional chromatographic extraction procedure was performed; the final yields were 75.5%, 78.5% and 79.5%, respectively showing that large quantities of DTPA can be tolerated.

Taking into account these results, 1 g of DTPA was added to 500 ml of urine containing the other radionuclides and three analyses were carried out to check the effect of DTPA on the final yields. Table II summarizes the obtained results and it shows that DTPA does not interfere with the analysis, except for americium and curium for which the extraction takes place at pH 3.

TABLE I  
DETERMINATION OF ACTINIDES IN THE URINE BY BATCH EXTRACTION  
WITH MICROTHENE SUPPORTING SOME ORGANIC EXTRACTANTS

NU- CLIDE	HNO <sub>3</sub> CONC.	STATIONA- RY PHASE	ELUTING AGENT	RECO- VERY %	SENSITI- VITY LI- MIT	ME- THOD (a)	TIME H.	REF.
Th	4.0 M	0.5M TOPO	0.3M H <sub>2</sub> SO <sub>4</sub>	98.2	0.2 /ug/1	Col.	4	4
Pa	4.0 M	0.1M TOPO	6M HCl+0.2M HF	-	-	ZnS	4	5
U	4.0 M	0.5M TOPO	1M HF	70.0	1 dpm/1	ZnS	4	6
Np	6.0 M	0.1M TOPO	6M HCl+Cl <sub>2</sub>	83.2	0.05 pCi/1	SSD	8	7
Pu	4.0 M	0.3M TOPO	6M HCl+0.01 HI	70.5	0.07 pCi/1	SSD	8	8
	4.0 M	0.3M TOPO	6M HCl+0.1 HI	76.5	0.10 pCi/1	ZnS	4	9
Am (Cm)	0.001M	1.5M HDEHP	3M HNO <sub>3</sub>	35.9	0.05 pCi/1	SSD	8	10

(a) Col. = colorimetry; ZnS = alpha counting with a ZnS(Ag) detector;  
SSD = alpha counting with a solid state detector after electroplating.

TABLE II  
RECOVERIES OF ACTINIDES BY EXTRACTION CHROMATOGRAPHY WITHOUT  
AND WITH DTPA

RADIONUCLIDE	FINAL RECOVERY % WITHOUT DTPA (a)	δ %	FINAL RECOVERY % WITH 1 g OF DTPA IN 500 ML OF URINE (b)	δ %
Th	98.2	11.5	93.0(c)	1.7
Pa	-	-	91.1(d)	2.7
U	70.0	3.0	70.2	2.2
Np	83.2	6.5	79.8	4.7
Pu	76.5	5.7	76.7	2.5
Am(Cm)	35.9	7.6	0	-

(a) Average of 10 analyses; (b) Average of 3 analyses; (c) Th-234 radimetric determination; (d) % Recovery of the only extraction.

It appears that in 4-6 M  $\text{HNO}_3$  the actinide elements are not complexed by DTPA.

A study was then performed to find out the possibility to extract also americium and curium in the presence of DTPA by decreasing the pH of the solution: in fact in this case there are two opposite effects, i.e. the extraction of Am which increases by increasing the pH value, and the complexing of Am by DTPA which also increases with the pH. Therefore it was decided to study the extraction of americium as a function of the DTPA concentration and of the pH of the solution. As Fig. 1 shows by stirring for 60 minutes 500 ml of urine without DTPA with 7 g of Microthene supporting 5 ml of 1.5 M HDEHP in xylene, the Am extraction is complete above pH 2.5; if some DTPA is present an extraction maximum takes place at a pH which decreases by increasing the quantity of DTPA. In any case it is always possible to obtain a sufficiently good extraction (56% to 86%) by using a suitable pH.

By fixing the pH at 2 and by increasing the DTPA concentration from 10 to 100 mg (fig. 2) the Am extraction decreased from 73 to 60%. Similar results were obtained with curium.

### Discussion

From the experimental results it appears that by using the extraction chromatography it is always possible to obtain sufficiently good recoveries of the actinides from the urines, even in the presence of DTPA.

To be sure that the clearing of the urine by boiling with  $\text{HNO}_3$  was not the cause of the DTPA removal<sup>13</sup>, some analyses were carried out by adding the complexing agent after the wet mineralization of the urine: also in this case the actinides extractions were the same, showing that the DTPA present does not form complexes with these elements at high  $\text{H}^+$  concentrations<sup>14-15</sup>.

On the other hand, the methods for the complete mineralization of the organic matter<sup>11-12</sup> are very time-consuming.

It has also been reported<sup>13</sup> that a wet-15 minute mineralization with 85 ml of conc.  $\text{HNO}_3$  and 8 ml of  $\text{H}_2\text{O}_2$  for 1.5 liter of urine could destroy the DTPA and permit plutonium to coprecipitate quantitatively at pH 3.5-9.0 with calcium and magnesium phosphates. On the contrary, we have found that this treatment is not sufficient to eliminate the effect of DTPA, as fig. 3 shows: in fact, boiling 500 ml of urine containing DTPA (0-1 g) with 100 ml of conc.  $\text{HNO}_3$  and 20 ml of  $\text{H}_2\text{O}_2$  for 60 minutes, the precipitation yield decreased from about 98% to about 39%. This is due to the precipitation pH (8.5-9.0) where the complex Pu-DTPA does take place.

Concluding, the described methods of extraction chromatography are very simple and rapid and they do not require any dry mineralization or coprecipitation.

The only drawback which exists for the americium and curium determination is to know roughly the DTPA concentration in the

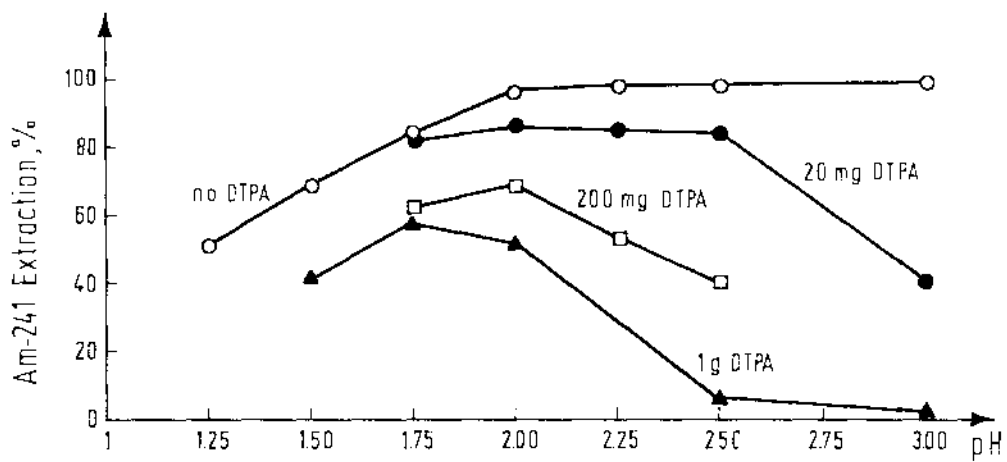


Fig 1 Extraction of Am-241 from 500ml of urine by Microthene-RDEHP as a function of the DTPA concentration.

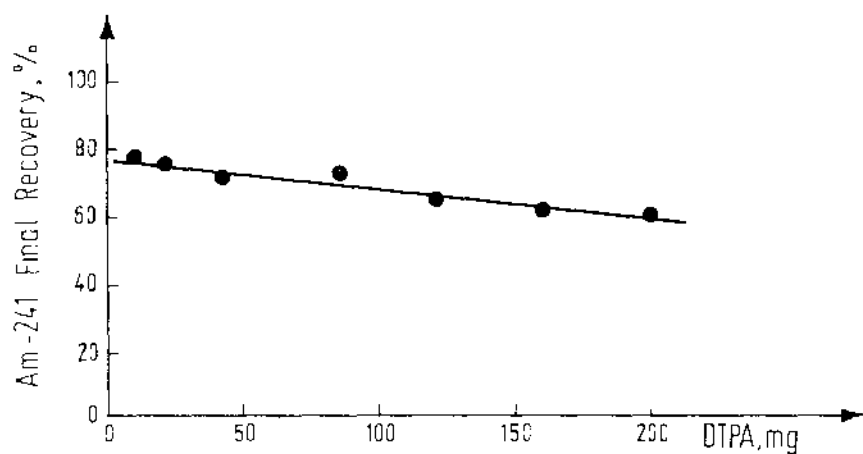


Fig 2 Am-241 final recovery with Microthene-RDEHP from 500ml of urine at pH 2 as a function of the DTPA concentration.

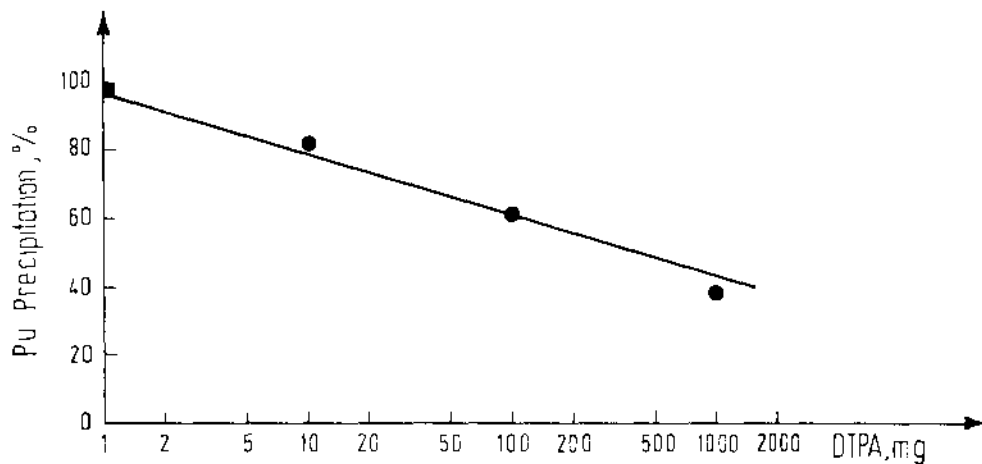


Fig 3 Precipitation of plutonium from 500 ml of urine wet mineralized with  $\text{HNO}_3$  and  $\text{H}_2\text{O}_2$  as a function of DTPA concentration

urine; however this is easily estimated by considering that almost all the injected DTPA is excreted in the first 24 hour urine sample<sup>16</sup>.

#### Reference

- 1) Testa C., *Analytica Chim. Acta*, 50, 447, 1970.
- 2) Testa C., Staccioli L., *Analyst*, 97, 527, 1972.
- 3) Testa C., "Assessment of Radioactive Contamination in Man", IAEA, Vienna, 1972.
- 4) Testa C., "Radiological Health and Safety in Mining and Milling of Nuclear Materials", IAEA, Vienna, 1964.
- 5) Santori G., Testa C., to be published.
- 6) Testa C., De Rosa D., Salvatori A., CNEN Report RT/PROT(68)6, 1968.
- 7) Santori G., Testa C., to be published on the *J. of Radioanalyt. Chemistry*.
- 8) Testa C., Santori G., *Proceedings of the 16th AIFSPR Congress*, 596, 1970.
- 9) Testa C., Santori G., *Minerva Fisiconucleare*, 16, 1, 1972.
- 10) Testa C., Delle Site A., Santori G., *Proceedings of the Regional Conference on Radiation Protection, Jerusalem*, 1973.
- 11) Low-Beer A.G., Parker H.G., *Health Physics*, 11, 61, 1965.
- 12) Horn I.F., *Health Physics*, 21, 41, 1971.
- 13) Valentin N., *Health Physics*, 12, 1933, 1966.
- 14) Piskunov E.M., Rykov A.G., SRARI-P-92, 1970.
- 15) Delle Site A., Baybarz R.D., *J. In. Nucl. Chem.*, 31, 2201, 1969.
- 16) Foreman H.: "Metal-binding in Medicine", J.B. Lippincott Ed., p. 82 and 160, 1960.

EXCRETION OF TRITIUM IN WORKERS PREPARING  
LABELLED COMPOUNDS

Houšková, M., Čajková, A., Kunz, E., Náměstek, L.

Institute of Hygiene and Epidemiology  
Praha, Czechoslovakia

Abstract

Analysis of results of tritium excretion follow-up in workers preparing labelled compounds proved the applicability of the method of measuring tritium in exhaled breath to the evaluation of HTO excretion, and showed the need to determine organically bound tritium or the total activity of tritium in urine. The mean ratio of exhaled breath to distilled urine activities amounted to  $0.88 \pm 0.18$ . The biological half-life  $T_b$  of HTO excretion was  $11.4 \pm 2.1$  days on the average. In a worker followed up for 600 days following exposure another half-life component was determined of  $240.2 \pm 8.4$  days. Organically bound tritium was excreted with a half-life lower than 2.5 days.

Starting in 1971, we have systematically followed up excretion of tritium in workers engaged in the synthesis of labelled compounds and in workers operating neutron generators, with the aim to improve the methods of measurement and evaluation of exposure to tritium.

Methods

Sampling of urine and exhaled breath was carried out in an inactive environment as a rule before starting work, but also after termination of individual operations and during a longer absence of the subject at the workplace. HTO from exhaled breath was trapped by molecular sieves (Nalsit 4A), desorbed by vacuum distillation at  $400^\circ\text{C}$  and frozen at  $-80^\circ\text{C}$ . Tritium was also determined in samples of non-treated native urine or in distilled urine following removal of interfering substances by sorption on powdered activated charcoal. Distillation was carried out to dryness in order to exclude the isotopic effect. Organically bound tritium was estimated from the difference of the native urine and distilled urine tritium activities, or by determining the tritium activity of lyophilized urine samples. The latter method seems to be more accurate. All measurements have been carried out with a Tri-Carb Liquid Scintillation Spectrometer, model B 3003 (Packard) with the use of a scintillator Insta-Gel for samples of native and lyophilized urine, or of a dioxane scintillator composed of POP, dimethyl POPOP and naphthalene for distilled urine. For evaluation of the

specific activity the method of internal standard has been used. The reproducibility of the method is  $\pm 3$  per cent, the lowest detectable activity of tritium in distilled urine is 3.5 pCi/ml for the counting time of 500 min and the error of measurement  $\pm 10$  per cent.

### Results and Discussion

#### Tritium in Exhaled Breath

Fig. 1 illustrates the distribution of values for the ratios of tritium specific activity in HTO from exhaled breath and distilled urine. The data represent results of 40 simultaneous exhaled breath and urine samples taken from 6 persons after a sufficiently long time following possible internal contamination. The mean ratio of  $0.88 \pm 0.18$  is in agreement with data of other authors<sup>1,2</sup> and indicates a probable isotopic effect in the lung. Repeated determinations in the same persons prove the reproducibility of the method. The reliability of the method is indicated by an excellent agreement of the course of tritium excretion by urine and exhaled breath during the long-term follow-up (Fig. 3). The advantage of the method as compared with the measurement of tritium in distilled urine consists in the possibility of choice of the sampling time, in shorter treatment and easier storing of samples, as for instance when sampling outside the workplace. However, the workers examined prefer urine sampling.

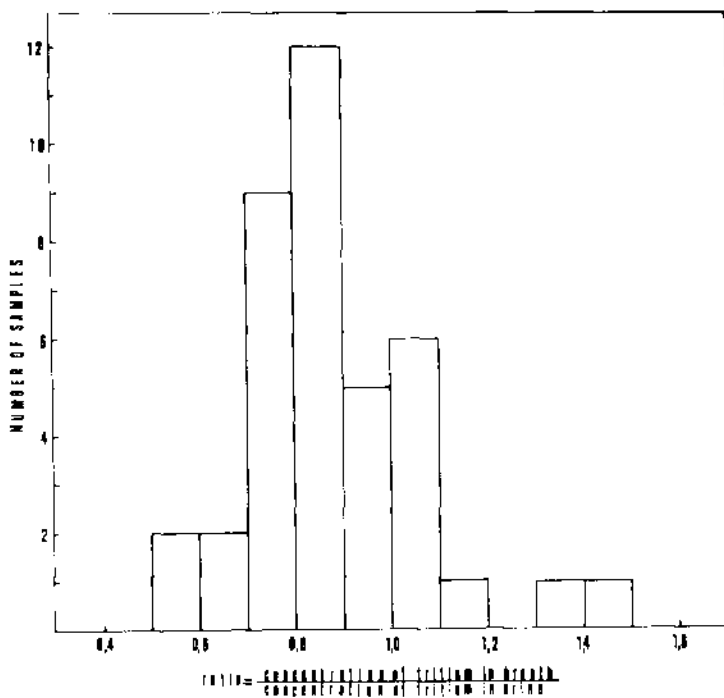


Fig. 1. Comparison of breath and urine samples.



### The Course of HTO Excretion

The values for HTO biological half-lives calculated from the results of measurements of distilled urine or exhaled breath samples taken at the periods when the subjects were removed from work are shown in the following Table.

Biological half-lives of HTO

Subject	$T_b$ (days)	Water phase	Sampling time
V	8.6±0.5	D.U.	May 1971
	10.2	D.U.	August 1971
	23.0±5.4	D.U.	April 1972
	17.6±8.9	D.U.	May 1972
B	13.6±2.9	D.U.	March 1971
	16.1	D.U.	July 1971
	12.0	D.U.	February 1972
S	9.3±0.4	D.U.	September 1972
K	8.9	D.U.	March 1972
M	10.1	D.U.	March 1972
J	12.2±0.9	D.U.	March 1972
H	11.7	D.U.	March 1971
	11.4	D.U.	April 1971
Č	12.67±0.03	D.U.	September 1971
	12.64±0.06	E.B.	September 1971
	240.2±8.4	D.U.	
	268.7±14.7	E.B.	

D.U. = distilled urine  
E.B. = exhaled breath

Except one case (subject Č), the persons examined remained out of exposure only for relatively short periods of time. The mean value of  $T_b = 11.4 \pm 2.1$  days corresponds to the data published elsewhere.<sup>3,4</sup> The subject V exhibited higher values of  $T_b$  during his two stays in high mountain conditions (Fig. 2 B,C), contrary to his previous value of  $T_b$  (Fig. 2 A). This finding cannot be explained reliably and the data obtained were not included in the calculated mean. The data of a systematic follow-up in the period prior to his leave of absence exclude the possibility of a longer component of excretion to exert itself. In subject Č it was possible to follow up excretion of tritium for 600 days following discontinuance of work in preparation of labelled compounds (Fig. 3). Analysis of the results led to the determination of another component of excretion. The value of  $T_b$  (see Table) agrees with the findings of other authors.<sup>5,6,7</sup> The equation characterizing the HTO excretion by Č is of the form

$$A_t = 5658.0 e^{-\frac{0.693}{12.67} t} + 14.3 e^{-\frac{0.693}{240.2} t},$$

where  $A_t$  is activity in time  $t$ .

The relative contribution of tritium excreted with a longer half-life (0.25 per cent of the total amount of tritium) to the dose commitment is negligible.

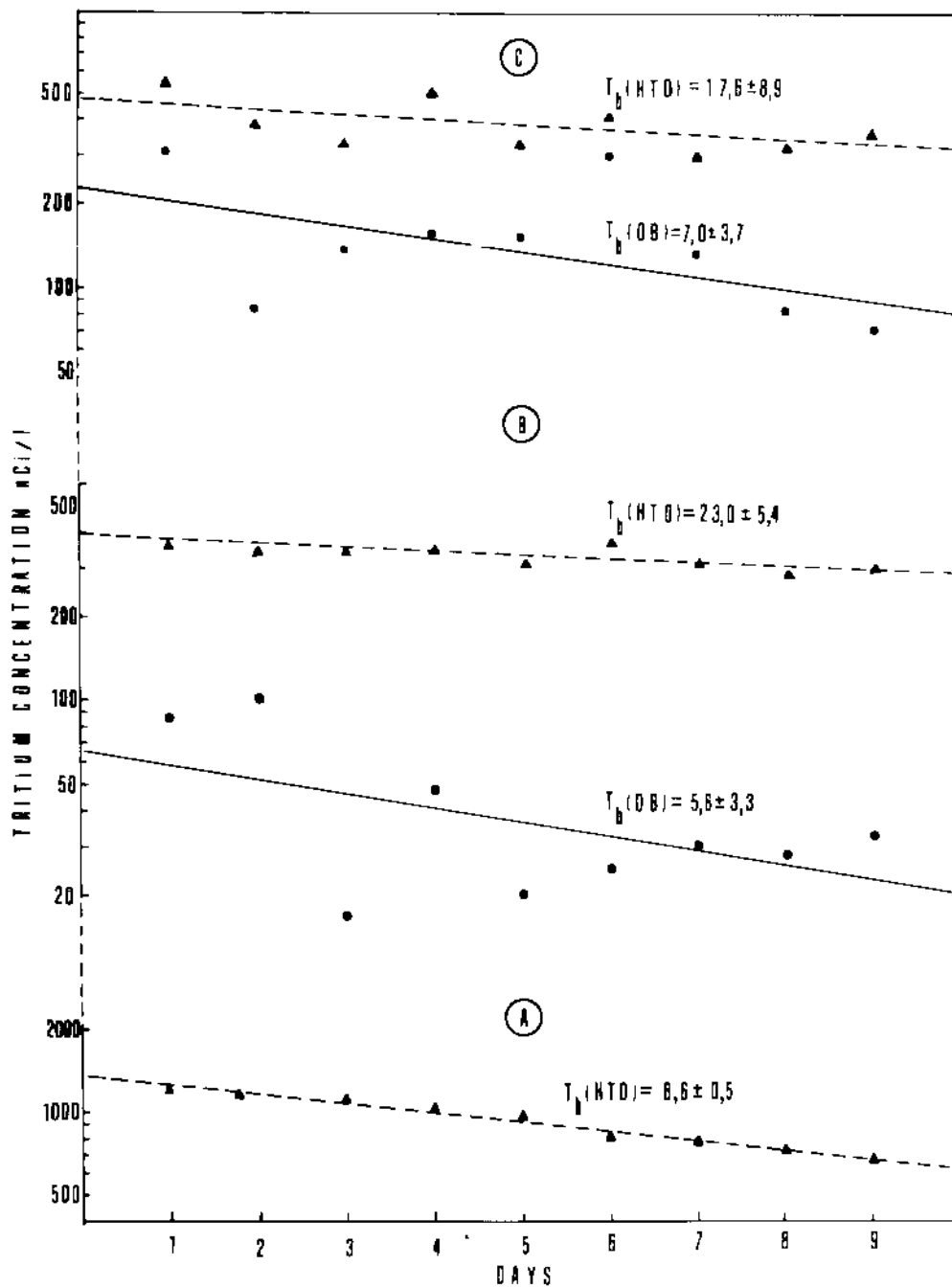


Fig. 2. Urinary excretion of tritium by subject V.  
 ▲ distilled urine,  
 • organically bound tritium

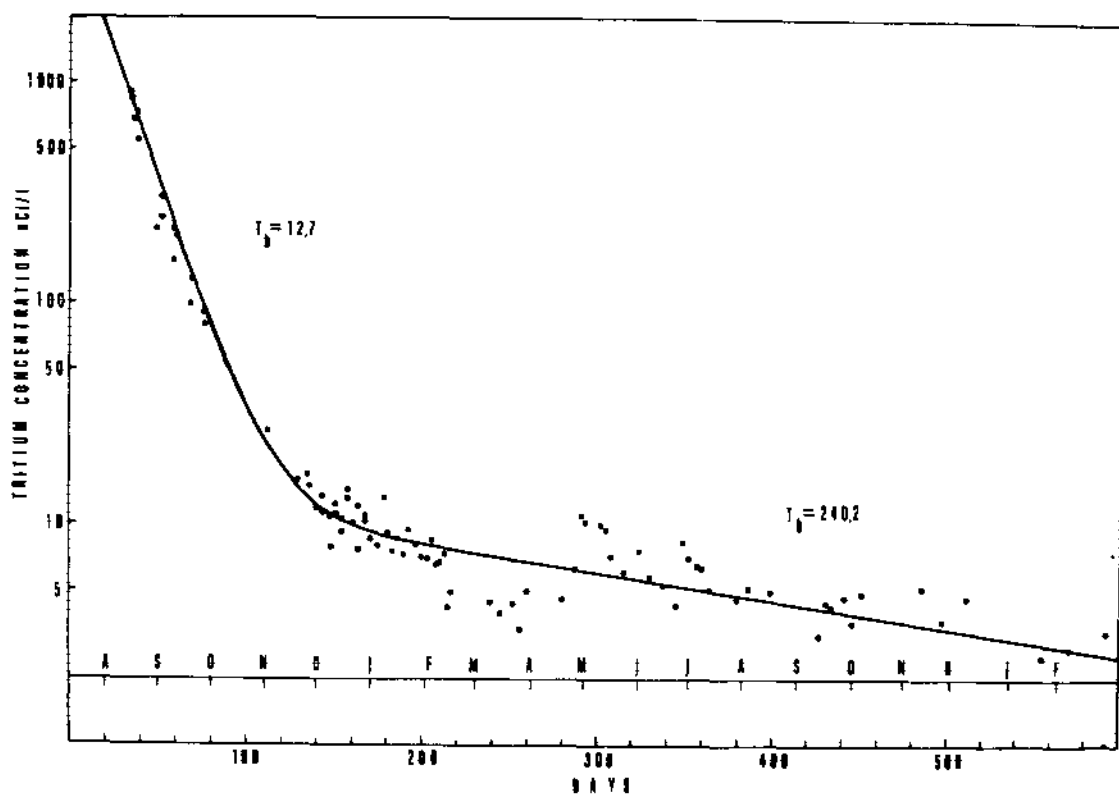


Fig. 3. Excretion of tritium in urine and exhaled breath by subject C.  
 • distilled urine,    • exhaled breath

#### Organically bound tritium

In workers engaged in the synthesis of labelled compounds varying values of organically bound tritium were observed. Organically bound tritium amounted, as a rule, to tens per cent out of the total amount of tritium in urine, in some cases, however, it exceeded many times the activity of HTO. In Fig. 4 the values are plotted of tritium activity in native and distilled urine as well as the difference of these values corresponding to the organically bound tritium. The values were obtained from two persons during the period of intensive follow-up. The biological half-lives of organically bound tritium were  $2.1 \pm 0.4$  for S and  $2.5 \pm 0.4$  for B. The real values might be lower, since additional contamination of the workers who continued in tritium work cannot be excluded. Following experimental percutaneous absorption of gaseous tritium, Eakins et al.<sup>8</sup> observed urinary excretion of organically bound tritium (mixture of organic acids) of 2 half-lives, 5 hours and 1.7 days. Koch et al.<sup>9</sup> found very short biological half-lives of various defined organic compounds labelled with tritium in persons engaged in work with these substances. Both possible sources of organically bound tritium in urine must be considered at the followed up workplaces. In conditions of our study it was not possible for the time being to prove the existence of a very short component of excretion ( $T_D$  of a few hours), nor to identify individual organic compounds in urine. Fig. 2 B,C shows the course of

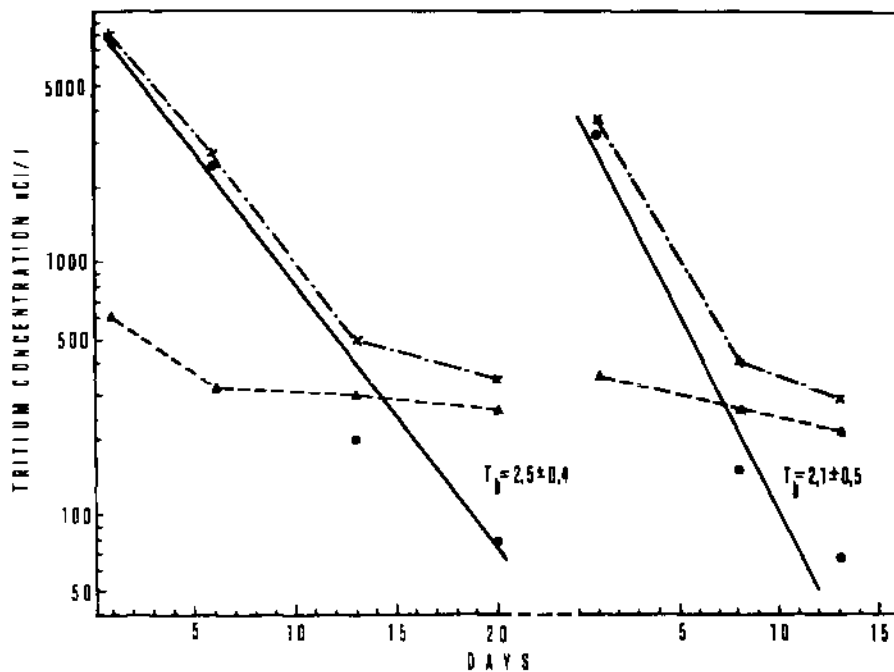


Fig. 4. Urinary excretion of organically bound tritium by subjects B and S.  
 -.-.- native urine, - - - distilled urine,  
 ——— organically bound tritium

excretion of organically bound tritium (according to activity of lyophilized urine) in subject V during his high mountain recreation. The biological half-lives were  $5.6 \pm 3.3$  and  $7.0 \pm 3.7$  days for the two periods of follow-up respectively. The values are higher than those observed in subjects B and S during tritium work. In workers operating neutron generators, the activity of organically bound tritium was below 1 per cent of the total activity of tritium in urine.

The possibility of intake of tritiated organic compounds or their formation in the organism during exposure to gaseous tritium is of importance when deciding on the methods, frequency of sampling and interpretation of results in monitoring of tritium exposure. The determination of tritium only in the form of HTC can lead to serious underestimation of the exposure, the evaluation of native urine activity according to HTO kinetics parameters can result in its overestimation.

#### References

1. Chiswell, W.D., Dancer, G.H.C., Health Phys. 17, 1969, 331-334.
2. Siri, W., Evers, J., Tritium in the Physical and Biological Sciences, IAEA, Vienna, 1962.
3. Butler, H.L., Leroy, J.H., Health Phys. 11, 1965, 283-285.
4. ICRP Publ. 10, 1968.
5. Moghissi, A.A., et al., Health Phys. 23, 1972, 808.
6. Reinig, W.C., Sanders, S.M., DP-MS-66-77, 1967.
7. Minder, W., Strahlentherapie 137, 1969, 700-704.
8. Eakins, J.D., et al., Congr. of IRPA, Brighton, 1970.
9. Koch, G.F., et al., 2. Congr. of IRPA, Brighton, 1970.

## OCCUPATIONAL EXPOSURE TO IONIZING RADIATION IN IRAQ

MOUSA S. ABDAS  
Radiation Control Office  
Ministry of Health  
Baghdad - Iraq

### Abstract

The external exposure of radiation workers in the Republic of Iraq are checked by requiring from the workers to wear film badges. There are only two laboratories of film dosimetry concerning the control of occupational exposures to ionizing radiation.

Analysis of the data obtained in 1972 for the yearly exposures of persons at medical service in comparison to persons engaged in research and in industrial radiation works is available.

The evaluation of external radiation exposures received by more than 897 individuals has been for 1972 in comparison with that for the period 1962 - 1971.

During the period 1969-1972 the annual exposures did not exceed 500 mrem in 95% of the occupationals and less than 1% exposures were higher than 5 rem were received and the total is about 400 man-rem in 1972. The exposures are very low in average.

It was found that exposures in men is significantly higher than in women, and the technical assistants were exposed significantly represent the major occupational personnel (about 70%).

In addition the present paper includes the total number of radiation workers, total personnel exposure and percentages of yearly exposures received.

### I- Introduction

Exposure to any harmful levels of ionizing radiation can not be directly detected by the human senses. Therefore, it was necessary to look for instruments to fulfill this job. These instruments are called the dosimeters. One of the most important dosimeters for personnel monitoring is the film Badge/ I /. In Iraq there are now two well equipped laboratories dealing with occupational exposure control.\*

The first laboratory was established at the Iraqi Atomic Energy Commission (IAEC) in 1962, This laboratory is now responsible only for the exposure assesment of the IAEC employees. The second it is concerned with the occupational exposures of the employees of the country except the IAEC. This is the Radiation Control Office

This paper discusses the results of the two laboratories and

---

\* The term "occupational exposure" is used in this paper as used by the ICRP/ 2 / to apply to all activities involving exposure of workers to ionizing radiation in the course of their work, regardless of whether the workers are directly engaged in radiation work or not .

gives some evaluations about the radiation exposure levels of about 900 persons in Iraq. It is expected that this service would be gradually extended to include more than 1200 persons working on radiation sources at the end of 1973.

## II. Materials and Methods

### I. Number of radiation sources in Iraq

The ionizing radiation sources in Iraq are as in other developing countries, consisting mainly of the sources and apparatus which are used for medical ( diagnosis and therapy ), dental, veterinary, industrial, research and educational, and atomic energy purposes. Table I lists the radiation sources served by Film Badge in Iraq. Most of these sources are medical and dental purposes. The number of radiation installations used for veterinary, industrial, and research and educational purposes, is at present very small compared with those used for medical purposes.

Table I : Number of Radiation Sources up to the end of 1972 served by film badges.

Radiation Source	Total number
X-ray diagnostic machines	300
X-ray therapeutic	17
Dental X-ray machines	36
X-ray veterinary machines	2
Industrial X-ray radiographic machines	7
Gamma-ray therapeutic units using Co <sup>60</sup>	3
Gamma-ray industrial radiographic units using Co <sup>60</sup> and I <sup>192</sup> Ir.	7
Gamma-ray agricultural unit using Co <sup>60</sup> Research 2 MW Reactor.	1
Isotope production laboratory and some radioactive research laboratories.	1
Isotopes clinics	3
Some laboratories using radioactivity for educational purposes.	

### 2. Number of radiation workers

Workers who are supplied with Film Badges are distributed all over the radiation fields of the country.

Nearly about 75% of the total number of persons dealing with the radiation sources have been supplied with Film Badges.

This number increased from 73 to 397 during the past eleven years ( 1962 - 1973 ).

The occupational persons up to end of 1972, are classified according to the nature of radiation work as summarized in Table 2 .

### 3. Dose Measurements

The photographic film is advantageous since it integrates the radiation exposure irrespective to exposure rate. In

Iraq, we use dosimeter film pocket type M.R.I. 30 Black spot Film Badge with Kodak Radiation Monitoring Combined Emulsion Film (3.2 x 4.1 cm). It is possible to measure exposures to different types of radiation from 10 mR up to more than 20 R, with an error in our processing conditions and evaluating of the films within the range  $\pm 20\%$ . Therefore, the minimum exposure recorded in a film badge report is usually 20 mR/ 3-4/....

The film badge usually worn while the workers are on duty. These films are used for one month by wearing them on the part of worker's body which might receive the largest amount of radiation.

Exposed films before developing must be checked for radioactive contamination. Films are developed with exposed film to  $^{228}\text{Ra}$ ,  $^{60}\text{Co}$  and  $^{137}\text{Cs}$   $\gamma$ -rays and unexposed control film.

The number of films sent for use and the actual films evaluated over the past five years are shown in Table 3.

The results of the film measurements are recorded in personnel cards containing also a description of the work, the place of work, the source of radiation used, the qualifications and the ages of the workers and the starting date of working in the radiation field. The results are usually sent back to different offices (work place).

Table 3. Total number of films sent to be used and the number of films returned back.

Year	1968	1969	1970	1971	1972
Sent abs	1542	4817	7384	7615	6477
Returned %	34.5	34.6	34.7	34.7	34.2
Returned abs	528	1668	2563	2628	2217
%	34.5	34.6	34.7	34.7	34.2

### III- Results and Discussion

Data collected for the external radiation exposures received by the occupational workers for the all period of the film badge service in Iraq is summarized in Table 4. The table shows the distribution for six ranges of doses received.

From this table especially in the last nine years the percent ages distribution of the followed up persons were in principle the same. One point emerges fairly clearly, namely the great majority of these persons receive less than one-tenth of the maximum permissible annual dose.

Cases of over-exposure are very few. In about 0.5% of workers the measured exposures amounted to more than 5 rem.

Table 4 also showed average figures for the annual occupational exposure to individuals. The extrapolated yearly averages of the doses to all the radiation workers were obtained between 445 m Rem for the last nine years. This is because the great the detection limits of the film badges (20 m rem/month). These results

Table 2. The followed up persons in 1972 according to work places.

Type of work	pers.	organ.
1. Medical X-ray		
Diagnostic	465	41
Therapy	19	6
Dental	26	20
Veterinary	4	2
Isotopes (Medicon)		
Diagnostic	61	2
Therapy	33	2
2. Industry	6	4
3. Reactor	267	1

reflected on the total annual dose in man-rem which was in 1972 for 897 persons about 360 man-rem/year.

Table 4. Distribution of occupational by years in according to the six ranges of annual doses in rem.

Year	< 0.5	0.5-1	1-1.5	1.5-3	3-5	5-10	>10	Total	average man-rem/yr
1962	abs 37	19	13	5	4	-	-	87	646
	% 45.3	22.1	22.1	5.9	4.7	-	-		
1963	abs 126	30	7	6	2	-	-	171	554
	% 73.7	17.5	4.1	3.5	0.3	-	-		
1964	abs 105	4	6	3	-	-	-	130	357
	% 93.5	2.0	3.0	1.5	-	-	-		
1965	abs 263	19	5	8	3	1	-	304	445
	% 88.2	6.3	1.6	2.6	1.0	0.3	-		
1966	abs 355	4	4	2	1	1	-	370	358
	% 96.7	1.1	1.1	0.5	0.3	0.3	-		
1967	abs 346	3	2	-	6	2	-	359	391
	% 96.6	1.6	0.4	-	1.1	0.3	-		
1968	abs 613	22	5	7	2	2	1	639	377
	% 94.0	3.4	0.8	1.1	0.3	0.3	0.1		
1969	abs 549	27	6	1	6	1	3	639	347
	% 93.7	3.9	0.9	0.1	0.9	0.1	0.4		
1970	abs 713	62	9	2	1	1	2	790	368
	% 91.4	7.8	1.0	0.3	0.1	0.1	0.3		
1971	abs 749	32	3	6	-	-	2	790	367
	% 93.8	4.0	1.1	0.8	-	-	0.3		
1972	abs 649	33	7	7	1	-	-	697	399
	% 44.6	3.7	0.8	0.3	0.1	-	-		

The high results of the first two years after the establishment of the first laboratory of film badges may be as a result of the lack of experience of the staff.

Table 5 shows average figures for the annual occupational exposure to individuals for the six ranges of doses received from external radiation sources in various kinds of radiation work for the last two years (1971-1972). Significantly relatively higher exposures were found among medical personnel. The persons at industrial radiography never received a dose higher than one-tenth of the maximum permissible dose. In 1971 there were only two cases in which two persons received a dose of 200% of the maximum permissible dose only once. One of them was medical personnel while the second was employed in the Nuclear Research Institute.

The routine radiation test especially in the I.L.C. Laboratories in fact, a good indication of what we have achieved over the years in guiding operators towards safer working habits. The results of the mean annual doses (in r-rem) of the external ionizing radiation of occupationally exposed persons in comparison to that of persons engaged in other offices as shown in Table 6. The analysis of the



doses received by the employees of IIRC and other radiation workers show the gradual reduction in the mean annual doses of the IIRC employees.

Table 5. Distribution of workers by type of work carried out according to the magnitude of annual exposures in Rem for the two last years.

ranges of doses	<0.5	0.5-10	1.-1.5	1.5-3	3-5	5-10	>10	Total
Medical	1971	513	20	0	3	-	1	545
	1972	556	27	7	7	1	-	598
Dental	1971	20	2	1	-	-	-	23
	1972	25	1	-	-	-	-	26
Atomic	1971	210	10	-	3	-	1	224
	1972	262	5	-	-	-	-	267
Industry	1971	6	-	-	-	-	-	6
	1972	6	-	-	-	-	-	6
Total		749	32	9	6	-	2	798
		849	33	7	7	1	-	897

Table 6. A comparison of mean annual dose received by workers of IIRC and other radiation workers.

This gradual reduction is due to the good trained team of the Health Physics Department of the IIRC and the high percentage of college graduate personnel among the occupational.

Type of work:	in Rem/year				
	1968	1969	1970	1971	1972
Atomic	254	244	233	214	230
Others	314	401	398	272	471

Table 7. shows the distribution of workers by sex and educational level. Two important points emerge, however, namely that out of 897 radiation workers at the end of 1972, there were 86.3% of the total workers men and 13.7% women.

Higher exposures is found among men than women. The higher exposure of men may be due to the work connected with greater risks of irradiation carried out by men.

The second point is that the number of ungraduated persons were more than graduated, and the unqualified were exposed in all branches of both sexes more than graduated personnel. The lower exposures in qualified persons may be due to their awareness of the nature of their work.

#### IV- Summary

The measurement of personnel exposures by film badges in Iraq is undertaken by two laboratories. Dose of exposure in the range of 10  $\mu$ rem to 20 rem can be detected, with the error not exceeding  $\pm$  20%. The minimum dose recorded is 20  $\mu$ rem/month. Statistical assessment of measurements was carried out in relation to character of work, the source of radiation used, the sex and qualifications of the workers.

Table 7. Distribution of workers by sex and education according to the magnitude of annual exposures in Rem for the last two years ( 1971-1972 ).

Sex	Annual Exposure	< 0.5	0.5-1	1-1.5	1.6-3	3-5	5-10	>10	Total
Female	A	1971	36	2	1	0	0	0	39
		1972	51	1	2				54
	B	1971	42	4	2	1			49
		1972	68	1	-	-	-	-	69
	Total	1971	78	6	3	1			88
		1972	119	2	2				123
Male	A	1971	204	10	3	2			219
		1972	239	6	1	-	1		247
	B	1971	457	16	3	3	-	-	491
		1972	491	25	4	7	-	-	527
	Total	1971	671	26	6	5		2	710
		1972	730	31	5	7	1	-	774
Total	1971	749	32	9	6	-	-	798	
	1972	849	33	7	7	1	-	897	

A= College graduates: operators, physics engineers, physicians, etc.

B= Technicians, laboratory assistants, janitors, mechanics, etc.

The annual exposures were very low in average, and did not exceed one-tenth of the maximum permissible dose in 94% of the followed up persons in average. The mean annual exposure dose per person nearly the same for the last nine years (it ranges from 347 to 445 mrem/year). For good control in IALGO the average year by dose per man generally tended to decrease.

#### References

1. Hugh F. H. Enry-Fundamentals of radiation protection wiley - interscience U.S.A. 1969.
2. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation, Un, No. 16 (A/5216), New York (1962).
3. Recommendations of the ICRP. ICRP-publication No. 6. Pergamon Press (1962).
4. Morgan K.Z. Dosimetry Requirements for Protection from ionizing radiation. Selected topic in radiation dosimetry. IAEA-1961.

A NEW TECHNIQUE FOR IMPROVED TRACK RECOGNITION IN  
NUCLEAR EMULSION FILM DOSIMETRY USING SOFT X-RAYS

V. Balasubrahmanyam and L. Szegedi  
Radiation Protection Bureau  
Health and Welfare Canada  
Ottawa, Canada

The property of grain size and granularity variation with X and gamma radiation of different energy is used to improve the recognition of proton tracks in nuclear emulsion. Suitable x-ray qualities and exposure level have been investigated for this purpose. It is found that using x-ray of 10 kV and pre-exposure of 30 mR, the sensitivity of the neutron film could be increased by 15%. This study is based on exposures made using a Pu:Be neutron source.

This enhancement relieves considerable eye strain of the scanner and in addition, one is able to pick out easily shorter tracks in neutron monitoring films.

### Introduction

In the Radiation Protection Bureau (RPB) film dosimeters have been used for many years for monitoring neutrons. In the course of regular scanning of these films, very often films are found with slight exposure to gamma radiation. It is thought that this gamma radiation comes from standard neutron sources containing a small percentage of gamma contamination (2-4%) or from a reactor environment where mixed neutron gamma radiation fields exist. Further, this effect is also noticed in film dosimeters that are accidentally exposed to extremely small amounts of light. The operators noticed a very pronounced "Contrast Effect" when scanning such film dosimeters through a microscope or a TV video set up. Films of this sort were found to require shorter scanning time and there was less strain on the eye. It was observed that especially the shorter tracks were picked out easily.

It is this latter observation that suggested the application of the "Contrast Effect" into practical dosimetry. This paper studies the results of the visual and quantitative effect on contrast as a function of x-ray exposures and varying qualities of soft x-rays.

### Neutron Dosimeter

As for dosimetry of fast neutrons, a conventional neutron monitoring film from Kodak called NTA Type "A" is used. Neutron films were exposed to 22°C at a relative humidity of 45%. To avoid fading problems, they were then processed promptly after exposure; the processing time in a standard fresh Kodak liquid x-ray developer was 12 minutes and the corresponding time for fixing was 24 minutes.

### Radiation Sources

A Pu:Be neutron source in free air was used for all neutron exposures, which were made perpendicular to the film plane. The films were not in their normal holders and the accuracy of neutron exposure were within  $\pm 10\%$ .

For the x-ray exposures, two different x-ray machines were used. The one used at RPB for 25 kV measurements is a Philips tube with kV selection from 25 to 100 having 1 mm Be window. The x-ray exposures were determined with an accuracy of  $\pm 5\%$  using a calibrated EIL chamber. The 10 and 15 kV measurements were made with an x-ray machine available at the National Research Council (NRC). This machine is also a Philips special tube for precise x-ray output calibration

type 150 kV with 1 mm Be window. The x-ray exposure measurements at NRC were made to an accuracy of less than 1% using a special nylon chamber.

High energy x-rays were obtained from a 250 kV Philips therapy tube with 2 mm Al inherent filtration. A Ra-226 source enabled gamma exposures to be made.

### Microscopy

The fast neutron dose was related by calibration to the proton recoils density per unit area. A metallurgical research microscope Model Cooke 40487 with a TV monitor was used for this purpose. One field of view corresponded to  $4.3 \times 10^{-4}$  tracks  $\text{cm}^2$  with a dry objective x 20. The tracks were counted in an area covering 150  $\text{mm}^2$  in three different films for each exposure and then averaged out.

The optical quality of a recoil proton track in a neutron film is generally influenced interdependently by the following factors. (1) Transparency of the film, (2) Lack of uniformity of development with depth and area, (3) Poor discrimination for desired tracks in the presence of unwanted details, (4) Distortion of track trajectories, (5) Fogging of the emulsion due to gamma or electron radiations, (6) Illumination of the field of view.

### Technique

The technique is based on a well known physical property of radiation interaction with photographic emulsion. In the case of x-ray interaction with nuclear emulsion, depending on the incident quantum energy, electrons of different energies are produced; these lead to a variable distribution of silver centres. This in turn leads to the formation of different sizes of silver aggregates depending on the developing conditions. Further, the granularity of the film will be very different depending upon the incident energy. For example, extremely low energy electrons develop only a few silver grains per unit area, whereas, a high energy electron develops a relatively larger number of grains. Usually total absorption of a quantum leads to island like clusters of grains due to the short curved paths of electrons in the emulsion. Eggert and Schopper (1938) showed that this leads to increase in granularity and grain size of the photographic emulsion.

By exposing conventional nuclear emulsion film dosimeters to soft x-rays of a few mR, a distribution of developed silver grains are artificially produced. When the film is further exposed promptly to fast neutrons, the proton recoil tracks are produced additionally. The x-ray developed grains and those forming the proton track are of different sizes, different mean diameters and granularity. It therefore became clear that the optical quality of the track and hence contrast, may be controlled by the application of a suitable exposure using a defined quality of x-rays.

### Experimental Procedures

After the films were exposed to x-ray, there was a gap of about four days before they were exposed to neutrons and then they were promptly processed. For films exposed to neutrons first, the delay period was two days for x-ray exposures and then processing was carried out after a maximum storage time of one day.

Generally both categories of x-rayed films, before or after neutron exposure, were processed simultaneously for convenience and to maintain processing conditions constant.

Generally, there appeared to be a critical dose and quality of x-rays at which the distribution of developed grains due to x-rays did not interfere with the recognition of normal proton tracks, consistent with providing a good contrast as seen through the microscope. If the x-ray exposure was increased beyond a certain value, the well known effect of train distribution, due to fogging, interferes with proton track recognition, whether the proton track was short or long. In order to investigate these aspects, the following experiments were conducted using x-rays of 10, 15 and 25 kV. The 10 kV was chosen due to its ready availability for providing a source of low energy electrons in the emulsion; 25 kV was chosen for comparison. The following experiments were conducted. (1) Determination of optimum time/temperature combination for development and visual recognition (quality) of developed grain distribution, (2) the effect of pre or post x-ray exposure on sensitivity and quality of tracks for 10 kV x-ray beam, (3) The effect of pre or post x-ray exposure on sensitivity and quality of tracks for 25 kV x-ray beam, (4) Distribution of tracks at 10 kV for different post x-ray exposures, (5) Distribution of tracks at 15 kV for different post x-ray exposures (6) Distribution of tracks at 25 kV for different post x-ray exposures, (7) Track distribution as a function of optical density and quality of radiation, (8) Optical density at 10 kV as a function of x-ray exposures, (9) Calibration of NTA Type "A" film with and without pre x-ray exposures using 10 kV x-ray beam, (10) Photomicrographs of the "Contrast Effect".

### Results and Discussion

Experiment one showed that background grain counts in unexposed neutron films developed for 12 minutes at 64°F provided clear and good contrast of developed grains in 150 mm<sup>2</sup> area. The other temperature and time combinations gave rise to haziness and lack of sharpness of grain distribution. Hence in further experiments films were developed at 64°F for 12 minutes and fixed for 24 minutes.

It is observed that the "Contrast Effect" is seen in neutron film dosimeters that are exposed to x-rays either before or after neutron exposure. The visual effects are slightly different in each case. Therefore, in order to investigate this aspect, a set of three films were given 250 mRem of neutron exposure from a Pu:Be source and then subjected to different post x-ray exposures of 10 kV x-ray quality. Another set was also exposed to x-rays of the same quality using the same geometry and then to the same magnitude of neutron exposure. The results of the actual number of proton tracks mm<sup>-2</sup> in these sets of films as a function of x-ray exposures for a constant neutron exposure of 250 mRem are shown in Figure 1. A 15% enhancement of sensitivity is quite clearly seen from the curve of pre x-ray exposures. It should be pointed out that both sets of films were processed simultaneously. The differences in trend arise mainly due to varying degrees of fading of proton tracks in post and pre x-rayed neutron films. It should be recalled that after neutron exposure films wait for about four days before processing in the case of post x-ray exposure and in the other case films were processed promptly after the neutron exposure. The optical quality of tracks in post x-rayed neutron films were found to be good for exposures up to about 50 mR and in pre x-ray films to about 75 mR.

The results of experiment 3, 4, 5, 6 & 7 showed that the optical quality of tracks were not comparable to that based on 10 kV x-ray exposures although the enhancement effect (approx. 15%) was of the same magnitude. Figure 2 shows the result of 10 kV x-ray measurements. It is well known that increased addition of gamma or x radiation on neutron film reduces the number of observable tracks, Beeker (1963), Carallini and Busholi (1967). The present measurements revealed that for 50% reduction of track density 5 R of gamma (Ra-226)

and 550 mR of 250 kV (HVT 2.8 mm cu) at 1400 mRem of fast neutron dose from Pu Be source, are required.

The results from experiment 8 showed that the optical density of 10 kV x-ray exposed films did not vary up to about 150 mR. It was noticed however the microscopic distribution of grains superimposed on recoil proton tracks is found to be completely different. This results in different visual effects in a microscope ranging from clear recognition to blurring of tracks.

Figure 3 referring to experiment 9, shows the results of track density measurements for the purposes of calibration with and without pre x-ray exposure. As experimentally determined earlier, one set of neutron films were pre x-rayed using 10 kV and 30 mR exposure with an accuracy of better than 1%. These films were promptly calibrated to neutron exposures from a Pu:Be source and the proton track density distribution was compared with those neutron films of another set normally calibrated without any pre or post x-ray exposure. These observations were made by scanner A and confirmed by B. It is quite clear from this figure that the sensitivity of neutron films increases by about 15% over the useful range from 50 mRem to 5 Rem.

Experiment 10 revealed the variation in the size of grains due to pre x-ray exposure and proton tracks as seen in the photomicrographs.

#### Conclusions

Experimental measurement of quality and density of tracks with varying x-ray qualities of 10, 15 and 25 kV and pre or post x-ray exposures from about 5 to 100 mR on Kodak Type "A" neutron monitoring film promptly exposed and processed showed clearly identifiable contrast effect in the quality of tracks as seen in a microscope and also marked improvement in detection sensitivity (15%). Soft x-rays from a 10 kV spectrum and an exposure of 30 mR are found to be suitable to realize this effect in practice. The extension of this technique, with reference to other low energy neutron sources producing short proton tracks, employing film dosimeters for monitoring purposes appears thus feasible.

#### References

1. Cross, W.G. and Tomasino, L.; Radiation Effects, 5, 85 (1970).
2. Fleischer, R.L., Price, P.B., and Walker, P.M.; Science, 149, 383 (1965).
3. Eggert, J. and Schopper, E.; Wiss Photogr., 47, 221 (1938).
4. Becker, K.; Atomkernenergie 8, 74, (1963).
5. Cavallini, A. and Buaholi, G.; Radiation Measurements ENEA, 293, (1967).

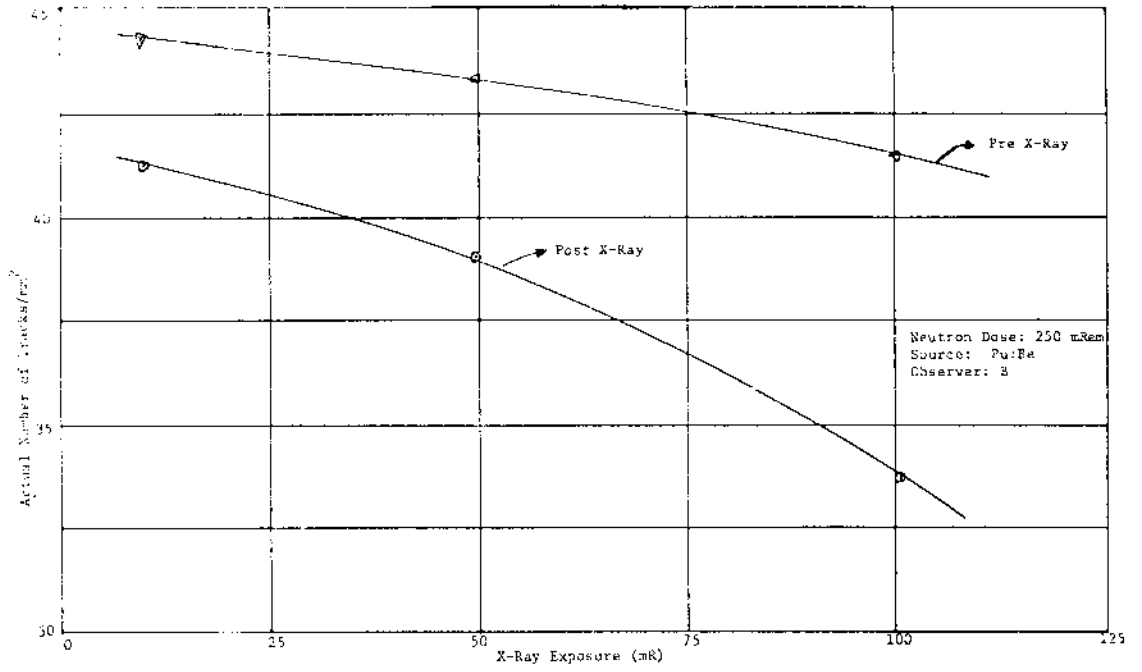


FIG 1 Effect of 10 KV X-Ray on Track Density in  $^{252}\text{Cf}$  Film Due to Pre or Post X-Raying

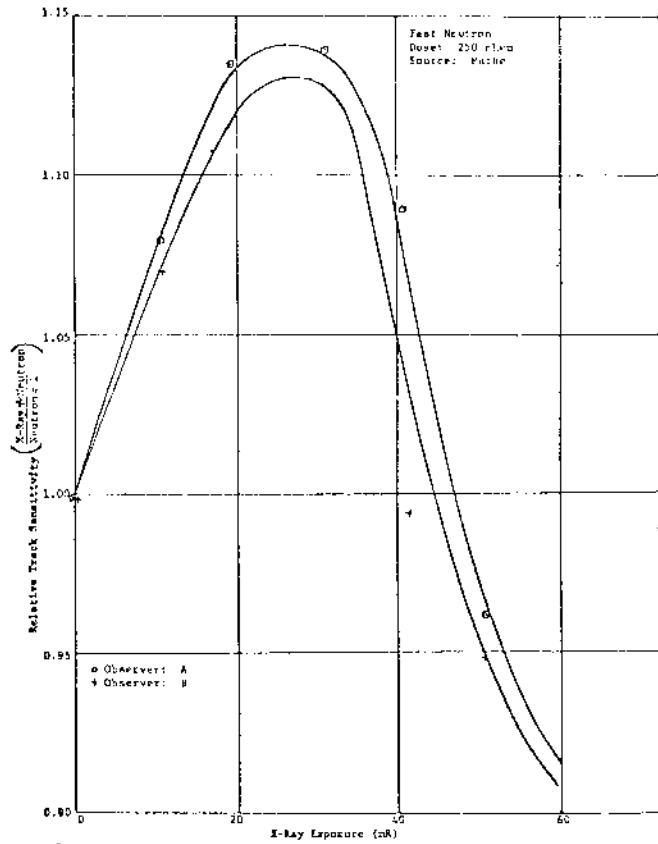


FIG 2 Track Distribution as a function of 10 KV x-ray exposure

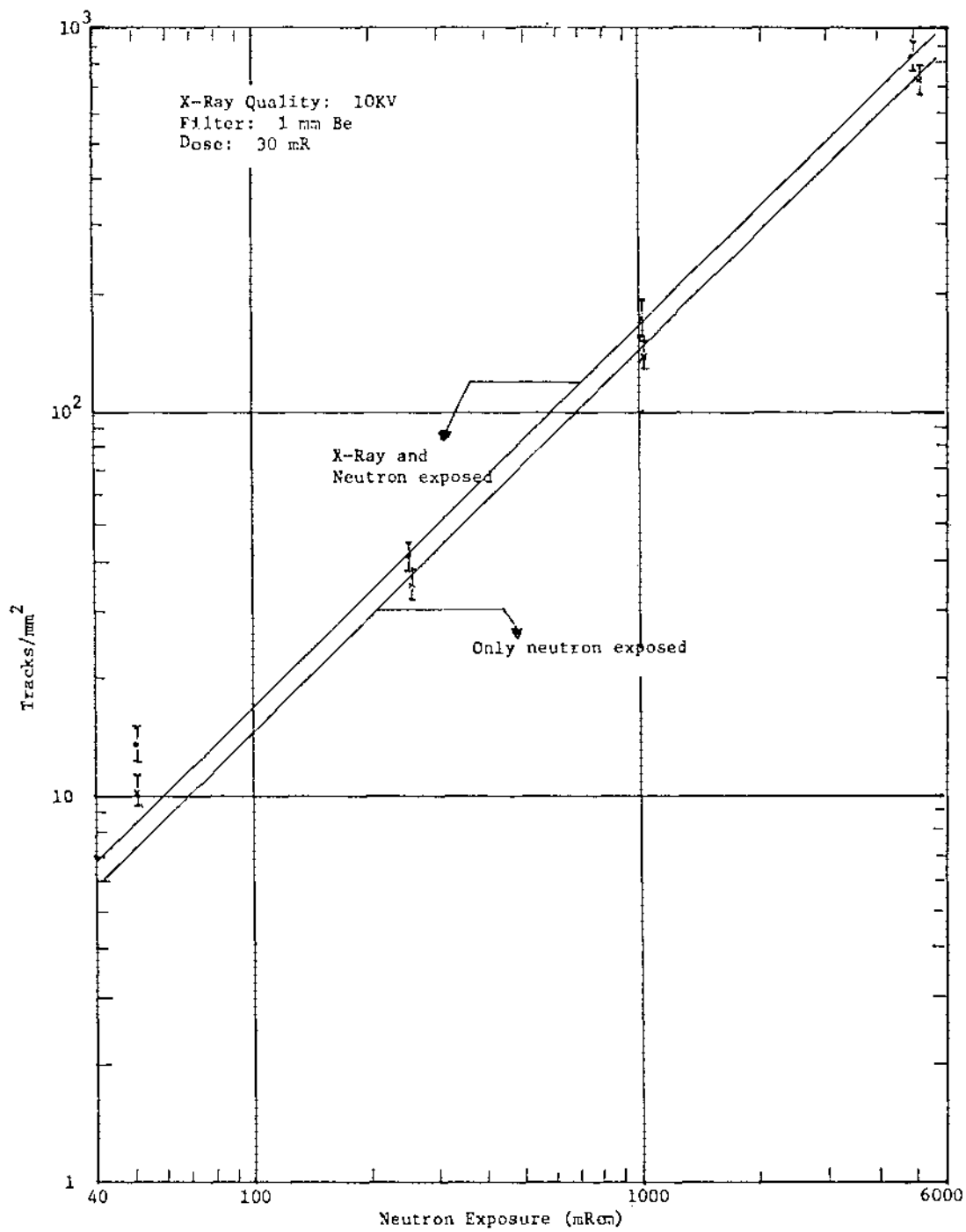


Fig. 3 Pu-Be Calibration Curve of NTA Type "A" Film with and without pre exposure of 10 KV Soft X Radiation



A THERMOLUMINESCENT PERSONAL DOSEMETER COMPATIBLE WITH AUTOMATIC  
PROCESSING AND THE CENTRAL RECORDING OF DOSE HISTORIES

K.B. Shaw, T.O. Marshall and P.N. Casbolt

National Radiological Protection Board,  
Physics Department, Harwell, Didcot,  
Berkshire, England.

Abstract

Factors influencing the choice of a thermoluminescent dosimeter are discussed. The dosimeter must be economic to produce, process and despatch: it must be convenient to the user and provide reliable dose information. The dosimeter must be compatible with automatic processing and the central recording of dose histories. A prototype dosimeter has been designed.

The dosimeter is required to measure separately low-penetrating and penetrating radiation doses and a filter system has been designed for this purpose. The low-penetrating and penetrating dose concepts are discussed together with the problems associated with achieving this requirement. The energy response data influencing the choice of filter system are given.

Introduction

In the United Kingdom over one million personal dosimeters are processed annually. The film badge is the most widely used dosimeter although thermoluminescent dosimeters are preferred for extremity monitoring<sup>1</sup>.

The present film badge service operated by the National Radiological Protection Board relies largely upon manual operations and uses little automatic equipment. The financial and technical advantages of an automatic thermoluminescent dosimeter system and an automatic film badge system have been considered and a decision made to develop for the future an automatic TLD system.

This paper deals with the factors which influenced the design of a thermoluminescent dosimeter or badge, suitable for use in a large scale personal monitoring service (up to  $10^6$  dosimeters evaluated per year) that uses the postal services for the despatch of dosimeters to the user.

It has been suggested, in ICRP Publication 12, that a basic two-element dosimeter is satisfactory in almost all practical cases<sup>2</sup>, particularly since the majority of doses are hardly greater than background: e.g. in a recent survey of 10,000 film badges issued by the NRPB, 96% recorded doses less than 40 mrad and only 16 results were over 500 millirads. The two-element dosimeter should provide information on low-penetrating and penetrating

radiations. Additional dosimeters can be worn if further information is required.

The large throughput required by the NRPB dosimeter service necessitates a dosimeter compatible with automatic processing and the recording of dose histories by computer.

The design of the dosimeter or badge is critical to the whole system and largely dictates the eventual cost and complexity of the dosimeter service.

Since the two-element dosimeter is considered satisfactory for the vast majority of users, the complete system is built around this conceptual design. The system is assumed to be largely automatic with a capacity of up to  $10^6$  dosimeters per year. The economic arguments are governed by the cost of the existing film badge service.

#### Design of a Thermoluminescent Dosimeter

The thermoluminescent dosimeter envisaged by the authors consists of the following major components:

- (a) Thermoluminescent materials.
- (b) Plaque to contain thermoluminescent materials and to carry number identification (could be entirely thermoluminescent material).
- (c) Plastic cover to exclude light, dust, grease, etc.
- (d) Outer plastic holder to contain required filters and to hold the plaque.

#### Thermoluminescent Material

The material chosen for the basic thermoluminescent dosimeter is  ${}^7\text{LiF}$  since its characteristics have been well investigated and supplies are readily available.  ${}^7\text{LiF}$  is suitably neutron insensitive and although the energy response is not ideal it is acceptable. There are, at present, three forms of  ${}^7\text{LiF}$  readily available in the U.K., viz. as powder, in teflon<sup>3</sup> and extruded into chips<sup>4</sup>. At a throughput of  $10^6$  dosimeters per year, the handling problems associated with powder make this form least attractive. The 3 mm x 3 mm chips with thickness from 0.25 mm to 0.90 mm are used extensively in the U.S.A. The teflon disc or foil forms are available in a range of sizes, however the standard is 12.9 mm diameter x 0.4 mm thick: the ratio of thermoluminescent phosphor to teflon content can be varied. Either the teflon disc or the chip form are suitable for the basic two-element dosimeter. However in the case of the chips two thicknesses are required to achieve the desired energy response<sup>5</sup>. The 0.4 mm teflon disc has adequate energy response as is shown below. Both forms of dosimeter can be re-used. The consistency of the discs over repeated cycles of use has been shown satisfactory<sup>6</sup> and the chips have been tested and proved acceptable<sup>5</sup>.

At present the ease of mounting and general robustness of the teflon discs together with lower cost give them a significant advantage over the chips. A calibration check as part of the processing sequence can indicate any change in sensitivity and serve as a calibration for the next cycle, this is particularly easy with automatic and computerised operations and has some cost advantages in the initial selection of the thermoluminescent material.

### Plaque

The plaque is required to contain the thermoluminescent materials and to carry the identification number. A method of insuring correct orientation of the plaque in the holder is required.

The material considered for the plaque has to be capable of withstanding 300°C annealing without distortion. It is generally accepted that a high temperature anneal outside the read cycle is not generally required. Any high dosed badges can be removed and annealed or alternatively discarded. Nevertheless it is considered wise to retain the option to anneal. The cost of retaining this option is small compared to the cost of having to change from a low temperature resistant plaque should this be proved necessary. Aluminium or a high temperature resistant plastic are considered suitable materials.

### Plastic cover

A plastic cover is required to exclude light, dust, grease, etc., from the thermoluminescent materials. This cover must be thin in order to preserve adequate  $\beta$ -ray response but must act as an adequate barrier. A label containing wearers name or number, issue period and employers code will need to be adhered to the plastic, prior to despatch to the customer. The label is considered an aid to the user in ensuring that the correct dosimeter is worn. A carbon loaded plastic, 11 mg/cm<sup>2</sup> thick, has been successfully used in operational tests to cover the teflon discs.

### Plastic holder

The holder contains a thick filter for the penetrating radiation measurement and an open window area for the low-penetrating measurement and will hold the plaque. The system is designed so that the holder is kept by the dosimeter wearer who exchanges his plaque at regular intervals. This ensures that postal costs are kept to a minimum. A hinged holder attached by a safety pin would be suitable. The maximum thickness of the holder will be determined by the thick filter requirements.

For low cost and ease of manufacture an injection molded plastic holder is suggested for the basic dosimeter. If thermal neutron measurement, low background or other measurements were required, then a holder capable of taking an additional special plaque could be used, appropriate filters could be incorporated in this holder and the design would ensure that appropriate plaques could only be loaded in the correct orientation.

### Low-penetrating and Penetrating Radiations

For a basic two-element dosimeter it is important to decide what should be considered as low-penetrating and penetrating radiations. In order to do this it is sufficient to consider the permissible doses and the depths of the basal layer of the epidermis, the lens of the eye and the gonads.

The maximum permissible dose<sup>7</sup> for skin is 30 rem/year and the depth of the basal layer of the epidermis is considered at a depth of 7 mg/cm<sup>2</sup>. The level for the lens of the eye is 15 rem/year, considered at a depth of 300 mg/cm<sup>2</sup>. The maximum permissible level for the gonads is 5 rems/year and they are assumed to be at a minimum depth of 1000 mg/cm<sup>2</sup> (higher depth values have been

used, e.g. 2 cm<sup>8</sup>).

To differentiate between low-penetrating and penetrating radiation we must consider the attenuation of the radiation in the tissue covering the organ in question and its permissible level. If, for example, a particular radiation is attenuated more than a factor of 2 in penetrating to the lens of the eye, then this will become a surface dose since, in controlling the skin dose one ensures that the lens of the eye are not over exposed. A similar argument can be used for the gonads except that the attenuation factor should, in this case, be 6.

Figure 1<sup>9</sup> shows the attenuation of photons in tissue with energies in the range 7.3 keV to 35 keV in terms of percentage depth dose. The vertical lines indicate the depth of the lens of the eye and the gonads and the horizontal lines indicate 50% transmission corresponding to an attenuation factor of 2, and 17% transmission corresponding to an attenuation factor of 6. It can be seen that the demarcation energy is 12.5 keV and that above this energy photons must be considered as penetrating radiations and, below this energy, as low-penetrating radiations.

Figure 2<sup>10</sup> shows percentage depth dose data for  $\beta$ -ray spectra in the range of maximum energies from 0.067 to 2.27 MeV. It can be seen that these radiations can be treated as low-penetrating and this can be said of most  $\beta$ -rays likely to be met in practice.

It would seem that the filter above the element to measure penetrating radiation should simulate the tissue ideally above the gonads, i.e. minimum 1000 mg/cm<sup>2</sup>. This could either be about 3 mm of aluminium or the appropriate thickness of plastic as far as  $\beta$ -rays are concerned but aluminium would be less suitable in the photon case. The optimum filter would be about 1000 mg/cm<sup>2</sup> of tissue equivalent material such as plastic but in practice this would result in a very thick dosimeter. The effect of using a thinner filter would be to over-estimate the penetrating dose for low energy radiation.

As far as the measurement of the low-penetrating radiation is concerned, the thermoluminescent elements themselves are generally too thick and the minimum window we are able to place over this element will be governed by considerations of robustness of the badge and opacity to light. Ideally, the window should be about 7 mg/cm<sup>2</sup> and the elements a few mg/cm<sup>2</sup> thick.

#### Energy Response Data for the Disc Form

The X-ray response of 0.4 mm thick <sup>7</sup>LiF in teflon discs under various filters is given in Figure 3. The data has been plotted as response relative to <sup>226</sup>Ra gamma rays. Plastic and aluminium filters were used up to 550 mg/cm<sup>2</sup>. Below 100 keV the disc response rises to a peak around 30 keV. The effect of the filters is to reduce this peak but increase attenuation below 30 keV. The  $\beta$ -ray response of the discs is given in Figure 4. The sources used were Tungsten-185, Thallium-204, Yttrium-91, Strontium-90 + Yttrium-90 and Ruthenium-106 + Rhodium-106. The perspex and aluminium filters have similar  $\beta$ -ray absorption and the 550 mg/cm<sup>2</sup> filter is sufficient to stop  $\beta$ -rays below 2 MeV.

The 470 mg/cm<sup>2</sup> was the highest surface density plastic filter used, further work is underway with filters up to 1000 mg/cm<sup>2</sup>. For the 170 mg/cm<sup>2</sup> plastic, the response to 12.5 keV photons is 70% of that due to <sup>226</sup>Ra. It seems probable that a 600 mg/cm<sup>2</sup> plastic is practicable and further measurements are in hand.

The 0.4 mm thick teflon discs under about 10 mg/cm<sup>2</sup> and 600 mg/cm<sup>2</sup> filtration should meet the requirements for the measurement of low-penetrating and penetrating radiations. Additional work is in hand on the effect of orientation with the discs mounted in a plaque contained in a holder with the appropriate filters.

#### Prototype Dosemeter

After careful consideration of all the published information on thermoluminescent dosimeters and with regard to the specification and results given in this paper, a prototype badge has been designed.

The economic considerations have led to a basic two-element badge with <sup>7</sup>LiF in teflon discs as the thermoluminescent material. The plaque, shown in figure 5, securely retaining the materials is aluminium and an 8 x 4 hole matrix is used for identification.

The plaque would be sealed in a light-tight and dust/grease proof plastic pouch which would also provide sufficient filtration for the low-penetrating radiations. An adhesive label with name of user and issue period would be attached to the cover, away from the teflon discs.

A plastic holder containing an open window area and a 600 mg/cm<sup>2</sup> filtered area would be retained by the customer. Plaques would be exchanged and the holder designed such that the label was visible through the holder. The plaque and holder can be made so that the correct plaque orientation can easily be achieved.

#### Conclusions

1. A basic two-element dosimeter to measure low-penetrating and penetrating radiations is adequate for the majority of practical applications.
2. Low-penetrating radiation is defined as photons less than 12.5 keV and most  $\beta$ -rays met in practice i.e. below about 3.5 MeV. Penetrating radiations are defined as photons above 12.5 keV.
3. <sup>7</sup>LiF in teflon discs are adequate for the dosimeter and are the most economic of presently available forms in the U.K.
4. A thermoluminescent dosimeter has been designed to be
  - (a) Produced economically
  - (b) Convenient to the user
  - (c) Processed economically
  - (d) Reliable
  - (e) Suitable for automatic processing and computerised record keeping.

#### Acknowledgements

The authors wish to thank Mr. H.J. Dunster for suggestions and encouragement. Mr. J.A. Dennis has provided advice and guidance on the project. The Sira Institute have provided most useful stimulation and their involvement is acknowledged. Messrs. R.J. Pattison, K. Cliff and A.G. Dyke and Mrs. P. Smith have provided valuable contributions to the project.

### References

1. T.F. Johns. Use of Energy-independent Dosimeters for Measuring Partial and Whole-body Doses. ENEA Symposium on Radiation Dose Measurements, Stockholm 1967.
2. ICRP Publication 12. General Principles of Monitoring for Radiation Protection of Workers 1968.
3. Teledyne Isotopes, Westwood, New Jersey, USA.
4. The Harshaw Chemical Company, Solon, Ohio, USA.
5. A.R. Jones. A Personal Dosimeter System Based on LiF Thermoluminescent Dosimeters. Proc. 3rd. Int. Conf. on Luminescence Dosimetry. Risø 1971.
6. T.O. Marshall, K.B. Shaw, E.W. Mason. The Consistency of the Dosimetric Properties of  $^7\text{LiF}$  in Teflon Discs over Repeated Cycles of Use. Proc. 3rd. Int. Conf. on Luminescence Dosimetry. Risø 1971.
7. ICRP Publication 9. Recommendations of the International Commission on Radiological Protection 1966.
8. A.R. Jones. Measurement of the Dose Absorbed in Various Organs as a Function of the External Gamma Ray Exposure. AECL - 2240, 1964.
9. British Journal of Radiology. Supplement 10. Depth Dose Tables for Use in Radiotherapy, 1961.
10. T.M. Francis and R. Seymour. Dose Rates and Depth-dose distributions for Beta Particles emitted by commercially available  $^{90}\text{Sr}/^{90}\text{Y}$ ,  $^{204}\text{Tl}$ ,  $^{147}\text{Pm}$  and  $^{63}\text{Ni}$  sources. NRPB-R4, October 1972.

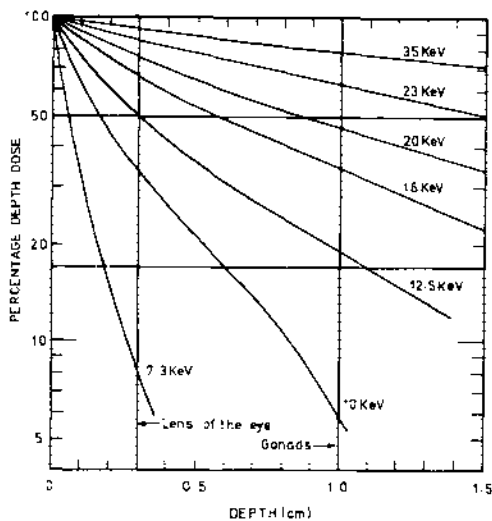


Figure 1 PHOTON ABSORPTION IN TISSUE

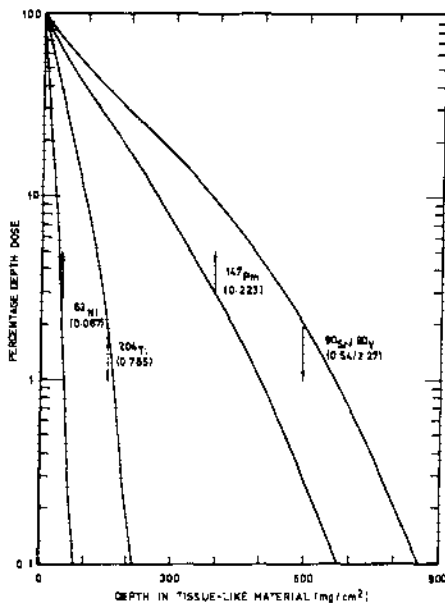


Figure 2 PARTICLES FROM PLANE SOURCES VIRTUALLY IN CONTACT WITH THIS MATERIAL [THE MAXIMUM ENERGIES OF BETA PARTICLES IN MeV ARE SHOWN IN PARENTHESES.]

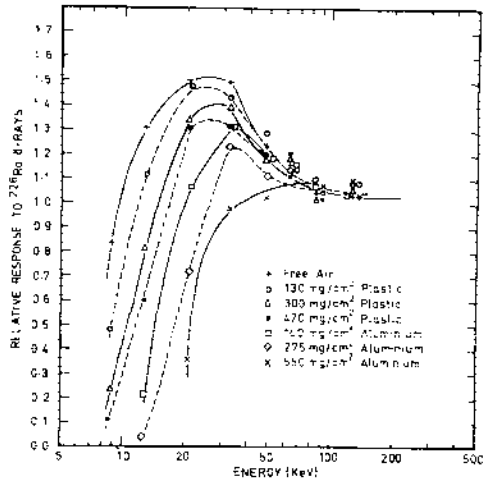
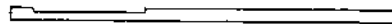
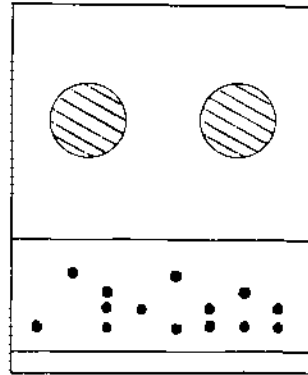


Figure 3. X-RAY RESPONSE OF 0.4mm THICK  ${}^7\text{LiF}$  IN TEFLON DISCS UNDER VARIOUS FILTERS.



TWO TEFLON/LITHIUM FLUORIDE DISCS IN AN ALUMINIUM ALLOY HOLDER WITH A 7 DIGIT BCD CODED NUMBER AND ROW EVEN PARITY HOLE. (INSERT No. 1872935).

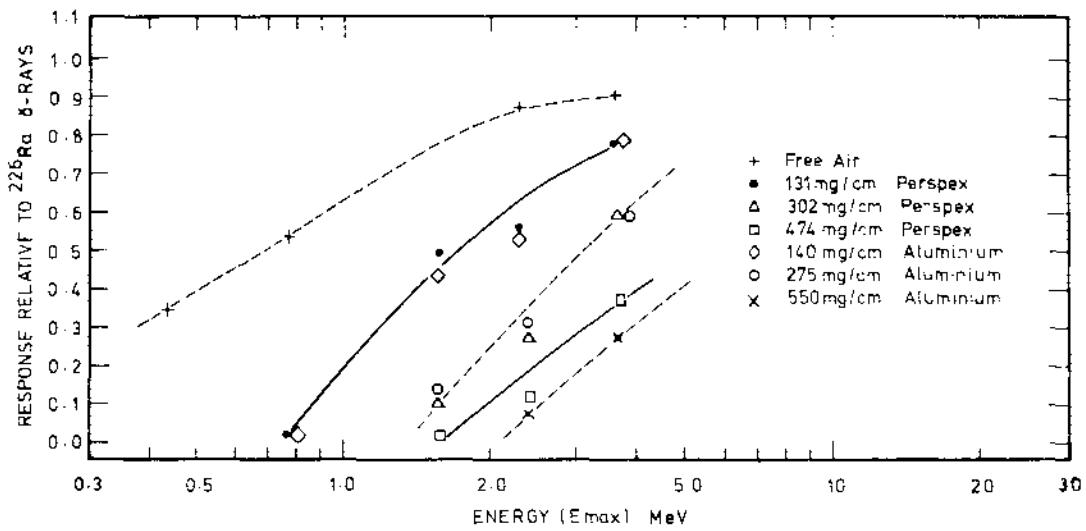


Figure 4. B-RAY RESPONSE OF 0.4mm THICK  ${}^7\text{LiF}$  IN TEFLON DISCS.

## A UNIVERSAL AUTOMATIC TLD-READER FOR LARGE-SCALE RADIATION DOSIMETRY

H.W. Julius, C.W. Verhoef and F. Busscher  
Organization for Health Research TNO  
Radiological Service Unit TNO  
Arnhem, The Netherlands

### Abstract

A universal automatic TLD-reader has been developed that can accept most of the present and probably many future types of solid TL-detectors. Perfect temperature contact, low background signal, high sensitivity, short cycle-times (about 18 sec., sample changing included) and good reproducibility are secured by heating the detectors with jets of hot nitrogen gas. A special manipulating device enables the reader to accept TLD's as offered by any sample changer (e.g. rotating disc or conveyor belt), making the instrument applicable in large scale personnel monitoring, environmental control, radiation therapy etc. The automatic selection of dosimeters with uniform sensitivity is one of the features of this instrument.

### Introduction

Although the daily practice of handling TLD in routine radiation dosimetry is not yet completely without problems, the advantages of this technique are clear enough. Especially in personnel monitoring TLD has in many cases proved to be more reliable than film dosimetry. For this and several other reasons, such as the need for automation, many institutes and dosimetry services are considering the possibility of using TLD in radiation protection, or have adopted this technique already. Unfortunately, most of the TLD-readers now commercially available are intended for manual operation. The few automatic systems developed so far suffer from the drawback that they are completely closed systems, since they are adapted to special types of personnel dosimeters and hence are not necessarily compatible with other types of TL-detectors.

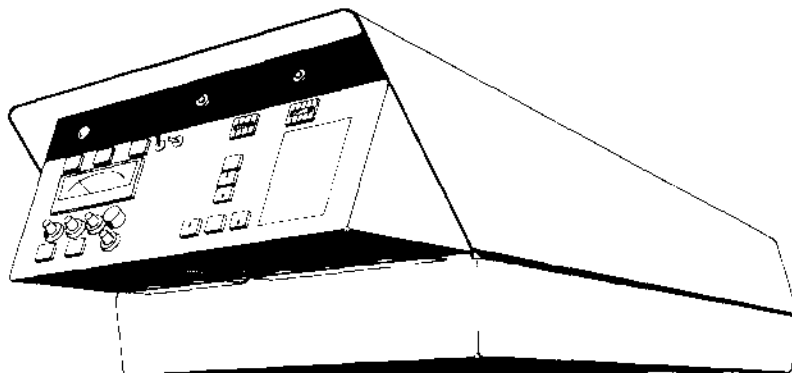
TNO, also anticipating the use of TLD in personnel monitoring (for about 10,000 people on a bi-weekly base) therefore developed an automatic and versatile TLD-reader that leaves complete freedom in the design of a future, automatically processable, dosimetry system. The instrument accepts most of the present (and probably many future) types of solid TL-detectors.

After an experimental set-up had been tested in the laboratory of the Radiological Service Unit TNO with satisfactory results, a more sophisticated prototype (Fig.1) has recently been developed



in cooperation with one of the other institutes\*) of the Organization for Scientific Research TNO. This instrument might be the basis for serial production by some commercial manufacturer.

Fig.1.  
Automatic TLD-  
reader (prototype).



The reader

Thermo-optical part. We use hot nitrogen gas as the heating agent - as has previously been described by Petrock and Jones<sup>1</sup> and is now being used by some others<sup>2</sup> - because of the well known advantages of this method: perfect temperature contact between heating medium and detector independent of its shape, low background signal, constant starting conditions and short cycle times (200 dosimeters can be read per hour).

The TL-detector, fixed on top of a thin suction needle (see below) is exposed to three hot-gas jets which are placed under angles of  $120^\circ$  (see Fig.2). The heating cavity has been designed to accept TLD's up to a diameter of 13 mm (teflondiscs). As a consequence the nitrogen consumption is relatively high (4 to 5 liter  $N_2$  per minute per jet, i.e.  $\pm 1 \phi$  per reading). The gas temperature can be varied and stabilized within a wide range to meet the requirements for different detector materials.

Except for the three gas in- and outlets, the heating cavity is completely closed during readout, so that no spurious light can interfere with the TL-signal and the gas be kept free from impurities.

The TL-light is measured with a cooled and temperature stabilized PM-tube. An ellipsoidal mirror (Fig.3) is used to avoid thermal contact between heating cavity and PM-housing. In the lightpass a diaphragm is placed, to shield light emitted by other sources than the TLD itself.

TLD-manipulator. The problem of putting a TL-detector from any holder or sample changer into the heating cavity has been solved by developing a special manipulating device. This TLD-manipulator may roughly be described as follows (Fig.3): In a cubical piece of metal a rotatable drum is fitted. The drum has a radial bore in which a hollow piston can be moved up and down. The bottom of the piston is closed, the other end is provided with a removable cap in the centre of which a hollow needle is mounted. When the piston is retracted within the drum, the latter can be rotated freely. With the drum in vertical position the needle may protrude either through a hole in the top or through a hole in the bottom of the

\*) Instrumentum TNO, Delft, The Netherlands

cube. If, in the latter position, the piston is evacuated through a duct in the cube, the suction needle can pick up a TLD. The TLD may now be brought into the heating cavity by successively: retracting the piston into the drum, rotating the drum over  $180^\circ$ , and moving the piston outwards again. After readout the TLD can be brought back into the position it came from but, if desired, the TLD may be dropped at another site by using a third opening ("emergency exit") in the cube, at which the drum can be stopped. This feature may be used if the detector has to be removed from the series for some reason (see the description of the electronics).

The construction of the TLD-manipulator indeed leaves the user complete freedom as to the way of offering a TLD to the reader. For this purpose any simple sample changer will do, such as a rotating disc or a conveyor belt. This means that there are essentially no restrictions in the way a TLD-batch is designed, except in that the detectors must be presented freely to be picked up after the holder was opened.

Electronics. The TL-signal is handled in a conventional way. The PM current is digitalized by means of a current-to-frequency converter (for which in the prototype a set-up as proposed by Shapiro<sup>3</sup> is used). Light output is integrated during the readout time. In principle a dose-range of about seven decades can be covered without range-switching. To get sufficient accuracy at low doses and to avoid saturation of the converter, the range will in practice be limited to six decades by setting the sensitivity, for example from 5 mR to 5000 R.

Because the temperature to time relationship is not linear - the gas temperature being constant - the response curve (Fig.4) has no direct scientific significance, but still it can be used as an indication for proper working of the instrument. The readout process can be followed on a logarithmic rate meter, or compared with a standard curve as stored in a computer memory.

The reader can be used as well in automatic as in semi-automatic (only cycle) and in manual mode. The instrument is

Fig.2. TLD (teflon disc) in centre of heating cavity, exposed to three jets of hot nitrogen gas.

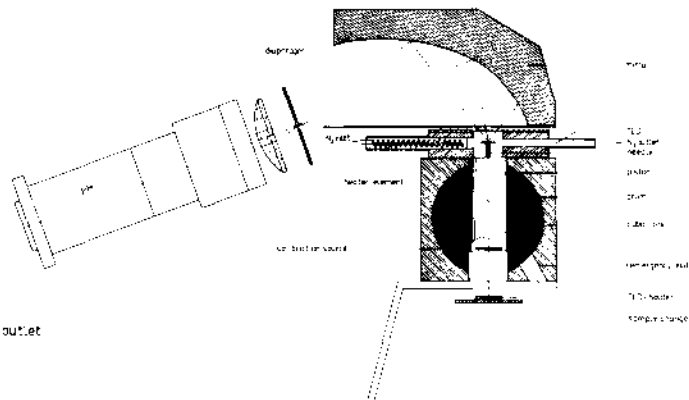
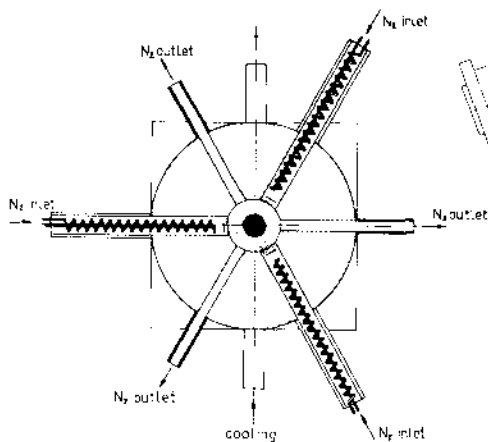


Fig.3. Diagram of the principle parts of the reader.

designed as a fail-safe system. This is of great importance in TL-dosimetry, where the dose-information is essentially lost by the readout procedure. Therefore checks are continuously performed for nitrogen flow, gas temperature, false release of the TLD from the needle, etc. Before each cycle the optical and electronic parts can be checked by means of a radioactive ( $^{14}\text{C}$ ) lightsource fixed at the bottom of the piston.

The rear panel is provided with in- and output connectors, so that additional modules of conventional nuclear electronics and even a programmable processor (such as the PDP/8) for external control and data handling can be used. Nevertheless, to make the system as selfsupporting as possible, several features have been built into the prototype, such as timers, display, sample changer control, etc.

The reader can be used for automatic selection of dosimeters of uniform sensitivity out of a batch of equally exposed TLD's, by using a lower- and an upper count (dose-)level. If these levels are set, for example, according to  $\bar{x} - 2\sigma$  and  $\bar{x} + 2\sigma$  respectively (assuming that the average response  $\bar{x}$  is known), then dosimeters showing a sensitivity within the chosen range are repositioned (thus collected), while detectors responding outside this range are ejected through the above mentioned "emergency exit" (thus separated from the others).

During normal automatic readout, the lower level can, if desired, be used to cause a prolonged, adjustable, heating time (post-read anneal) after a relatively high dose has been measured. In this mode, doses above the upper limit will cause the TLD to leave the reader by the "emergency exit", to be annealed in an oven.

### Experiments

Up to now, experiments have been carried out in some detail for LiF hot-pressed ribbons (Harshaw,  $1/8 \times 1/8 \times 0.035$  inch, 25 mg) and LiF teflon-discs (Teledyne Isotopes, 12.7 mm dia., 30 mg),

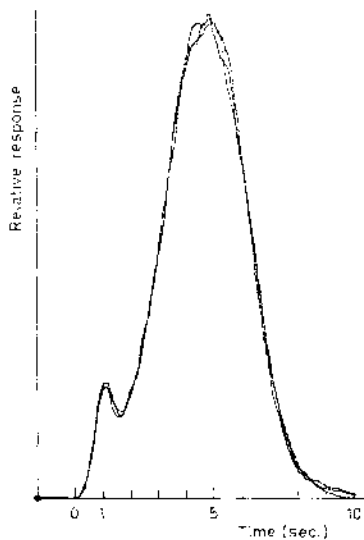


Fig.4. Successively recorded glow curves of 3 LiF-ribbons (Harshaw, 25 mg) exposed to 0,5 R  $^{60}\text{Co}$  gamma radiation.

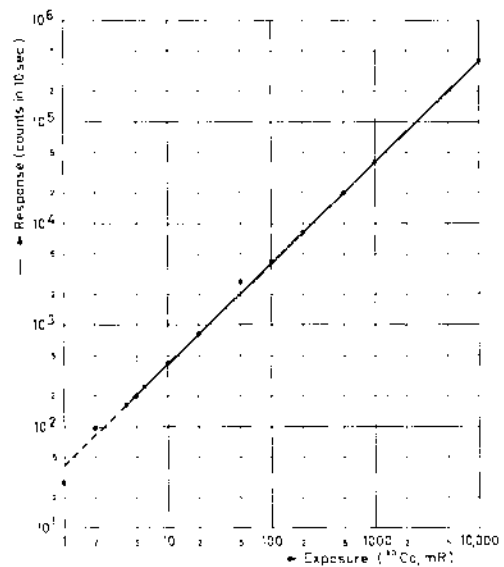


Fig.5. Dose-response curve of hot-pressed ribbons (Harshaw TLD-100, 25 mg) read in automatic reader.

because these detector materials are possibly used in a future TLD-batch. Fig. 5 shows the dose-response curve for the Harshaw, LiF-chips. The data points represent the averages of eight TLD's. For this experiment - in which non-selected TLD's from one manufacturing batch were used - standard deviations appeared to be 9% at 5 mR and 2-4% above 20 mR.

The sensitivity of the instrument would make it possible to detect doses far below 1 mR, but unfortunately we did not succeed in finding the proper annealing procedure to make the signal of non-irradiated TLD's sufficiently low and constant. This was especially true for the 12.7 mm diameter teflon discs which, moreover, show substantial light-sensitivity. These phenomena were responsible for the fact that, for this material, the detection threshold was as high as about 5 mR, although these dosimeters were found to be roughly twice as sensitive as the hot-pressed ribbons.

The dosimeters are almost completely emptied, as can be proved by a second readout, which gives a response corresponding to less than 0.1% of the original dose. If desired, this remaining signal may be used to prove the significance of an unexpected high dose.

### Conclusion

Our experience with the experimental version of the TLD-reader as described above, proved that the apparatus satisfies our requirements as to fast evaluation of different types of TLD's, although it was not supplied with the features as available in the more elaborate prototype that has just been finished. This universal automatic TLD-reader being available now, the way is open to us (as it is, in principle, to anyone) to design an automatically processable personnel TL-dosimeter for large-scale radiation protection measurements.

Irrespective of the fact whether this is a good thing or not, in practice most institutions prefer to have their own input in the dosimetry system used, as far as philosophy and choice of the applied TL-materials are concerned. Up to now this was impossible without designing all parts of the equipment. We think that this universal instrument will help to overcome the hesitations to use TLD on a large scale and will make this attractive technique more a tool than a toy.

### References

- (1) Petrock, K.F., Jones, D.E., "Hot nitrogen gas for heating thermoluminescent dosimeters", Proc. 2nd Int. Conf. on Luminescence Dosimetry, Gatlinburg, Tenn., Sept. 1968, USAEC Rep. CONF 680920 (1968) 652-669.
- (2) Bøtter-Jensen, L., "Read-out instrument for solid thermoluminescence Dosimeters, using hot nitrogen gas as the heating medium", Proc. IAEA Symp. on Advances in Physical and Biological Radiation Detectors, Vienna (1971) 113-124.
- (3) Shapiro, E.G., "Electronics for Automated TLD Reader System", Lawrence Liv. Lab., UCRL-50007-69-2, Hazards Control Progress Rep. No. 34 (1969) 20-29.

ANNEALING TLD'S EXPOSED TO HIGH  
LEVELS OF  
NEUTRON AND GAMMA RADIATION\*

G. W. R. Endres, F. N. Eichner  
and L. L. Nichols  
Battelle  
Pacific Northwest Laboratory  
Richland, Washington 99352

ABSTRACT

Experimental programs involving radiation dose measurements at high levels (greater than 100 rads) have led to problems in reuse of thermoluminescent dosimeters (TLDs). Accepted procedures for annealing the TLDs in many cases do not entirely eliminate all of the stored energy trapped in the crystal lattice during the high level exposures.

Several procedures for annealing  ${}^7\text{LiF}$ ,  ${}^6\text{LiF}$  and  $\text{CaF}_2:\text{Dy}$  were studied to determine which one would best restore the TLDs to the original background readings and sensitivity.

Results are given in numerical and graphical form to show that many of the dosimeters are restored to usefulness after long-term annealing in a nitrogen atmosphere. For  ${}^7\text{LiF}$  in a period of 16 hours at  $400^\circ\text{C}$  in a nitrogen fed furnace plus 2 hours at  $100^\circ\text{C}$  in air satisfactorily anneals dosimeters exposed to as much as 3500 rads from radium and  ${}^{252}\text{Cf}$  radiation sources. These dosimeters have the same sensitivity to a 500 mR calibration exposure as unused dosimeters from the same batch of LiF.

All dosimeters used were in the form of  $1/8 \times 1/8 \times 0.035$ " blocks, and all annealing was accomplished in vicor glass dishes. The dosimeters were placed through the standard anneal of 1 hour at  $400^\circ\text{C}$  plus 2 hours at  $100^\circ\text{C}$  to determine the completeness of the special long-term anneal.

INTRODUCTION

One of the problems encountered when using thermoluminescent dosimeters for very high-level dosimetry measurements is the annealing of the dosimeters to return them to their original sensitivity before exposure. In

particular,  ${}^7\text{LiF}$  and natural LiF used in reactor irradiations and burst exposures have a different sensitivity after readout and a standard anneal as prescribed for low-level exposures (less than 10 rads). This work was conducted in an effort to find annealing procedures which would restore the original sensitivity and properly empty the traps in TL materials exposed to radiation levels greater than 100 R.

In our previous experiments many times the dosimeters had to be discarded after the very high-level exposures. An increase in the use of LiF dosimeters at high levels has made it necessary to find a procedure for annealing which will allow the return of at least a portion of the dosimeter to usefulness.

THEORETICAL CONSIDERATIONS

Various heat treatments have been studied for routine handling of thermoluminescent (TL) materials. Preirradiation and postirradiation annealing were studied by Zimmerman<sup>1</sup> in great detail for LiF. The glow curve for LiF is a rather complicated function of not only temperature and time but also cooling rate. There are at least five peaks which represent energy states occupied by trapped electrons in the irradiated crystals. The relative populations of these energy states depend in a complicated manner on the annealing processes used to prepare the crystals before exposure to ionizing radiation.

For practical dosimetry, the annealing process should minimize the population of the low-level traps which tend to fade (or self-anneal) more rapidly at normal ambient temperatures than does the population of the higher level traps. The procedure must also preserve the sensitivity of the device to ionizing radiation so that a new calibration is not necessary each time a group of devices is exposed and read out.

For  ${}^7\text{LiF}$  (TLD-700) the plot of a typical glow curve is shown in Figure 1. Although there are a number of lower temperature peaks, the peak at about  $80^\circ\text{C}$  is the lowest peak that creates a problem in normal usage. The three peaks clustered around

200°C, numbers 2, 3 and 4, normally merge into a single peak after a proper annealing. This reduces the fade problem of peak number 2. A high temperature peak, peak number 5 which is not shown in Figure 1, is populated by electrons in deeper traps which may not be completely emptied during an anneal procedure.

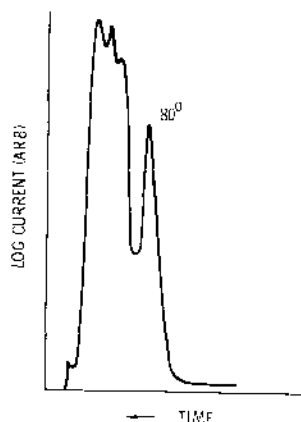


FIGURE 1. Glow Curve for  ${}^7\text{LiF}$

After much experimental effort by a number of researchers<sup>2,3,4</sup> an anneal procedure of 1 hour at 400°C followed by 2 hours at 100°C has become a standard at many laboratories using the TL materials for more or less routine work. This anneal procedure seems to produce the desired results as indicated above for most exposures below the 100 R level. Above 100 R the standard anneal may not properly empty the trapped electrons and restore the original sensitivity of the TL material to ionizing radiation, particularly if the exposure included neutron radiation.

#### EXPERIMENTAL PROCEDURES

A Harshaw Model 2000 thermoluminescence analyzer was used in our work to read out all of the TL materials studied in this program. Most of the materials were in the form of 0.32x0.32x0.089 cm blocks of either  ${}^7\text{LiF}$  or  ${}^6\text{LiF}$ . These blocks each contain about 22 mg of material. Both the  ${}^7\text{LiF}$  and  ${}^6\text{LiF}$  are sensitive to gamma and neutron radiation at least to a limited extent.

The normal readout of the blocks includes heating them with an approximately linear temperature ramp to about 250°C and holding that temperature for

a few seconds. For all readings in this study a total cycle of 30 seconds was used. Glow curves were recorded for some of the readout cycles but not for all. Typical glow curves are given in Figure 2.

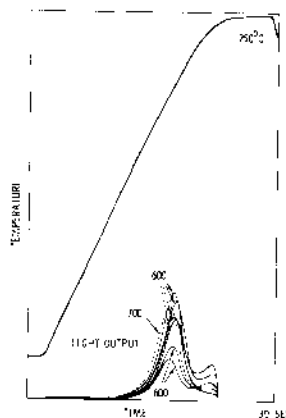


FIGURE 2  
Glow Curves and Temperature Ramp

All TLD-700 ( ${}^7\text{LiF}$ ) blocks were from the same batch of materials except those exposed to radiation from a burst reactor.

A group of  ${}^6\text{LiF}$  and  ${}^7\text{LiF}$  blocks which had been exposed to high levels of radiation from the Health Physics Research Reactor (HPRR) at Oak Ridge National Laboratory were exposed to 1 R gamma radiation after a standard anneal procedure of one hour at 400°C and 2 hours at 100°C. Many of the dosimeters yielded a higher reading than dosimeters not exposed to the high radiation levels. The differences were 10 percent or more in a number of cases. Thus, the standard anneal did not appear to adequately restore either the sensitivity or trap site structure or both. The same problem occurred after high level exposures to  ${}^{252}\text{Cf}$  and  ${}^{226}\text{Ra}$  sources.

After several time-temperature anneal cycles were tried a 16-hour period at 400°C followed by 100°C for 2 hours was determined to be a reasonable annealing procedure to study further. The same dosimeters exposed at the HPRR mentioned above were annealed for 16 hours at 400°C and 2 hours at 100°C. The 400°C furnace was flushed with  $\text{N}_2$  gas during the annealing. The  $\text{N}_2$  atmosphere was provided to minimize

any possible changes in sensitivity caused by the presence of hot oxygen.

Results of a comparison between the specially annealed dosimeters and a control group not exposed to high levels of radiation are shown in Table I.

Table I

	<u>TLD-700 <sup>7</sup>LiF</u>	High Level Sensitivity After Exposure & 16-Hr Anneal at 400°C
	Exposure Sensitivity of Control Group	
1 R	120.8 ± 1.8	118.8 ± 2.5
5 R	523.3 ± 15.5	532.0 ± 5.7
10 R	1016.7 ± 26.8	1002.5 ± 25.6
	<u>TLD-600 <sup>6</sup>LiF</u>	
50 mrem thermal neutrons	340.0 ± 22.5	343.9 ± 21.9

Dosimeters exposed to the high levels of radiation now have the same sensitivity as those exposed only to low levels within the uncertainty of the measurements.

Another set of dosimeters, <sup>7</sup>LiF only, was exposed to varying amounts of gamma radiation from 1 R to 30,000 R. These dosimeters had widely varying pre-radiation. Results of several comparisons of dosimeter groups with different exposure histories are shown in Table II.

Table II

<u>Control Dosimeters</u>	
1 R Exposure After 16-Hr Anneal	1.32 ± 0.08
Group 1 Previous Exposure 0.2-5000 R	1 R Exposure After 16-Hr Anneal
	1.28 ± 0.08
Group 2 Previous Exposure 1000-5000 R	1000 R Exposure After 16-Hr Anneal
	1173 ± 51
Group 3 Previous Exposure 5000-30,000 R	1000 R Exposure After 16-Hr Anneal
	1056 ± 63
Group 4 Previous Exposure 1000-5000 R	5000 R Exposure After 16-Hr Anneal
	9643 ± 340
Group 5 Previous Exposure 5000-30,000 R	5000 R Exposure After 16-Hr Anneal
	8644 ± 550

Table II (Cont'd)

Group 6 Previous Exposure 1000-5000 R  
30,000 R After 16-Hr Anneal  
100,400 ± 5400

Group 7 Previous Exposure 5000-30,000 R  
30,000 R Exposure After 16-Hr Anneal  
90,600 ± 3500

There is no significant difference in the sensitivity of the control group and group 1 which had previous exposures from 0.2 R to 5000 R. When the previous exposure is between 5000 R and 30,000 R the sensitivity decreases. Groups 4 and 5 show a significant difference in sensitivity even with the 16 hour anneal. Exposures above 5000 R appear to cause a permanent damage in the dosimeters or the 16 hour anneal at 400°C plus 2 hours at 100°C does not restore the sensitivity. It is possible that another procedure would restore the sensitivity above 5000 R.

To determine how well the dosimeters might function as low-level (1 R or less) dosimeters, the same seven groups were cycled again through the 16 hour anneal at 400°C and 100°C anneal and then exposed to 1 R gamma radiation. These results are shown in Table III.

Table III

All Groups After 16-Hour Anneal And 1 R Exposure

<u>Group</u>	<u>Reading</u>
1	1.13 ± 0.06
2	1.16 ± 0.06
3	1.08 ± 0.06
4	0.98 ± 0.02
5	0.98 ± 0.07
6	0.93 ± 0.05
7	0.79 ± 0.02

(Uncertainties are 1 standard deviation.)

Group 1 is the control group for this set since it showed no significant difference in sensitivity compared to controls in the previous set. The uncertainties shown are 1 standard deviation or 67% confidence limits. At 95% confidence, groups 1 through 5 are the same. This indicates that long-term anneal is effective in restoring the sensitivity of TLD-700 <sup>7</sup>LiF exposed to radiation levels as high as about 30,000 R. Groups 6 and 7 are definitely less sensitive indicating

some "permanent" damage. Materials in groups 6 and 7 received exposure in the range 35,000 R and greater. Particularly, some dosimeters in group 7 received as much as 60,000 R total.

#### CONCLUSIONS

The annealing for 16 hours at 400°C with N<sub>2</sub> gas plus 2 hours at 100°C does adequately restore sensitivity and remove trapped electrons in TLD-700 material exposed to high levels of gamma and neutron radiation. Although only a limited amount of work has been done, the procedure also seems to work for TLD-600 (<sup>6</sup>LiF). The procedure did not work for CaF<sub>2</sub>·Dy due to deterioration of the blocks after 16 hours at 400°C. For some reason the blocks crumbled after the anneal cycle was completed.

Future investigation includes a determination of the effect of the N<sub>2</sub> gas during the 400°C anneal by conducting a set of annealing studies without the N<sub>2</sub>. We also plan to study annealing in more detail after high-level neutron exposures.

#### REFERENCES

1. D. W. Zimmerman, C. R. Rhyner and J. R. Cameron, "Thermal Annealing Effects on the Thermoluminescence of LiF," Health Physics 12, p.525 (1966).
2. A. M. Harris and J. H. Jackson, "On the Low Temperature Annealing of TLD LiF," Health Physics 18, p. 162 (1970).
3. Per Spanne and C. A. Carlsson, "Efficiency Variations of Thermoluminescent LiF Caused by Radiation and Thermal Treatments," Proceedings of the Third International Conference on Luminescence Dosimetry, Riso Report #249, Part I, p. 48, Riso, October 11-14, 1971.
4. L. F. Kocher, G. W. R. Endres, L. L. Nichols, et al, "The Hanford Thermoluminescent Dosimeter," BNWL-SA-3955, May 1971.



THIRD INTERNATIONAL CONGRESS OF THE INTERNATIONAL  
RADIATION PROTECTION ASSOCIATION  
9-14 SEPTEMBER 1973, WASHINGTON D.C.

A TLD PERSONNEL MONITORING SYSTEM  
WITH AUTOMATIC PROCESSING

Lars Bøtter-Jensen, Poul Christensen and Benny Majborn  
Danish Atomic Energy Commission  
Research Establishment Risø  
DK 4000 Roskilde, Denmark

Abstract

The development work carried out at Risø on a TLD personnel monitoring system with automatic processing is summarized.

The system is based on identification coded personnel badges and includes an automatic TLD reader. Experimental results are presented and the interpretation of monitoring data is discussed.

1. Introduction

Thermoluminescence dosimetry (TLD) has been studied intensively for more than a decade, and is gradually being accepted as a useful method for routine personnel monitoring.

Experience gained at Risø has shown that a TLD system can favourably replace photographic film dosimeters for routine  $\beta$ - and  $\gamma$ -monitoring. In order to exploit the benefits of TLD most economically with respect to large groups (~ 1000 workers) an automatic read-out system is preferable.

This paper describes the automatic TLD system developed at Risø including a personnel dosimeter badge, and results obtained with the system are given.

2. Dosimeters and the TLD badge

The personnel TLD monitoring system has been based on combinations of  $LiF$  and  $Li_2B_4O_7:Mn$  solid tablets; the latter have been developed and analysed extensively at Risø. The main features of  $Li_2B_4O_7:Mn$  are good tissue equivalence, low cost and the possibility of re-use after read-out without annealing requirements.

The personnel TLD badge consists of an identification coded dosimeter holder and a matching cover with photograph and name for visual identification of the employee, see fig. 1.

The dosimeter holder (molded from styrene butadiene acrylonitrile) contains solid TL dosimeters in four depressions together with an identification number and a corresponding binary hole code. A sandwich shielding consisting of 1 mm aluminium is provided with a beta window thus situated that it corresponds to one of the dosimeter positions.

For the majority of the workers at Risø (~ 900) the dosimeter combination will be two  $Li_2B_4O_7:Mn$  dosimeters for routine gamma and beta recording and one  $LiF$  dosimeter to enable detection of occasional slow neutron exposures and to ensure proper interpretation of beta-/gamma monitoring results by comparison of the  $LiF$  and  $Li_2B_4O_7:Mn$  responses. A fourth dosimeter is reserved for read-out in case of instrument failure and for verification of unusually high doses.

However, for the small minority of workers (~ 100) who may be exposed to neutrons more regularly two  $^7\text{LiF}$  dosimeters will be used for  $\beta/\gamma$ -monitoring in order to ensure a proper  $\beta$ -evaluation, and an additional  $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$  dosimeter will be used for the detection of slow neutrons.

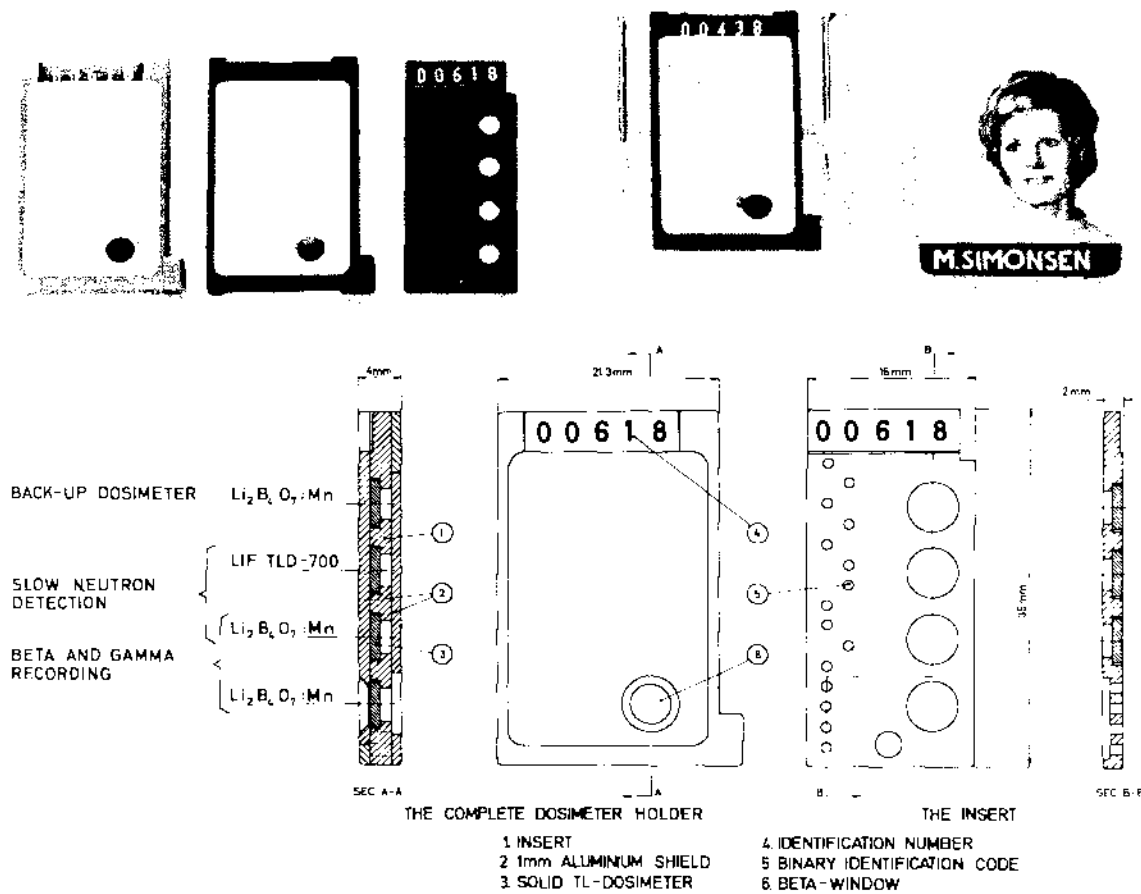


Fig. 1. At the top, an exploded view of the entire TLD-badge and at the bottom a schematic diagram of the TLD-holder.

### 3. The automatic reader

The photograph in fig. 2 gives a view of the complete reader and of the encoding punch for individual coding of the dosimeter holders.

Fig. 3 shows the entire read-out system diagrammatically. The automatic functions are pneumatically controlled and the read-out technique is based on heating the dosimeters with hot nitrogen gas.

Dosimeter holders are stacked in magazines from which they are fed, one by the other, into the reader where three dosimeters automatically are lifted in sequence from the holder into the read-out chamber. A nitrogen gas flow of 4 litres/min. is maintained at approximately 250°C. The total loading capacity allows for continuous automatic read-out of 450 individual TL holders.

During the transfer of holders the identification hole code is optically

scanned and the information together with the dosimeter readings are transmitted to a Teletype writer for printing and punching on paper tape.

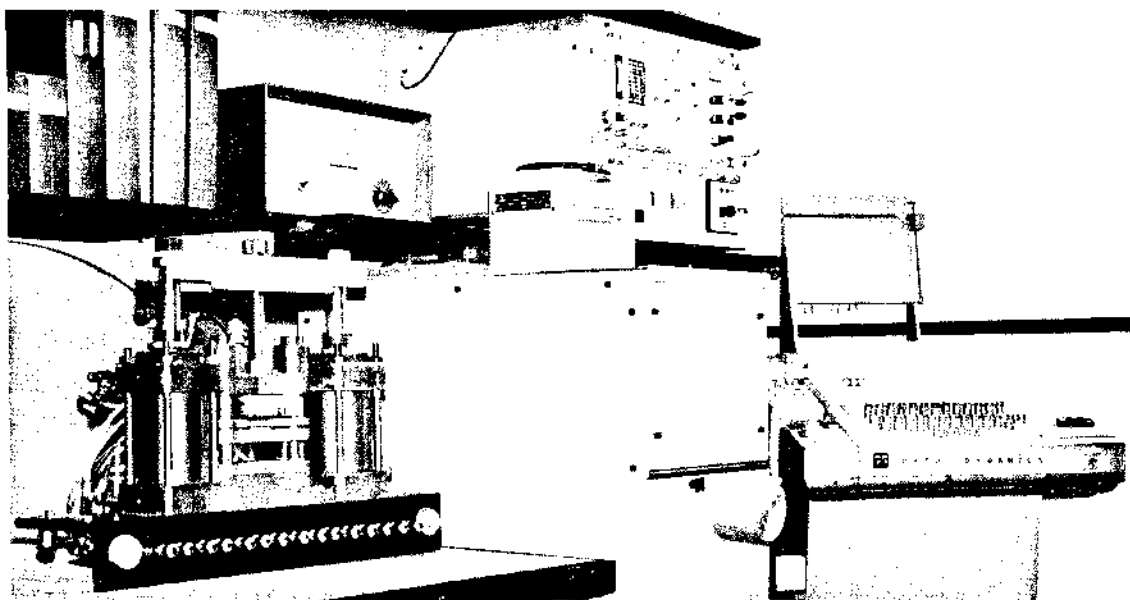


Fig. 2. Photograph showing the entire set-up.

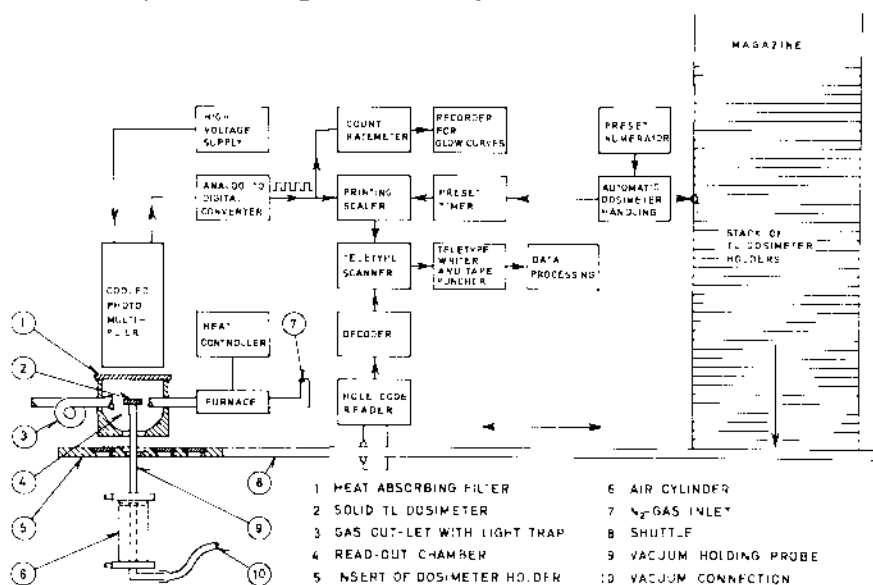


Fig. 3. The automatic TLD-reading system, schematically.

#### 4. Radiation response

##### 4.1. X- and $\gamma$ -rays

The response ( $^{60}\text{Co}$ ) of the  $LiF$  and  $Li_2B_4O_7:Mn$  dosimeters is linear for exposures from a few mR and up to about 1000 R, whereafter supralinearity is observed. Based on the glow-curve area, the  $LiF$  dosimeters are about two times as sensitive as the borate dosimeters (P.Y. Lubc IMA 9514-S).

The badge-response as a function of the angle of incidence was measured for  $^{60}\text{Co}$  and for different X-ray energies with the badge placed in free air and mounted at chest height on an Alderson average-man phantom, respectively. Ex-

posure rates were determined in free air with calibrated Baldwin-Farmer ionization chambers. Results are given in figs. 4 and 5.

From fig. 5 it will be seen that the overresponse for frontal incidence ( $0^\circ$ ) amounts to 50-60% for 100 keV X-rays. This overresponse arises from the combined effect of the dosimeter responses and the interaction with the badge and the phantom (compare figs. 4 and 5). From behind the badge is shielded by the body thus underestimating the dose. The energy dependence of the response (as can be deduced from fig. 5) compares well with the energy dependence of the absorbed dose to the testes.

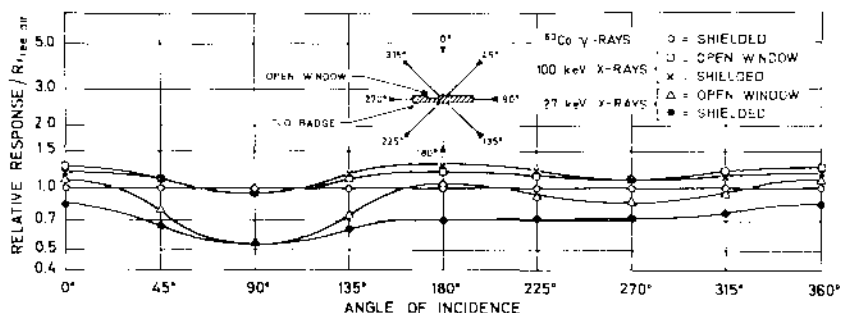


Fig. 4. Directional dependence curves for the TLD-badge.

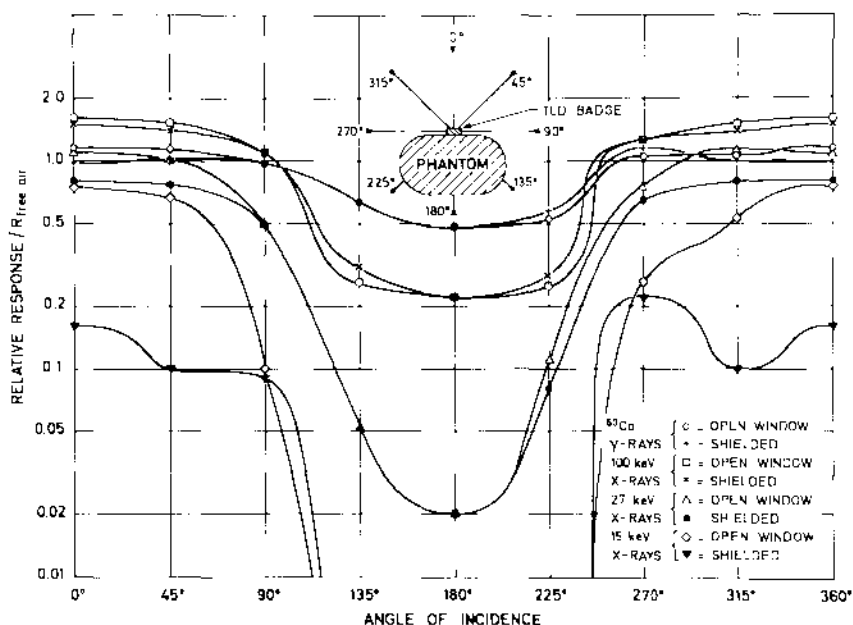


Fig. 5. Directional dependence curves for the TLD-badge mounted on a phantom.

#### 4.2. $\beta$ -rays

It appears that high energy  $\beta$ -rays, e.g. from  $^{90}\text{Y}$  will to some degree (app. 20%) penetrate the Al-shield and give rise to an overestimation of the  $\gamma$ -dose.

The relative open window response of the TLD-badge to  $^{90}\text{Y}$  beta rays at incident angles of  $0^\circ$ ,  $45^\circ$ ,  $90^\circ$ ,  $135^\circ$ ,  $180^\circ$ ,  $225^\circ$ ,  $270^\circ$  and  $315^\circ$  was measured to be 1.00, 0.71, 0.03, 0.41, 0.95, 0.41, 0.02 and 0.60 respectively.

#### 4.3. Neutrons

Owing to the large difference in the TL-response of  $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$  and  $^7\text{LiF}$  to a thermal-neutron exposure, the TLD-badge may be used for thermal-neutron dose estimation.

With the badge attached to an Alderson average-man phantom and exposed to an incident thermal-neutron fluence of  $10^{10}$  n/cm<sup>2</sup>, TL responses of 52% and 4.7% equivalent R have been measured for the  $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$  tablets and the  $^7\text{LiF}$  chips respectively.

### 5. Operational experience

#### 5.1. Stability

Investigations have been carried out on the reproducibility of the dosimeter-readings for  $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$  dosimeters when exposed in the TLD badge to 20, 100 and 500 mR  $^{60}\text{Co}$   $\gamma$  irradiation. Two dosimeter groups were analysed, one treated with the normal annealing procedure (pre-irradiation annealing 30 min. at 300°C and post-irradiation annealing 5 min. at 100°C) and the other without annealing. The results are given in table 1.

Table 1. The mean value and SD obtained from 10 successive  $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$  readings with varying arbitrary time intervals. Each reading is determined from the mean value of 9 dosimeter responses. Two groups of dosimeters, one treated with standard annealing and the other without annealing, were exposed to 20 mR, 100 mR and 500 mR of  $^{60}\text{Co}$   $\gamma$  irradiation.

	With standard annealing		Without annealing	
	Mean (Response/mR)	SD (%)	Mean (Response/mR)	SD (%)
20 mR	2.00	12.5	2.15	9.7
100 mR	1.81	2.7	1.88	3.7
500 mR	1.89	1.7	2.01	2.8

#### 5.2. Field experiments

Field experiments have been carried out at different areas at Risø. The employees were wearing TLD badges for monthly periods and doses were automatically recorded.

As earlier reported,<sup>2</sup> good agreement is on the whole found concerning the comparison of TLD and personnel films at higher doses (> 100 mR), whereas some disagreements are observed in the low-dose region, which may be attributed to inaccuracy of the film dosimetry system. An overresponse for  $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$  compared with the response of  $^7\text{LiF}$  observed in reactor areas indicates contribution from thermal neutrons.

### 6. Discussion and conclusion

On the basis of experience and experimental data the following advantages of TLD over film dosimeters have been recognized: 1) The monitoring cycle may be extended owing to less fading; e.g. from 3 months to one year for the non-critical group of workers. 2) Immediate results can be obtained with TLD, which may be important in case of unusual incidents, 3) The TLD system has a wide range of sensitivity allowing for emergency measurements and in addition for evaluation of mixed field  $\beta/\gamma$ /slow-neutron exposures 4) TLD offers tissue equivalence so that no energy corrections are required and 5) TLD is less sensitive to ambient humidity variations.<sup>8</sup>

Moreover, it should be emphasized that the easy handling and read-out technique makes TLD very attractive for automation which to some degree might be time saving (read-out during the night) and thereby also man-power saving. Experience has shown that for  $\beta/\gamma$  monitoring  $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$  is attractive, in particular owing to the simple annealing procedure which is convenient for automatic processing. Where neutron exposures contribute to the total dose more regularly,  $\text{LiF}$  is preferable.

The main importance of the thermal neutron dose estimation is 1) that occasional neutron exposures are detected and 2) that  $\beta/\gamma$ -monitoring results may be properly interpreted also if neutrons have contributed to the total dose.

Finally, it is concluded that the read-out technique based on hot  $\text{N}_2$ -gas heating has turned out to be very reliable with excellent reproducibility.

#### References

1. ATTLIX, F.H., A current look at TLD in personnel monitoring, *Health Physics* 22 (1972) 287.
2. CHRISTENSEN, P., A combined lithium borate and lithium fluoride thermoluminescence dosimeter for routine personnel monitoring, *Proceedings of the Symposium on Advances in Physical and Biological Radiation Detectors, IAEA, Vienna, (1971) 101.*
3. BØTTER-JENSEN, L., Read-out instruments for solid thermoluminescence dosimeters, using hot nitrogen gas as the heating medium, *Proceedings of the Symposium on Advances in Physical and Biological Radiation Detectors, IAEA, Vienna, (1971) 113.*
4. BØTTER-JENSEN, L., CHRISTENSEN, P., Progress towards automatic TLD processing for large-scale routine monitoring at Risø, *Proceedings of the Third International Conference on Luminescence Dosimetry, Risø Report No. 249 (1971) 851.*
5. JONES, A.R., *Health Physics* 12 (1966) 663.
6. MAJBORN, B., BØTTER-JENSEN, L., CHRISTENSEN, P., Thermoluminescence dosimetry applied to areas with mixed neutron and gamma radiation fields, *Proceedings of the Symposium on Dosimetry Techniques applied to Agriculture, Industry, Biology and Medicine, IAEA, Vienna, (1972) 169.*
7. BØTTER-JENSEN, L., CHRISTENSEN, P., and MAJBORN, B., Interpretation of monitoring data from a personnel TLD-badge exposed to mixed neutron and gamma radiations, *Symposium on Neutron Monitoring for Radiation Protection Purposes, IAEA, Vienna, (1972) IAEA/SM-167/4-6.*
8. CHRISTENSEN, P., BØTTER-JENSEN, L., MAJBORN, B., Influence of ambient humidity on TL dosimeters for personnel monitoring, *Regional Conference on Radiation Protection, Jerusalem, Israel, 5-8 March, 1973.*

## SOME PRACTICAL APPLICATIONS OF SINTERED BeO AS TL DOSIMETER

G. Benincasa, S. Caravolo, G. Scarpa  
Laboratorio di Dosimetria e Standardizzazione, CNEN  
Casella Postale 2400, Roma, Italy

### Abstract

Following the investigations reported in previous papers and symposia, some new experiments have been carried out in order to get further information about the possible application of the thermoluminescence of ceramic undoped beryllium oxide to some specific practical fields, such as health physics measurements and dose intercomparisons.

The results of these experiments are described and a conclusion is drawn that ceramic BeO, even of commercial electronic grade, can be used with success and advantage in the specific dosimetric fields referred to above, in addition to its typical use in mixed radiation fields of neutrons and gamma rays.

### Introduction

The general dosimetric characteristics of BeO as a thermoluminescent material have been extensively described and discussed in previous papers and reports 1,2,3.

From these investigations it has become rather evident that BeO, even in the form of plain, undoped, ceramic material as commercially manufactured for the electronic industry, possesses so many interesting properties to put itself among the most promising TL materials for routine dosimetric use. This applies in the first place to dosimetry of mixed fields of neutrons and gammas 3, because of the very low sensitivity of BeO to thermal neutrons.

But also in the field of conventional dosimetry of X and gamma rays BeO exhibits considerable advantages over other TL materials: low cost, absence of low-temperature or spurious peaks, simplicity of annealing cycle, high insensitivity to mechanical shocks and most chemical agents and long-term stability of response. These features are of special relevance in some specific fields of applied dosimetry, such as radiation protection measurements and dose intercomparison experiments.

In order to better define and study the possible application of BeO to the above mentioned dosimetric activities, a new set of specific experiments have been undertaken.

The three main topics covered by these experiments were: a) the linearity of response-to-exposure relationship with different radiation qualities; b) the energy dependence at various dose levels and c) the change of response with time elapsed from annealing to irradiation and from irradiation to readout.

All experiments have been carried out using the same instrumental equipment described in previous papers 2,3.

## Measurements and results

### a) Response-to-exposure relationship.

Figure 1 is a log-log plot of some results, already published, obtained using  $^{60}\text{Co}$  gamma rays, compared with a new set of measurements performed with 2 mm Cu HVL X-rays (effective energy: about 100 KeV). What is apparent is that the supralinearity, which begins in the region of 20-30 R, is more marked for gamma- than for X-rays.

This is better shown in figure 2, which is a plot of the linearity index versus exposure; it can be seen that the index is distinctly lower in the case of X-rays, at least for exposures of up to 1000 R, where the two curves approach each other again.

As linearity index the exponent  $n$  of the expression

$$A = K \cdot D^n$$

was assumed, in which  $A$  is the area of the glow-peak and  $D$  is the corresponding exposure. This index can be graphically derived as tangent of the angle between curve and abscissa axis.

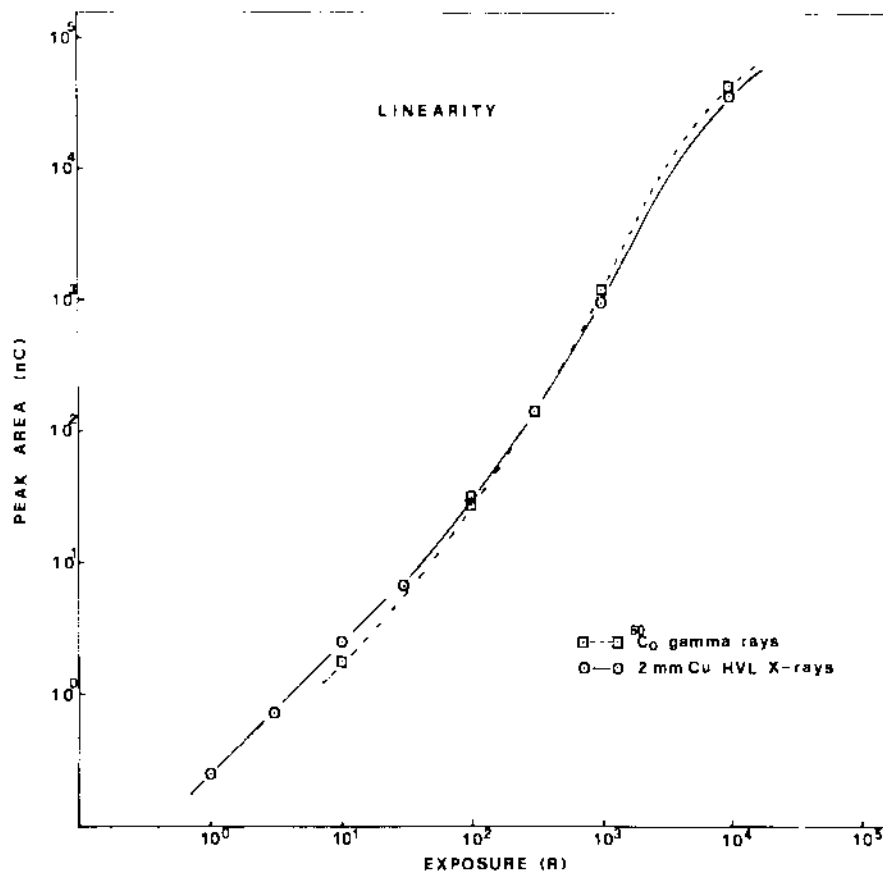


Fig. 1- Response-to-exposure relationship for gamma- and X-rays.



The different supralinearity of the two radiation qualities could be, at least partly, related to a dependence of supralinearity on LET, even if the difference between X- and gamma-radiation is relatively little in this respect. This dependence has been experimentally found by several authors in LiF and other TL materials <sup>4</sup>.

Tochilin et al. <sup>5</sup> have published a comparison of dose response curves of BeO exposed to <sup>60</sup>Co gamma rays and 9 KeV X-rays: the agreement with the above results is remarkable.

b) Energy dependence.

The energy dependence of the response of BeO to X- and gamma-rays was investigated at two different exposure levels: 10 and 100 R. Gamma rays emitted by <sup>60</sup>Co and <sup>137</sup>Cs sources and well filtered X-rays were used.

The results, normalized to unity for <sup>60</sup>Co gamma rays, are shown in figure 3. For both exposure levels a broad peak is apparent between 100 and 150 KeV; the experimental points at 100 R, however, are lower than those at 10 R. This is simply due to the fact that BeO exposed to 100 R already behaves supralinearly and, as shown in figure 2, the supralinearity is less marked for X- than for gamma-rays.

As a consequence, only the 10 R points can be regarded as representative of the real energy response of BeO. These points roughly agree with the curve published by Tochilin et al. <sup>5</sup> and included in figure 3, even if most of them are slightly below Tochilin's curve. A comparison with data published for LiF commercial solid dosimeters gives a slight advantage of BeO in this respect.

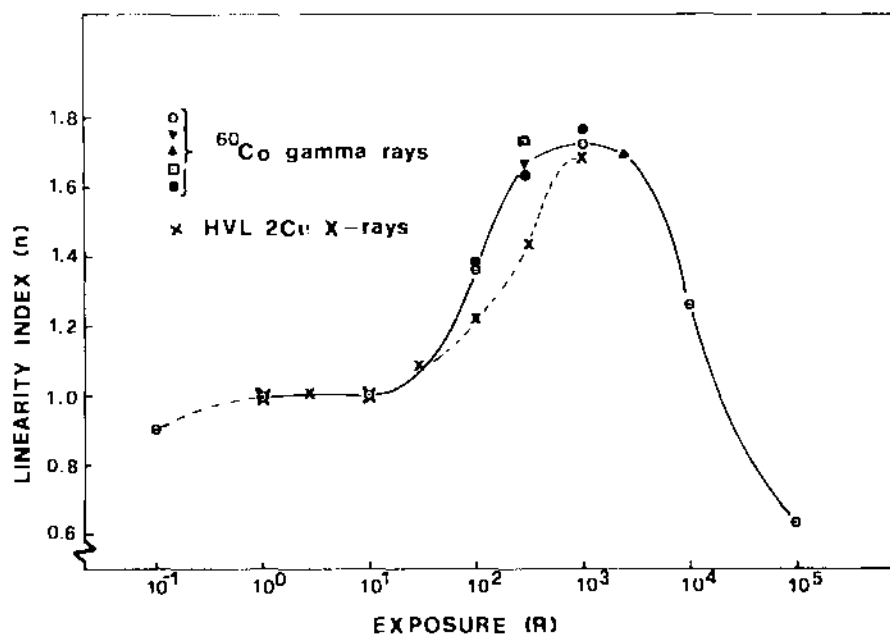


Fig. 2- Linearity index versus exposure for gamma- and X-rays.

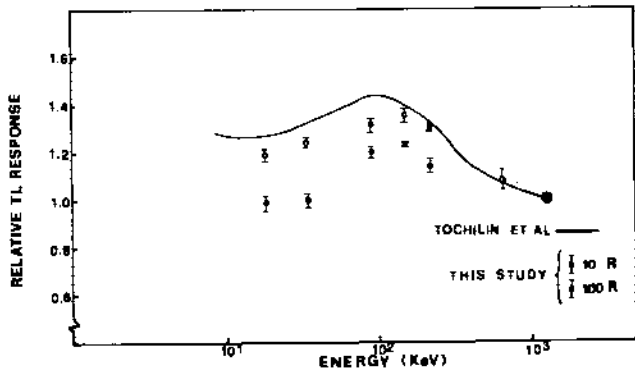


Fig. 3- Energy dependence of BeO irradiated to 10 and 100 R. Curve published by Tochilin et al.<sup>5</sup> is included.

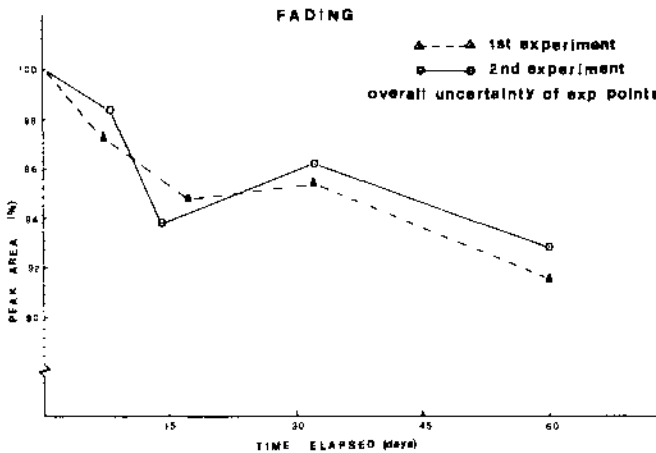


Fig. 4- Fading of BeO as resulted from two standard experiments.

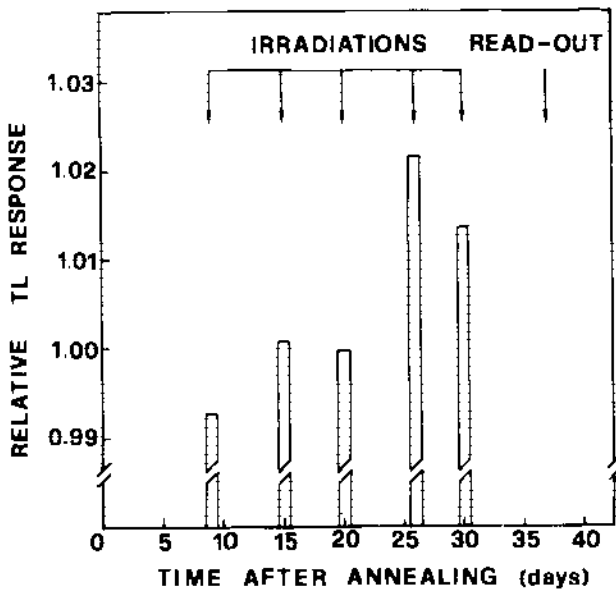


Fig. 5- Time-dependent variations of response of BeO in a typical time sequence simulating conditions met in health physics monthly measurements and dose intercomparisons.

c) Time-dependent variations of response.

A typical set of curves representing the fading characteristics of BeO is shown in figure 4. These curves were obtained by a standard experiment on fading, carried out by irradiating a number of freshly-annealed BeO discs and storing them (in the dark and at room temperature) for a variable period of time before readout. The initial drop of 5 to 6% during the first two weeks, the slight "recovery" at the end of the first month and the rather slow decay afterwards, reaching 8% at the end of the second month, are clearly illustrated.

Another experiment was then arranged in order to simulate more closely the actual time sequence of the above mentioned applications of TLD, i.e. health physics personnel dosimetry and postal intercomparisons of absorbed dose, in which all the dosimeters are initially annealed and finally read simultaneously, but irradiated at different time intervals in between.

As diagrammatically shown in figure 5, in this experiment a 37-days interval was selected between annealing and final readout, as an approximation to typical conditions met both in monthly badge checks and in international dose comparison runs.

The results are included in the same figure 5 in terms of average TL response of each single set of dosimeters, normalized to unity for the set irradiated on the 20th day after annealing.

Even if a slight build-up trend can be observed, the deviations are rather small, not exceeding  $\pm 2\%$ . From this relative stability of response a sort of compensatory effect can be inferred between fading on one side (as shown in figure 4) and something equivalent to an increase of sensitivity to radiation with annealing-to-irradiation time, on the other side.

Comments

Basing on the results of the present study and of previous contributions 1,2,3, and comparing these results with the ones reported for other TL solid dosimeters, it is apparent that ceramic BeO of commercial electronic grade could be used instead of doped LiF and  $\text{Li}_2\text{B}_4\text{O}_7$ , both in radiation protection measurements and in intercomparisons of dose by mailed dosimeters.

As far as health physics measurements are concerned a distinction should be made between routine personnel dosimetry (low dose levels) and accident dosimetry (medium and high dose levels).

Below 20-30 R the dosimetric characteristics of ceramic BeO are roughly the same as those reported for solid LiF: this applies to sensitivity (background equivalent to about 30 mR for 0.25 in. discs and model 2000 Harshaw reader), energy dependence (top value: 1.35 at 150 KeV) and fading in the dark (see above).

Irradiated BeO is highly sensitive to ambient light, with special reference to u.v. component, but this drawback can be easily overcome using the ordinary light-tight kind of badge used in film dosimetry.

On the other hand, as mentioned at the beginning of this study, distinct advantages of BeO over LiF and other TL materials are the very simple and quick annealing procedure (only 5 min at 600 °C), the absence of multiple and spurious peaks in the useful part of the glow-curve, the high insensitivity to mechanical shocks, the chemical inertness, the low cost (around 50 cents for 0.25 in. diameter discs) and the absence of appreciable changes of sensitivity even after very heavy irradiations (up to  $10^5$  R).

As to the accident dosimetry, a drawback of BeO could be the lower dose level at which supralinearity appears (20-30 R, compared to 500-1000 R for LiF and  $\text{Li}_2\text{B}_4\text{O}_7$ ). This requires a set of calibrations to be carried out in order to derive the whole response curve. On the other hand, all the above mentioned advantages still apply in this field.

Dose intercomparisons made by very small sized dosimeters, deliverable by ordinary post, are an ideal field of application of TLD. In this respect, the good mechanical and chemical properties of BeO, its high long-term stability, its moderate energy-dependence and its very good reproducibility in the medium range of doses, seem to put this material in a position of privilege. This had to be confirmed by practical experiments: for this purpose BeO ceramic discs have been added to ordinary LiF powder filled containers during the second dose intercomparison among radiobiologic laboratories organized by the European Late Effect Project Group (EULEP). The results of this experiment, though still under evaluation, seem to give further support to the concrete possibility of using BeO as transfer dosimeter in inter-comparisons.

#### References

- 1- G. Scarpa, Phys. Med. Biol. 15, 667-672 (1970)
- 2- G. Scarpa, G. Bonincasa and L. Ceravolo, Proc. Third Int. Conf. on Lumin. Dosimetry, Risø, 11-14 Oct. (1971)
- 3- G. Scarpa, G. Benincasa and L. Ceravolo, Symp. on Dosimetry Techniques applied to Agriculture, Industry, Biology and Medicine, Vienna, 17-21 Apr. (1972)
- 4- J.R. Cameron, N. Suntharalingam, G.N. Kenney, Thermoluminescent Dosimetry, The University of Wisconsin Press (1968)
- 5- E. Tochilin, N. Goldstein and W.G. Miller, Health Physics 16, 1-7 (1969)

COMPOUNDS OF TLDs AND HIGH-MELTING ORGANICS FOR FAST  
NEUTRON PERSONNEL DOSIMETRY\*)

Klaus Becker, T.D. Tham\*\*) and F.F. Haywood

Health Physics Division  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830

ABSTRACT

The search for a replacement of the NTA film in fast neutron personnel dosimetry has not yet produced a completely satisfactory result. Various investigators have attempted to increase the low fast neutron sensitivity of inorganic thermoluminescent materials by mixing them with liquid or solid organic recoil proton radiators, but most of the systems described so far either exhibited a very pronounced energy response, or they required the separation of the constituents prior to evaluation, because almost all hydrogen-rich organic compounds evaporate and/or disintegrate at temperatures below those which are required for the read-out and annealing of the more stable TLD materials.

In this feasibility study, a finely powdered, highly sensitive and fading-resistant phosphor ( $\text{CaSO}_4:\text{Dy}$ ) has been hot-pressed into pellets with p-sexiphenyl (melting point  $\sim 450^\circ\text{C}$ ) as a high-melting and stable hydrogenous radiator. Outgassing of the pellets in a vacuum furnace at  $150^\circ\text{C}$  removed residual volatile impurities. The response of detector pairs, consisting of such pellets and of Teflon-embedded phosphors, has been investigated.

The sexiphenyl-embedded phosphor is sensitive to UV light. An efficiency of about 50 % of the gamma radiation response was observed for recoil protons produced by fission and also for 14 MeV neutrons. The response is a linear function of dose. Desirable follow-up studies are outlined.

---

\*) Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

\*\*) IAEA Fellow, on leave from Vietnam.

In recent years, the discussion about various possible replacements of the NTA film as a fast neutron personnel dosimeter has intensified (1-3). Among the more promising approaches are:

1. track etching techniques, either based on recoil nuclei or on fission fragment registration (for a review, see ref. 4);
2. recoil proton registration with detectors based on stimulated exoelectron emission (5,6); and
3. recoil proton registration with TLD, which is the subject of this paper.

It was first shown almost ten years ago that the low inherent fast neutron response of inorganic TLD detectors can be increased by intimately mixing them with hydrogenous materials. This principle was first used (7) by mixing LiF:Mg,Ti during irradiation with alcohol, which can easily be evaporated prior to the reading. Further studies of the energy response of the system (8) were only partially encouraging, because the system required continuous stirring or shaking during exposure, and its response decreased rather rapidly with neutron energy (0.17 per 100 erg/g neutron kerma in tissue at 2.9 MeV, 0.66 at 14.9 MeV). A better response at low neutron energies could be expected with extremely fine-grained LiF, but this introduces other problems such as a drop in inherent radiation sensitivity. Naturally, mixing of LiF with alcohol or other liquids also affects its photon energy dependence (9). Mixing of fine LiF powder with water is not recommended because this induces a rapid fading (10).

Other investigators have, therefore combined TL phosphors with solid organic materials, either by just covering a solid TL phosphor layer with radiator foils (11, 12), or by intimately mixing a phosphor such as  $\text{CaSO}_4:\text{Tm}$  with a water-soluble radiator such as glucose (13). This mixture is exposed in a Teflon tube together with another tube containing  $\text{CaSO}_4:\text{Tm}$  only; the glucose is washed out prior to evaluation; and the difference in the reading of the two detectors is related to the fast neutron dose.

Obviously, it would be much more convenient in routine applications if compounds of the TL phosphor and a high-melting radiator could be made which are capable of undergoing multiple readout and annealing cycles without the need for separating the constituents. Some work along this line was carried out in the Soviet Union (14), but no details are known and attempts to reproduce the reported results have met with limited success. As there are several fairly easily available hydrogenous compounds with melting points above  $300^\circ\text{C}$ , a search for a material with desirable properties was carried out. Lithium hydride (M.P.  $\sim 690^\circ\text{C}$ ), ammonium aluminum chloride (M.P.  $304^\circ\text{C}$ ) and some other inorganic compounds had to be ruled out due to poor stability in humid air and/or low hydrogen content.

There are several organics which are not intensely colored and would not, therefore, absorb too much of the TL light. Some of them are: adenine (365°C); l-alanine (318°C); 1, 5 - dinitro-anthroquinone (384°C); 2-hydroxy-anthraquinone (302°C); m-azoxybenzoic acid (345°C); benzidine sulfone (327°C); p-carboxy-cinnamic acid (358°C); dinocotinic acid (323°C); guanine (360°C); isonicotinic acid (317°C); isophthalic acid (330°C); dl-isovaline (307°C); dl-leucine (332°C); d- or dl-norleucine (< 300°C); quercetin (310°C); 5-quinolinecarboxylic acid (339°C); taurine (328°C); theobromine (337°C); trimelic acid (~ 350°C); dl or d-tyrosine (316°C); uracil (338°C); methyl uric acids (360-400°C); l-valine (315°C); and dicoumarin (> 330°C).

Unfortunately, many of these materials undergo rapid deterioration (discoloration, sublimation, "outgassing" of volatile impurities, etc.) when heated repeatedly to temperatures close to their melting point. We concentrated, therefore, on high-melting polyphenyls such as p-quaterphenyl (~ 320°C), which is commercially available (Pilot Chemicals Div., Watertown, Mass. 02172), and p-sexiphenyl (~ 450°C) which has been prepared at ORNL (15). Polyphenyls are known to be stable compounds with a relatively high hydrogen content (5.9 % in quaterphenyl, 5.68 % in sexiphenyl) and interesting luminescence properties. Even the carefully purified material, however, exhibited some sublimation of impurities at high temperatures which led to a white condensate at the interference filter of the TL reader. After keeping the samples in a vacuum furnace at 150°C overnight, this disturbing effect disappeared.

Small pellets of about 20 mg weight, 6 mm diameter and less than 1 mm thick, have been produced by hot-pressing a mixture of very finely powdered (average grain diameter ~ 4 µm) TL phosphor with the powdered polyphenyl or Teflon at about 200°C and a pressure of about 2000 kg/cm<sup>2</sup>. As a phosphor, CaSO<sub>4</sub>:Dy, which has been prepared as previously described (16), was used. Some thermal neutron sensitivity of this material due to its Dy content was of no concern in these tests because it affects both the Teflon- and the sexiphenyl-embedded material to the same extent. Several mixing ratios have been tried, but a 1:1 ratio was chosen as a compromise between sensitivity and mechanical stability requirements. The detectors were annealed at 280°C (Teflon) and 350°C (p-sexiphenyl), respectively, for about two hours prior to re-use. No residual TL signal could be detected after this treatment, and there was no change in sensitivity during multiple use.

The response of both the Teflon and the p-sexiphenyl embedded detectors is a linear function of the gamma radiation dose between at least 1 and 1000 rad. The response of the Teflon-embedded detectors was 75 % higher due to a larger quantity of phosphor per detector. The standard deviation of the dose readings, both during multiple reading of the same and of different detectors, amounted to less than ten percent but could undoubtedly be reduced to 1-2 % for mass-produced detectors which are produced under more carefully controlled conditions.

For exposure, the Teflon-embedded detectors were placed in a Teflon shield, and the sexiphenyl-embedded detectors in a polyethylene shield of about 2 mm thickness in order to establish recoil particle equilibrium. The results of various exposures of the detectors to mixed neutron and gamma radiation fields are summarized in Table I.

Table I. Fast Neutron Response of  $\text{CaSO}_4:\text{Dy}$  Embedded in p-Sexiphenyl

Radiation Source	Sexiphenyl/Teflon response ratio *)	neutron/ $\gamma$ dose ratio (rad/rad)	recoil proton efficiency
HPRR reactor	3	7	$\sim 0.4$
14 MeV (d,T)	10	25	$\sim 0.4$

\*) Normalized for equal gamma radiation sensitivity

As can be seen, the relative efficiency of the phosphor to recoil protons amounts to about 40 % for both fission and 14 MeV neutrons, which is more than what one would expect from the known LET response of TL phosphors. Perhaps some of this response is due to an "indirect" response of the phosphor to light which is produced by the protons traversing the highly luminescent sexiphenyl. Indeed, a relatively high sensitivity of the sexiphenyl-embedded material to UV light was observed, which made it necessary to handle the detectors in red light instead of the standard laboratory fluorescent light illumination. It is not known whether this UV sensitivity is due to an optical or an electronic energy transfer between the sexiphenyl and the phosphor.

In further studies, it is intended to employ some other constituents, with low-Z phosphors with good sensitivity and stability, but low thermal neutron response ( $\text{Mg}_2\text{SiO}_4:\text{Tb}$ ,  $\text{BeO}$  etc.) replacing the  $\text{CaSO}_4:\text{Dy}$ , and other organic compounds including some hydrogenous, temperature-resistant polymers (polyimides, Dextsil, polyaryl sulfone etc.) replacing the p-sexiphenyl. Also, the phosphor to radiator ratio has to be varied within wider limits, and the effect of protective coatings of the detector pellets be studied. The practical threshold of the system for lower neutron energies (estimated to be presently around 0.1 MeV) has to be established, and the photon energy response of the optimized detector must be measured.

It is hoped to arrive, after these studies have been completed, on a system for fast neutron dosimetry which is as simple to employ as the presently used  $^6\text{LiF}/^7\text{LiF}$  pairs for thermal neutron dosimetry.

Acknowledgements. The authors would like to thank P.T. Perdue for providing the p-sexiphenyl used in these experiments, and R.L. Shoup for the 14 MeV neutron exposures.



## References

1. K. Becker, ORNL-TM 4279 (1973).
2. Proceed. Symp. Neutron Monitoring for Radiat. Protect., IAEA Vienna (1972).
3. K. Becker, Solid-State Dosimetry, CRC Press (1973).
4. K. Becker, Chapter 2 in "Topics in Radiat. Dosimetry" (F.H. Attix, ed.), Academic Press, New York (1972).
5. K. Becker and K.W. Crase, Nucl. Instr. Meth. 82, 297 (1970).
6. K. Becker and M. Abd-el Razek, Nucl. Instr. Meth., in press (1973).
7. C.J. Karzmark, J. White, and J.F. Fowler, Phys. Med. Biol. 9, 273 (1964).
8. C.L. Wingate, E. Tochilin, and N. Goldstein, in: Luminescence Dosimetry, AEC Symp. Ser. No. 8, CONF-650637, p. 421 (1967).
9. G.W.R. Endres, R.L. Kathren, and L.F. Kocher, Health Phys. 18, 665 (1970).
10. Z. Spurny, J. Novotny, and L. Hedvicakova, Phys. Med. Biol. 16, 295 (1971).
11. R.A. Faccy, Paper, Ann. Meet. Am. Nucl. Soc. (1968).
12. C.M. Sunta, K.S.V. Nambi, and V.N. Bapat, Proceed. Symp. Neutron Monitor. for Radiat. Protect., IAEA Vienna (1972).
13. E. Blum, D.K. Bewley, and J.D. Heather, Phys. Med. Biol. 17, 661 (1972).
14. V.A. Kazanskaya et al., Jad. Priboestroenie 13, 118 (1970).
15. P.T. Perdue and W.H. Baldwin, Nucl. Instr. Meth., to be published.

## THE SIEVERT LECTURE

ROLF M. SIEVERT: THE PIONEER IN THE FIELD OF RADIATION PROTECTION

Karl Z. Morgan  
School of Nuclear Engineering  
Georgia Institute of Technology  
Atlanta, Georgia 30332

On this occasion we gather to honor Rolf M. Sievert. He laid the foundation for medical and radiation physics in Sweden in 1919 when he offered and made available unpaid collaboration with members of the medical profession. These efforts led to marked improvement in the diagnosis and treatment of malignant diseases and use of ionizing radiation not only in Sweden but throughout the world. From July 1924 the cancer society employed Sievert and gave him a salary as head of the Physics Laboratory of the Cancer Clinic in Stockholm. He used this salary and far more of his own money to develop this laboratory into a world famous radiation research institution. In 1941 Sievert was appointed Professor of Radiation Physics at the Karolinska Institutet. His main contributions to clinical physics were from 1920 to 1940. Although he was an active contributor to the science of radiation protection for over 40 years until his untimely death at the age of 70 on December 3, 1966, the work he inspired among his colleagues at the Institutet still continues and his influence will be felt among radiation physicists and radiation protectionists as long as man is concerned with his well being. His important paper in 1921 on the distribution and intensity of primary gamma radiation of radium preparations as used in medicine was a major advance in the subject and was a first step toward qualification of radium dose to patients. This work resulted in the scientific community naming in his honor the basic shielding integral as the Sievert integral. By 1925 he was actively discussing problems of radiation protection and had set up a mobile measuring laboratory which standardized dosimetry and measured the intensity of x-ray facilities placing Sweden in the forefront in radiation physics.

During the Second International Congress of Radiology in Stockholm in 1928, plans were made leading to the formation of the International Commission on Radiological Protection and it is no surprise that Rolf Sievert along with L. S. Taylor (U.S.), Grossman (Germany), Kaye and Melville (Great Britain) comprised the membership of this first Commission. In 1932 Sievert published in *Acta Radiologica* his famous paper on the condenser chamber method of measurement and after this a series of publications on applied mathematics, experimental physics, radiological units and dosimetry, radiation protection, and radiobiology. In the late 1940s he became a pioneer in the development of his pressure ionization chambers for the measurement of gamma radiation from the human body and with these and other instruments made many measurements of natural background radiation.

Sievert was primarily responsible for developing the first Swedish radiation protection laws in 1941 and his Institutet, under his leadership, was in charge of radiation protection problems in Sweden until he retired in 1965. Sievert was not only the leader in radiation protection in his own country, but he was a giant in the world community. He was a member of ICRP, ICRU,

and of the UNSCEAR and was one of the organizers of IRPA that is meeting here today.

Sievert was a dynamo of new ideas and had an uncanny intuition which guided him to pursue and bring to fruition research activities, goals, and objectives which proved to be the ones most important. He was a generous man and sought ways of helping others. He was modest in regard to his own accomplishments and offered encouragement even to the least of those of us who might follow in his footsteps. He was gracious and kind and made all who knew him proud to be his associates and friends. He was jovial, good natured, and the life of the party, where his close friends soon realized he could drink any of us under the table, yet he carried a dignity that made him seem to stand just a bit taller in stature and to us more important than the others about him. He had a warm personality that endeared him to all of us who proudly honor him on this occasion of the Sievert lecture. Rolf received many high honors during his lifetime and shortly before his death received a copy of a special issue of the HEALTH PHYSICS JOURNAL in his honor. This man, who was born in 1896, the year in which Becquerel discovered radioactivity, had a major impact on the lives of all members of IRPA and especially on the life of Dr. Bo Lindell, one of his students and closest associate, whom we are proud to honor on this occasion. We are requesting that copies of this lecture and all the proceedings of this session be sent to Dr. Sievert's wife so that she, with us, can enjoy this occasion. I am proud at this time to present the first Sievert lecturer, Bo Lindell, of Stockholm, Sweden.

# RADIATION AND MAN

## THE 1973 SIEVERT LECTURE

Bo Lindell

National Institute of Radiation Protection  
Fack, S-10401 Stockholm 60, Sweden

The honour that has been bestowed upon me by selecting me as the first lecturer in this new series of lectures must be a reflection of the admiration that my colleagues still feel for Rolf Sievert and their hope that one of his pupils might be able to pay him a proper tribute in this first Sievert lecture.

Let me, however, begin by claiming that this is an impossible task: no pictures, no descriptions, no quotations can do Rolf Sievert justice. Only life could bring into his stout body the vitality and the magnetism by which he mesmerized his environment. Those who were never subjected to that forceful vitality and to the cascade of ideas, innovations, plans and solutions that flowed from Rolf Sievert in a glittering, boisterous torrent will never be able to see in the dead pictures of Sievert the man he was to us who knew him.

Furthermore, it would not have pleased Sievert to have a lecture of this kind focussed on himself. He would have felt warm at heart by the honour shown to his memory by the creation of the Sievert Prize, but since he was a man who liked results, he would have felt embarrassed if this lecture did not leave the personal field soon enough for the technical areas of radiation protection to which he devoted his life.

I have chosen to talk about the general subject of "Radiation and Man", a subject as wide as Sievert's interests. Let us begin by appreciating that, thanks to the pioneering efforts of men like him, radiation safety problems are dealt with in a special way and with much more concentrated efforts than any other occupational or environmental risk.

For example, thanks to the efforts of ICRP, the International Commission on Radiological Protection - founded in Stockholm already in 1928 - we have had for almost forty years an internationally applied set of dose limits which guarantee that no harmful acute effects will result from normal uses of radiation. We should recall that the prevention of immediate toxic effects is still the main problem in many conventional types of occupational or environmental protection, we may just recall substances such as mercury and DDT.

With the conventional standards of thinking, small doses of radiation would be considered not only safe but also often non-existent. Let us not forget that laws on food additives in many countries until recently have completely forbidden any presence of carcinogenic substances, but that the definition of a "zero quantity" has been "a non-detectable quantity". Had radioactive substances been chemically toxic instead of radioactive, many of them would, in the terms of the law, not have existed until new scientific detection methods had revealed their existence and complicated life for the health authorities.

Remember also that even the lung cancer risk from tobacco smoking, although now rather obvious, has been regarded with a good deal of skepticism until recently. Any possible genetic risk of smoking remains to be discussed. Risks of cancer from coffee, tea, alcohol, smoked food, etc are sometimes discussed but never in a quantitative way. To most people, these are hypothetical risks or more or less accepted risks of living.

In the field of radiation protection, risks of cancer and hereditary defects have been assessed quantitatively for almost twenty years and the possibility of such risks even after very small doses of radiation has been used as the basis for protective measures, although there is no proof that these risks really exist.

When we talk about risks we have a semantic problem. Different people mean different things when they say "safe". This may, indeed, be the heart of the matter in the current debates on radiation safety, and I shall take this point as the basis for my lecture.

I will start with a reference to table 1 showing the radiation doses which we receive from natural sources of radiation:

Table 1: Annual doses from natural sources

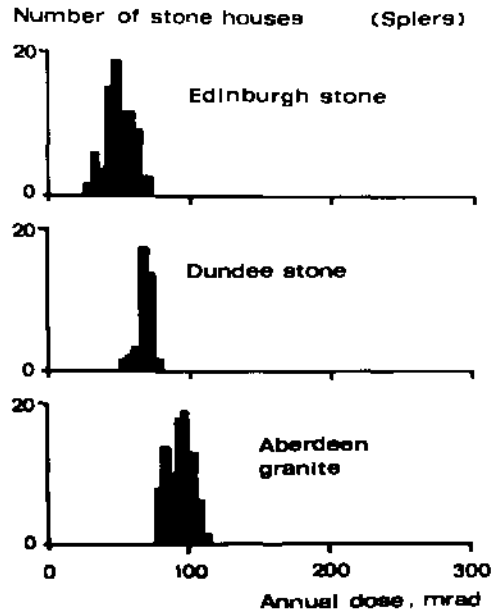
Source	Annual dose mrad
Cosmic radiation	30
Terrestrial radiation	50
Internal sources ( <sup>40</sup> K)	20

This is a rough approximation of more detailed information which you may find in, for example, the 1972 report of UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation).

Of the three components of the total natural radiation dose, the dose from terrestrial radiation shows the largest variations and is also usually the largest component. Even if we exclude data from areas with particularly high concentrations of radioactive materials in the ground, such as the monazite-bearing regions in Kerala and Brazil, there are wide variations in the exposure rate on the ground and inside buildings. Observations on the variation in the radiation dose in buildings of various materials were published in several countries in the 1950's, and I shall only show a few data that have been widely published.

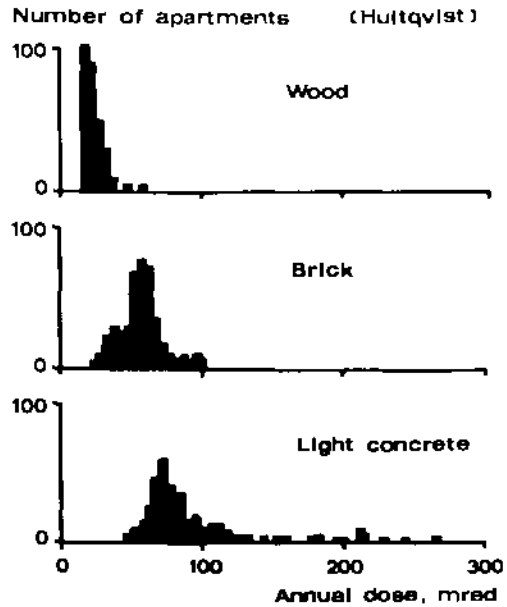
You can see from the UK data in figure 1, derived from Spiers, that a person in Scotland might get an extra 50 mrad per year if he stays indoors in a house of Aberdeen granite instead of in a house of Edinburgh sand-stone.

Figure 1: Annual gonad doses from gamma radiation in Scottish buildings



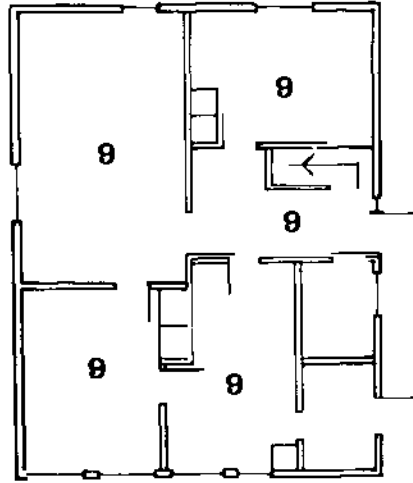
The Swedish data in figure 2, from Hultqvist, shows that a person in some Swedish light-concrete houses might get an extra 200 mrad per year as compared with a person in a wooden house.

Figure 2: Annual gonad doses from gamma radiation in Swedish apartments



These data may be familiar to many of you. I shall add a few more recent results which illustrate the variations in the natural radiation in buildings. You will see from figure 3 that there is little variation in the exposure rate in a house where no building material is particularly radioactive.

Figure 3: Exposure rate ( $\mu\text{R/h}$ ) in an apartment with walls of light concrete of low activity



I have marked exposure rates in microroentgen per hour with the contribution from the cosmic radiation subtracted but with no reduction for absorption in the body. You will obtain the annual depth dose in the gonads, assuming an absorption factor of 0.6 and 15 hours indoors per day, by taking each  $\mu\text{R/hr}$  equal to 3 mrad/year.

Figure 4 shows the situation in a building with mixed materials and you can see that the exposure rate is higher near those construction elements which have a higher activity concentration and which are marked with black in the figure. In such a house the annual doses may differ with 10 or 20 mrad per year between persons who stay for long periods in different rooms.

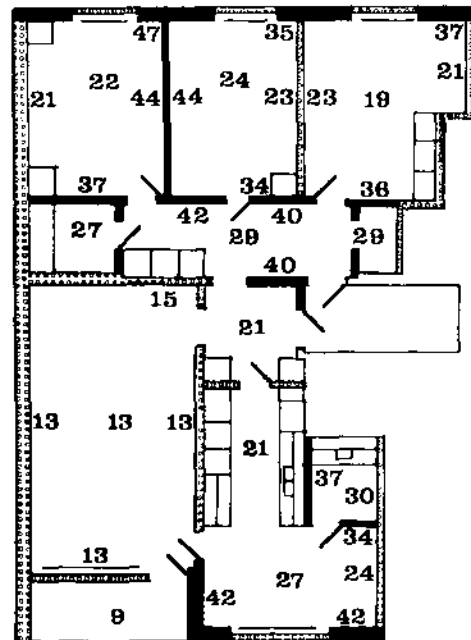


Figure 4: Exposure rate ( $\mu\text{R/h}$ ) in an apartment with walls of mixed materials

In figure 5 you will see the exposure rates in a house with extensive use of a material with comparatively high activity. Here the inhabitants may well receive actual depth doses which are more than 200 mrad higher than in a more normal house.

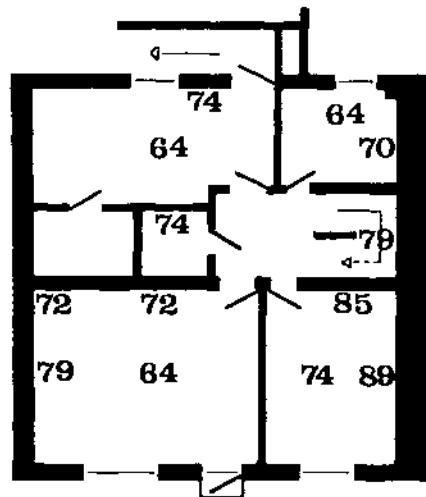


Figure 5: Exposure rate ( $\mu\text{R/h}$ ) in an apartment with walls and other construction elements of light concrete of high activity

From 1956, that is for 17 years, ICRP has given recommendations not only for the protection of workers but also for the protection of the public. All dose limits that have been recommended by ICRP have in common that they do not apply to doses from natural sources of radiation nor to doses received by patients in medical procedures.

These exceptions have been difficult for many to understand, but they are natural consequences of the cautious attitude that ICRP and radiation protection authorities are taking. If there were a threshold dose as indicated in figure 6, below which no cancer and no genetic harm could be caused by radiation, then the primary goal would naturally be to limit the total dose, irrespective of source, that would be received during the biologically relevant period. With that assumption, dose limits would have to apply to the sum of all doses, with no exceptions.

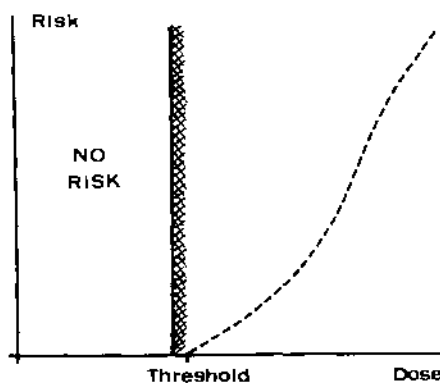


Figure 6: The concept of a dose/effect-relationship with a dose threshold for risks



Even without any threshold but with a non-linear relationship between risk and dose, the same principle would have to apply (figure 7).

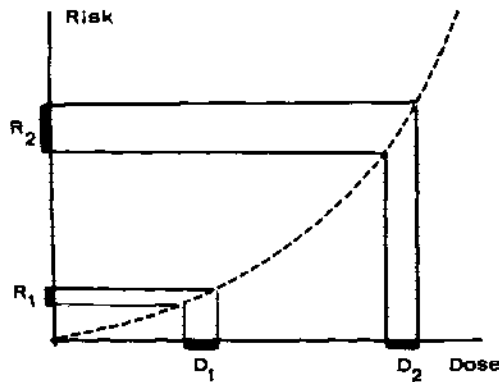


Figure 7: The non-linear dose/effect relationship

For the national authorities this would be a particularly nasty situation. The lack of threshold would mean that any additional dose, however small, would imply an additional risk. That risk, however, would not only depend upon the magnitude of the dose increment but also on the slope of the risk-dose curve. That slope would be different for each individual, depending upon his starting point on the curve, *i.e.* depending upon his total previous exposure. In risk-benefit assessments one would have to realize that one and the same dose might mean different risks to different individuals and in order to cope with this situation in a quantitative way one would need to have full records of all previous exposures. The potential bureaucracy that this could lead to is just frightening and it is possible that the authorities would have to stipulate an average slope of the risk-dose curve, to apply without knowledge of previous exposures. That would mean overestimating some persons' risk situation and underestimate that of others, but the assessment of the total expected harm would still be correct.

The present policy, however, is to assume that the risk-dose relationship is linear, even though this is by no means certain (figure 8).

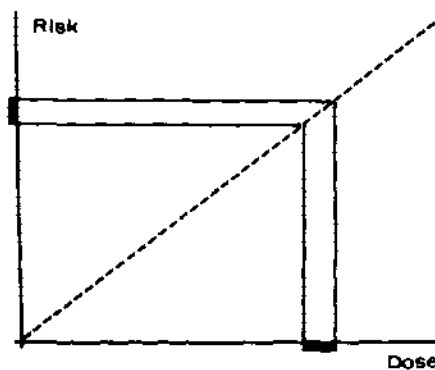


Figure 8: The linear dose/effect relationship

In this case the slope is the same at all points, therefore the risk per rad, which I shall call the risk coefficient, is the same for all persons independent of previous exposures, and the risk from any additional dose is directly proportional to the dose increment.

On this assumption, and we have to recall that it is an assumption for protection purposes, any radiation exposures which are controllable can be made subject to risk-benefit assessments without knowledge of previous exposures. If such assessment is carried out for each source or project, dose limits may be considered superfluous. Nevertheless, the present attitude is to consider dose limits a valuable convention even though the more basic recommendation must be the one given in paragraph 52 of ICRP Publication 9, namely that "all doses be kept as low as readily achievable, economic and social consideration being taken into account".

From what I have said now it can be concluded that it is not meaningful to give dose limits for natural radiation but that there is no reason to exempt natural radiation from cost-benefit assessments as far as countermeasures are concerned. Obviously no easily eliminated natural source of radiation should be accepted any more than other radiation sources.

I have now sketched the background for a general radiation protection philosophy. Any practice that results in radiation exposures should be subject to a risk-benefit assessment. If the possible risk is acceptable, then the practice should still not be accepted until it has also been shown by a cost-benefit assessment that the resulting exposure is so low that further dose reductions will not bring about a benefit which justifies the effort.

For the risk-benefit assessment, the risk coefficient, covering all expected incidence of cancer and all severe hereditary defects in the first generation offspring, is usually assumed to be a few times  $10^{-4}$  per rad after whole-body exposure, perhaps  $10^{-4}$  per rad after exposure at low dose rates and 3-4 times higher at high dose rates if the assumption of a linear dose-risk relationship is correct. In cost-benefit assessments there is some guidance in the knowledge that it is not unusual in current radiation protection practice to consider it reasonable to eliminate a radiation dose if it can be done at a cost of 100 \$ per rad and person.

These two quantitative assumptions characterize present radiation protection activities except where special public concern may have influenced the decisions. It is interesting to note that the combination of the risk coefficient and the dose elimination cost with the values that I have just mentioned implies a value of one million dollars for each human life which might, on statistical grounds, be expected to be saved.

I am now getting into dangerous grounds and do not wish to be misunderstood to mean that the reverse thing is true, namely that it would be justified to risk a human life in order to save or gain a million dollar to society. Lives and dollars are not exchangeable quantities.

The interesting thing with a low risk, however, is that individuals tend to treat it as a nuisance more than as a reality, if they understand that it is low and if they try to comprehend its significance. It seems to be a fact that many people have the feeling that they better "understand" what a low dose implies if they are told what it is worth to pay to eliminate the dose. In my experience many persons who are knowledgeable in radiation risk assessments react immediately, perhaps with their spinal cord, when they are told the dollar equivalent of a radiation dose (the PQR-cost<sup>1)</sup>), and interestingly enough their immediate reaction on the PQR-cost will prove to give about the

---

<sup>1)</sup>The arbitrary letters "PQR" were chosen as the symbol for this quantity by my friend Dr Arne Hedgran who invented the concept.

same result as their more scientific brain exercise when they make a direct risk assessment.

Take, for example, the natural radiation. I have shown that an extra annual dose of 10 mrad is not unusual in certain buildings. The individuals concerned may feel that this is rather insignificant when they are told that the PQR-cost (i.e. the cost which is justified in order to eliminate this extra dose) is as low as one dollar per year. It is not unlikely that they feel that the risk is indeed insignificant, when they are told that it may amount to an extra lethal risk of the order of 1:1 million per year. We are all subject to higher total risks than we wish to recognize, and we should really not care if our risk of dying in a given year is 1:389.453 or 1:389.301; for all we need to know in that case it is 1:400 - which, incidentally, in many countries is the annual risk of dying at the age of 40.

Now I am getting back to the semantic problem I mentioned when we talked about the word "safe". Obviously, in normal language an extra dose of 10 mrad per year is "safe". But that does not mean that the situation would still be acceptable if all individuals in, for example, the United States received the same extra dose. In reference to their individual risk situation this is still a negligible dose, but the total number of individuals that might be expected to suffer severely from this exposure - provided that our risk coefficient is true which we don't know - is

$$N = P \times D \times C = 250$$

if  $P = 250 \times 10^6$ ,  $D = 10^{-2}$  rad and  $C = 10^{-4}$  per rad.

Of course 250 cases of cancer and severe hereditary defects are not insignificant except in a statistical sense, and the practice that could cause this result is hardly "safe".

Why do we have this paradox, that a practice which causes an insignificant risk to each individual might still be unacceptable if we make a direct risk-benefit assessment of the practice as such? The reason is of course that when we checked the individual's risk we only really checked if the risk was insignificant in relation to his total risk situation and found that it was indeed negligible. We did not bother to check if that negligibly small risk was actually justified from the point of view of the benefit to each individual. That might not have been the case and if so we have no right to expect that the overall benefit from the practice outweighs the harm.

The lesson is that we have a practical risk threshold, below which extra risks are negligible to us as individuals. It takes rather high risks to change our total risk situation significantly. Therefore we may well accept relatively high variations in our radiation background without finding it necessary to check whether the extra doses are really justified. For the society as a whole a similar high risk threshold exists for additional risks to become so high that the harm becomes statistically significant and a social burden. But I maintain that in the risk-benefit evaluation of any given practice the only relevant factors to be compared are the total harm and the total benefit from the practice. If the benefit does not outweigh the possible harm we should be concerned. The degree of our concern should of course increase in proportion to the harm actually expected. If human lives are at stake, I think we are morally and ethically obliged to be worried long before the harm exceeds the threshold that makes it obvious.

Let me now complete the picture I have given of the natural radiation background by putting it in the frame of other exposures to which we are commonly subjected. The dominating one is the medical exposure, the major source being x-ray equipment for diagnostic examinations. According to UNSCEAR, the median value of the genetically significant dose from diagnostic x-ray

examinations is about 20 mrad per year for some 30 studies that have been reported from various countries. Since the genetically significant dose involves weighting for child expectancy and does not include the many exposures of old patients in the averaging, the per caput gonad dose may be twice as high. The per caput mean marrow dose is even higher in some countries.

Although the medical exposures are only partial body exposures, they are given with high dose rates and with extreme individual doses which deviate much more from the average than we have seen for the doses from natural sources. In table 2, I have indicated the significance of the medical exposures by showing you some PQR-costs based on 100 \$ per rad, i.e. without having tried to compensate for the partial body exposure and the higher dose rate, two factors which work in opposite directions.

Table 2: PQR-costs of some medical examinations

Type of examination	Gonad dose mrad	PQR-cost \$
Dental (male)	0.1	0.01
Chest mass survey (female)	3	0.30
Barium meal (male)	30	3
Urography (female)	600	60

It is obvious that the individual significance of a correctly performed dental exposure is negligible, but the large number of examinations may still justify general protection efforts aimed at improving equipment and procedures. A urographic examination for example gives a dose which is not insignificant, and it may be justified to make protection efforts for 30 \$ per examination in order to reduce the dose by 50%. The PQR-cost does not give direct guidance in the risk-benefit assessment; for this purpose the risk coefficients would have to be used.

It is worthwhile recognizing that, while an unnecessary dental exposure has a PQR-cost of one cent, a retake of a urographic film, when the necessary diagnostic information might still have been obtainable from the first imperfect one, spoils protection efforts for a value of 60 \$. If the PQR cost had been an extra tax on the film, many unnecessary exposures might have been avoided. At least one would wish that it becomes an imaginary tax in the radiologist's mind.

With regard to the diagnostic uses of radionuclides I shall only comment upon the frequent use of iodine-131 for thyroid studies. Of a total of about 70,000 radionuclide investigations per year in Sweden about 50% involve the use of iodine-131. These 35,000 examinations cause an average thyroid dose of about 30 rads. The average PQR-cost of a thyroid examination will then be 300 \$ on the basis of 10 \$ per rad since the risk is mainly limited to thyroid cancer. This is not an insignificant radiation burden but many doctors may be ignorant of its significance. The total PQR-cost of the uses of iodine-131 for diagnostic purposes in Sweden is, with these numbers, 10 million dollars per year. At the usual ambition level it would be worth one million dollars per year to reduce the average thyroid dose in Sweden by 10%.

It should be obvious from what I have said, that the justification of any medical exposure must be based upon the result of risk-benefit analyses rather than upon comparisons with any dose limits. It is, however, likely that substantial reduction of doses to patients may be obtained merely by strict adherence to the standards and procedures recommended by ICRP in Publications 16 and 17 on the protection of the patient. The PQR-value of reducing the patient doses in, for example, the United States by as little as 0.1 % seems to be about one million dollars per year.

Of course, risk-benefit assessments should also be made in the protection of the worker. Here, however, the risk-benefit situation is somewhat complicated since the worker stands a direct risk but is usually exposed to the benefit only indirectly, through his salary. It is against normal radiation protection practice to compensate higher doses by higher salaries, since that might eliminate the motivation for caution. On the other hand the protection authorities wish to induce the employers to keep the doses low.

The solution to this problem is to replace risk-benefit assessments by reference to agreed dose limits which guarantee that the individual risk will never be higher than risks which are accepted in other occupations, but to add the requirement that "all doses be kept as low as readily achievable", i.e. to retain the cost-benefit assessment.

To-day, the ICRP dose-limits are rarely exceeded in radiological work. Until relatively recently, nurses and assistants in gynecological radiotherapy departments at some major clinics used to be a group which received high doses because of the difficulties in applying shielding and distance protection. With modern techniques and less frequent use of radium, however, the doses have been much reduced, as you can see from table 3, giving an example from Radiumhemmet in Stockholm:

Table 3: Annual staff doses at Radiumhemmet

Dose range (rad)	Number of workers in each dose range 1961-1971					
	1961	1963	1965	1967	1969	1971
10 - 15	1	-	-	-	-	-
5 - 10	10	6	6	-	-	-
3 - 5	11	13	11	3	-	-
2 - 3	9	12	16	11	9	3
1 - 2	23	27	17	32	33	31
0 - 1	85	132	160	220	261	342

The elimination of the high doses corresponds to about 150 manrads, i.e. a reduced PQR-cost of 15,000  $\text{\$}$  per year after 1971 as compared to the situation ten years earlier. It is symptomatic, however, that this gain is neutralized by additional manrads because of an increased number of employees who work with new sources of radiation in other departments.

If it is now rare that dose limits are exceeded, there are on the other hand some less encouraging observations. Accidents still occur in radiation work, analytical x-ray equipment and equipment for industrial radiography with either x rays or gamma rays being particularly frequent sources. The occupational exposures at nuclear power stations are relatively high and may continue to be so, possibly causing more manrads than the environmental contamination from the same stations.

On the average, however, the occupational exposures are amazingly low, the over-all average being of the order of perhaps 200 mrad per year, causing a whole-population per caput contribution of the order of a few tenths of a mrad.

With this background it is remarkable that the working conditions in mines, and not only in uranium mines, are such that many workers will inhale larger quantities of radioactive particles than the maximum values that can be derived from the ICRP recommendations. Miners are also the only present occupational group for which the cancer rate seems to be clearly correlated to their radiation exposures. Yet non-uranium miners are not even legally classified as radiation workers in some countries, for instance Sweden. The protection measures also take time to put in effect, but slowly give results: (table 4).

Table 4: Radon levels in Swedish mines (from Snihs)

Highest level of radon daughters pCi/l	Number of workers in mines in each category	
	1970	1973
300	21	0
100 - 300	620	0
30 - 100	700	600
10 - 30	2000	2700
0 - 10	1400	1400

It remains for me to close this review by giving some references to environmental contamination. I would like to refer you to the reports of UNSCEAR and ICRP. Even though ICRP recommends dose limits to individual members of the public, it is obvious that there can be no follow up of individual exposures. Radiation protection of the public and control of the environmental contamination in general can only be achieved by control of the sources. This also implies that each source or practice that can lead to radioactive pollution must be subject to both an assessment of the dose to individuals in the critical group and a risk-benefit analysis.

In both cases the annual dose commitments rather than the annual doses should be assessed, since otherwise some long-lived radionuclides may cause non-controllable future exposures.

The dose commitment for any organ or tissue is the infinite time integral of the average value of the mean dose rate in that organ or tissue in the population of interest:

$$D_c = \int_0^t \dot{D}_{\text{average}} dt = \int_0^t \frac{1}{P} \sum_{i=1}^P \dot{D}_i dt$$

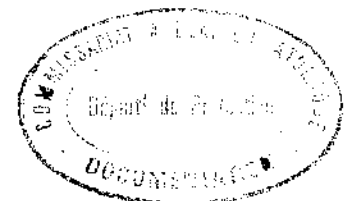
If the population is constant in time, the expected total number of individuals who will be affected by any late deleterious effect will be the product of the population number  $P$ , the dose commitment  $D_c$  and the risk coefficient  $C$ :

$$N = P \times D_c \times C$$

If the radiation exposures from the practice for which the dose commitment is calculated is not limited within any national or geographical borders, it is appropriate to calculate the dose commitment to the whole world population. This has been the practice of UNSCEAR. An alternative way to presenting the dose commitment as such is to present the product  $P \times D_c$ , which product may be called the population dose commitment from the practice and which is measured manrads.

The product of the population dose commitment and the risk coefficient may be called the harm commitment. The harm commitment is the true expectation of total harm from the given practice only to the extent that the assumption with regard to the value of the risk coefficient is correct.

It seems reasonable to expect that nuclear power from light water reactors



can be obtained at an annual population dose commitment of less than one manrad per installed MW of electric power, which will mean only a few millirads per year in year 2000. This, however, is on the assumption that there will be no significant contribution to the population dose by the mathematical expectation of doses from nuclear accidents and that the long-term waste disposal will also be so arranged that it will not contribute significantly to the population exposure.

According to the UNSCEAR reports, nuclear testing in the atmosphere has given a soft tissue dose commitment of about 0.12 rad to the whole world population, which corresponds to a population dose commitment of about 500 million manrads. In terms of either risk or PQR cost this is of little significance to the individual member of the population, the PQR cost per person being 12 dollars, assuming 100 dollars per manrad. The total PQR cost of nuclear testing as such, however, is as high as 50 billion dollars.

We should therefore be grateful to those who succeeded in reaching agreement on the cessation of the heavy atmospheric testing ten years ago. Had testing continued only one more year at the same rate as during the period 1961-1962, it would have caused an additional dose commitment of some 150 million manrads. With the expectation of less than one manrad per MWyear from the normal operation of nuclear power reactors, that corresponds to 150 million MWyears or one thousand years of operation of 150 power reactors at 1000 MW each.

As you have seen, radiation sources differ widely with regard to dose commitment and harm commitments, although we really only know what the harm might be, rather than what it is likely to be. In order to fill in the gaps in our knowledge and to make certain that radiation will be more beneficial than harmful to man in future years, yet another commitment is necessary. We have to commit ourselves to hard work, bright ideas and never-failing attention, just like the pioneers that we owe so much gratitude. Like Rolf Sievert.

## DOSE CALCULATIONS

### DOSIMETRIC ASPECTS OF A FORTHCOMING REPORT OF ICRP ON INTERNAL EMITTERS\*

W. S. Snyder  
Health Physics Division  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830

#### Abstract

The International Commission on Radiological Protection (ICRP) has recommended maximum permissible annual doses (MPAD) as a basis for protection of radiation workers. Committee 2 of ICRP attempts to provide secondary standards on annual intake which are based on these MPAD. This committee plans to issue a new handbook with complete text, but the first edition will contain only the radionuclides of some twenty elements which have been selected partly on the basis of their importance and partly to illustrate the method of calculation.

Some new features of the publication are the following: (1) If an intake of radioactivity occurs in a certain year, the total dose from this intake is attributed to this year, and this principle of dose commitment insures that workers do not become "unemployable" in radiation work due to early intake of a long-term emitter. (2) A new lung model provides more detailed information on the deposition and clearance rates of inhaled particulates and includes an adjustment of deposition with the activity median aerodynamic diameter. (3) Doses to active bone marrow and to endosteal cells are estimated for beta and gamma emitters depositing in bone. The methods of Spiers are used for these estimates. For alpha emitters, the Committee will continue to use the relative damage factor  $N$  pending more detailed information on the distribution of such emitters in the skeleton. (4) Dose from photons released in a large cloud of radioactivity is estimated by computing depth dose within the body by Monte Carlo techniques. (5) The report will use more accurate decay scheme data and improved retention models, and dose from photons will be estimated more accurately than in the old report.

---

To predict the course a committee may take is to predict the unpredictable, and this is especially true when one can only offer his own judgments. Nevertheless, my topic is that of expounding the dosimetric methods developed for the new report of ICRP Committee 2. Of course, the Committee can always change its direction or use a new bit of data that comes along, and thus it should not be cause for surprise if the judgments offered here turn out to be wrong sometimes. However, many of the dosimetric techniques the Committee plans to use have been developed at Oak Ridge National Laboratory by me and my colleagues, and this is what I will be speaking of for the most part.

---

\* Research sponsored by the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.



Present plans call for the Committee to publish an initial volume early in 1974 which is to contain a complete text and the procedure for dosimetry. This same report will contain recommendations on a certain number of radionuclides which will be chosen to illustrate the principles given in the text but which also are the more important ones for radiation protection. First of all, let us review the new recommendations that will be embodied in the report. These are:

- (1) The principle of dose commitment is used as the controlling factor for occupational exposure to radionuclides of long half-time in the body. This principle is implicit in ICRP Publications 1 and 2,<sup>1-2</sup> but its use explicitly results in greater simplicity in that it allots to the year during which an intake occurred all the dose to be received from that intake during the worker's remaining life span (conventionally taken as 50 years). This is a simple recognition of the fact that the consequences of an early accumulation of a long-term body burden should be attributed to the operation which led to this intake. The principle that such intakes are not to be considered as routine is a healthy one for radiation protection.
- (2) The total body as an organ supposedly uniformly exposed is de-emphasized, because this rarely, if ever, occurs. The average dose equivalent to the active bone marrow and the dose equivalent to endosteal cells lying near bone surfaces have been indicated as criteria for exposure of bone. In effect, these tissues replace bone insofar as one can implement the recommendation. The technical difficulties of estimating dose to these tissues will be discussed further below.
- (3) The use of 1 as the value of  $Q$  for electrons and beta rays of low energy instead of 1.7 as used formerly is a simplification. Now electrons are on a par with photons since, for all these, dose is numerically equal to dose equivalent.
- (4) Dose equivalent to pulmonary lymph nodes is not considered at this time. This decision of the Commission is based on biological evidence which, at present, indicates that exposure by inhalation appears to produce many more malignancies of lung tissue than malignancies associated with lymphatic tissue. Undoubtedly, experiments now in progress will be watched to assess more accurately the hazard to the lymph nodes.

There are other changes in the basic recommendations, and some of them will be referred to below; but none of these is as closely involved in the dosimetry of internally deposited emitters as are the foregoing. There will be many changes in retention models as will be seen, these applying for the most part to the early period postintake and, hence, being important principally for interpretation of excretion data or for radionuclides of short radiological decay times. Fortunately, relatively easy access to a whole body counter has made it possible in many cases for the health physicist to feel that he can determine intake and even retention to some extent, and this is a great help in meeting the recommendations which place considerable emphasis on control of intake. Unhappily, there are still radionuclides which cannot be measured with sufficient accuracy for the routine cases, and these offer considerable challenge to health physicists who must deal with them.

Several ICRP recommendations on internal emitters have been published since ICRP Publication 2 appeared in 1959, the most extensive being those in ICRP Publication 6.<sup>3</sup> Also, ICRP Publications 10<sup>4</sup> and 10A<sup>5</sup> provide some help in meeting practical problems. However, it is a tribute to Publication 2 that it has served so well for so many years.

A new model for deposition and retention, or clearance of particulates from the respiratory system, was formulated by the ICRP Task Group on Lung Dynamics (chaired by P. E. Marrow), and this was first published in *Health Physics* in 1966.<sup>6</sup> Although some of the parameters of the model have been revised several times since its first appearance, the model remains basically the same. The most notable of these revisions affects the clearance from lymph nodes. This had been conservatively chosen so that 90% of the deposition in these organs was eliminated only by radioactive decay and 10% with a long biological clearance time, as was indicated by the biological data available at that time. Since then, the long-term studies of dogs exposed to an aerosol of natural uranium<sup>7</sup> have given an indication that there is clearance from the lymph nodes, and the model has been revised so that now the situation is just the reverse--90% clearing with a long biological half-time and 10% eliminated only by radioactive decay. This is a striking example of how ICRP models are formed and changed as new data accumulate. At the moment, the Committee intends to average dose over the lungs as formerly. No doubt when the question of the lymph nodes is reconsidered and when the model is complete for gases and vapors, as well as for particulates, the question of reassessing doses to subregions of the lung will be reconsidered also.

A procedure for computing the time integral of activity ( $\mu\text{Ci-days}$ ) of the nuclide and of its daughters in the various subregions of the lung is derived, and the dose equivalent,  $H_k$ , to organ  $k$  is calculated by the formula

$$H_k = \sum_i \sum_j U_{ij} \times (\text{SEE})_i (k-j) \text{ rem}/\mu\text{Ci intake} \quad (1)$$

where  $U_{ij}$  is the time integral of activity of daughter  $i$  in the lung or other source organ indexed by  $j$ , and  $(\text{SEE})_i (k-j)$  is the specific effective energy for the  $i^{\text{th}}$  daughter with source organ  $j$  and target organ  $k$ , that is, the energy absorbed per gram in the target organ  $k$  per disintegration of the  $i^{\text{th}}$  daughter in the source organ  $j$ . In computing the  $(\text{SEE})_i (k-j)$ , the absorbed fractions of photon energy are computed as in MIRD Pamphlet No. 5.<sup>8</sup> In principle,  $j$  may be any organ of the body, and it is one of the unusual features of this dosimetry that cross irradiation of an organ by photons emitted in other organs is taken into account. The decay scheme information has been processed by the methods of Dillman<sup>9</sup> so full account is taken of internal conversion, Auger electrons, and other particles emitted. The  $U_{ij}$  are computed assuming the aerodynamic mean activity diameter (AMAD) is  $1 \mu\text{m}$ , but a procedure is indicated which makes it possible to adjust the dose to any desired particle size in the respirable range. The dose equivalent per  $\mu\text{Ci}$  inhaled will be given in the projected report for all organs for which the dose equivalent is 10% or more of its annual dose limit when the critical organ just attains its dose limit. An attempt will be made to list genetic dose especially, even though it may be only 1% of its limit, but the adequacy of the model will be taken into account also, that is, one requires that the biological information be reasonably adequate for activity contained in the gonads as well as for the surrounding organs.

The Committee intends to list also the maximum permissible annual intake (MPAI) by inhalation and the derived working level (DWL) which is the former  $(\text{MPC})_a$  in disguise. The reason for these changes is to emphasize that it is the annual intake and the dose commitment due to it that are basic in the Commission's recommendations, while there is no clear violation involved if the DWL is exceeded by a factor of 10 for a day or so. The report also will contain estimates of MPAI for intake by oral ingestion. However, no equivalent of the  $(\text{MPC})_w$  will be given, since the situation where the supply of drinking water is contaminated significantly rarely occurs in occupational exposure. In any case, the MPAI is the criterion for protection, and an equivalent concentration is easily obtained if it is wanted.

It will be noted that formula (1) only involves two variables, the  $U_{ij}$  and  $(SEE)_i(k-i)$ . The estimation of the first of these involves the model for retention and is computed for each daughter element and for each source organ, here indexed by  $i$  and  $j$ , respectively. This requires that one have a retention model for each daughter element as well as for the parent. Biological data on these daughter elements is sparse indeed, and for many elements one will be reduced to guessing the behavior of the daughter, usually assuming it will remain in the organ where it is produced subject to elimination as would material recently deposited in the organ. Thus, if

$$\sum_i a_i e^{-\lambda_i t} \quad (2)$$

is the retention function for a daughter element in bone or liver or some other organ when the element is injected into blood, then

$$\sum_i a_i e^{-\lambda_i t} / \sum_i a_i \quad (3)$$

will be the retention function for that element when it is produced in the organ as a daughter element. There are a few elements where some biological data indicate exceptions to this, but they are very few. In fact, the question has hardly begun to be explored by the biologists. For most daughter elements, formula (3) is the basis for the dosimetry of that element when it is produced in an organ.

The estimation of  $(SEE)_i(k-i)$  involves a chapter of its own, beginning with the Task Group on Revision of Reference Man. This report, which is in press, gives organ weights and other biological data needed for dose estimation. Most of these data are embodied in a mathematical phantom which has been used to estimate absorbed fractions of photon energy for various source and target organs.<sup>8</sup> The phantom representation approximates the size, shape, density, position, and elemental composition of the various organs. A computer code uses this information to produce the specific effective energy, and in a sense the use of SEE values replaces the effective energies and F values of Publication 2. The decay scheme data used have been produced by the method of Dillman as was noted above.

Explicit formulae are given for the time integrals of retention in the gastrointestinal tract. The basis for this procedure is the report of Eve<sup>10</sup> which is almost unchanged except for new data on the masses of the small intestine and contents. Then dose is given by formula (1) as before. For photons, the absorbed fractions are computed using the anthropomorphic phantom described above, and dose from electrons and from alpha particles represents only a surface dose as in ICRP Publication 2. The inclusion of a modifying factor of 0.01 for alpha radiation originating in the contents of the tract is continued, since it rests on rather firm biological evidence indicating that the hazard of irradiation by alpha particles emitted in the contents of the tract is very small.<sup>11</sup> The Committee plans to give a dose per  $\mu\text{Ci}$  of intake to organs with the same limitations on dose equivalent as for inhalation.

The dose equivalent for other organs is given by the same formula (1) as before, although the time integral of activities for the source organs will depend on the retention functions adopted. These retention functions are being completely re-examined and revised by the Committee. No attempt is being made to reduce them to a common form since each element will be treated separately on a few pages of the report, and thus the retention models will be independently developed. In each case there will be tables indicating the time integrals of retention and the SEE values for various source and target organs, and these will be given for the radionuclide and for each daughter element as required.

The only organ where the dosimetry has considerable novelty is the skeleton with its intricate intermixture of bone and bone marrow. Here the Committee is adopting the methods developed by Spiers for beta emitters.<sup>12</sup> Spiers has supplied to the Committee his latest estimates of the dose received by active bone marrow and by endosteal cells near cancellous bone or near cortical bone from a number of beta emitters of a variety of mean energies.<sup>13</sup> For other betas and electrons, the Committee will interpolate on the mean energy or unique energy of the particle to estimate the dose.

Unfortunately, Spiers' estimates are rather different when the source is in cancellous bone and when it is in cortical bone, and thus one should have independent estimates of the time integrals of activity in these two types of bone. There are very few radionuclides which deposit in bone for which we have this data, and these are nearly all due to the efforts of Jahn Marshall, Chairman of the ICRP Task Group on Alkaline Earth Metabolism in Adult Man.<sup>14</sup> Marshall and his task group have supplied to the Committee their estimates of the time integrals of activity for all the isotopes of Ca, Sr, Ra, and Ba. This is a notable contribution and represents years of work, yet it is only a beginning. Clearly such estimates are needed for all bone-seekers, and this puts the burden squarely on the shoulders of the experimental biologist who must undertake the arduous task of documenting the distribution of the radionuclide in bone in considerable detail. In the absence of this data, the Committee is planning to consider all the activity to be present in cancellous bone. For most radionuclides of short radioactive decay time, this may not be far from the truth, but, clearly, better data are needed.

For alpha emitters depositing in bone, the Committee is forced, for lack of adequate data on distribution of the radionuclide in bone, to give up for the moment any attempt to calculate dose to the endosteal cells. Thus the dosimetry of the alpha emitters remains essentially that of ICRP Publication 2--namely, average dose equivalent in bone and use the N factor for the various radionuclides. The value  $N = 5$  used previously still seems reasonably adequate and is retained. The ICRP Task Group on Metabolism of Plutonium and Related Elements and Their Compounds, chaired by Arthur Lindenbaum, has altered considerably the distribution of activity by recommending that deposition in the liver and in skeleton be considered as being equal. This may result in making liver the critical organ for some radionuclides.

In Publication 2 the modifying factor N has been given the value  $N = 1$  for radium when it is the parent. This is because  $^{226}\text{Ra}$  was considered a standard for bone-seeking radionuclides, and the carcinogenic potency was thought to be related to the distribution of dose within the skeletal system. However, the human data of Spiess and Mays,<sup>15</sup> as well as the animal data of Hug,<sup>16</sup> indicate that  $^{224}\text{Ra}$  is a more effective carcinogen than is  $^{226}\text{Ra}$ . The Committee plans to use the factor  $N = 5$  for the radiums of short radioactive decay times when they are the parent radionuclide.

There remains immersion dose, that is, the dosimetry associated with a person exposed to a semi-infinite cloud of a radionuclide. Here the essential step is due to Dillman<sup>17</sup> who has estimated the energy spectrum of the photons in such a cloud and has also given us the depth dose from the electrons and from beta rays.<sup>18</sup> Finally, he has developed the spectrum of bremsstrahlung produced by the betas. All of these spectra are allowed to impinge on the anthropomorphic phantom developed by the Oak Ridge group, and so for the first time we have dose distributed in depth in such a phantom from a cloud source and also estimates of dose to the various organs.

There are many other facets of the report as planned which remain much the same as previously, and the fact that they are not mentioned here is not to be construed to mean they

are omitted. Clearly in the allotted time, one can only mention principal points, and one can only hope the new report will serve health physicists as well as did ICRP Publication 2.

#### References

1. Recommendations of the International Commission on Radiological Protection (Adopted September 9, 1958) (Pergamon Press, Belfast, 1959).
2. ICRP Publication 2, Report of Committee II on Permissible Dose for Internal Radiation (1959) (Pergamon Press, Great Britain, 1959).
3. ICRP Publication 6, Recommendations of the International Commission on Radiological Protection (As Amended 1959 and Revised 1962) (Pergamon Press, Glasgow, 1964).
4. ICRP Publication 10, Report of Committee IV on Evaluation of Radiation Doses to Body Tissues from Internal Contamination due to Occupational Exposure (Pergamon Press, Exeter, 1968).
5. ICRP Publication 10A, The Assessment of Internal Contamination Resulting from Recurrent or Prolonged Uptakes (Pergamon Press, Exeter, 1971).
6. ICRP Task Group on Lung Dynamics, "Deposition and Retention Models for Internal Dosimetry of the Human Respiratory Tract," Health Phys. 12, 173 (1966).
7. L. J. Leach et al., "A Five-Year Inhalation Study with Natural Uranium Dioxide (UO<sub>2</sub>) Dust--I. Retention and Biologic Effect in the Monkey, Dog and Rat," Health Phys. 18, 599 (1970).
8. W. S. Snyder et al., "Estimates of Absorbed Fractions for Monoenergetic Photon Sources Uniformly Distributed in Various Organs of a Heterogeneous Phantom," J. Nucl. Med. Suppl. No. 3: 10, 5 (August 1969).
9. L. T. Dillman, "Radionuclide Decay Schemes and Nuclear Parameters for Use in Radiation-Dose Estimation," J. Nucl. Med. Suppl. No. 2: 20 (1969).
10. I. S. Eve, "A Review of the Physiology of the Gastrointestinal Tract in Relation to Radiation Doses from Radioactive Materials," Health Phys. 12, 131 (1966).
11. M. F. Sullivan et al., "Irradiation of the Intestine by Radioisotopes," Rad. Res. 13, 343 (1960).
12. F. W. Spiers, Radioisotopes in the Human Body (Academic Press, New York, 1968).
13. F. W. Spiers, Private communication.
14. ICRP Publication 20, Alkaline Earth Metabolism in Adult Man (Pergamon Press, Exeter, 1973).
15. H. Spiess and C. W. Mays, "Bone Cancers Induced by <sup>224</sup>Ra (Th X) in Children and Adults," Health Phys. 19, 713 (1970).
16. O. Hug et al., Radiation-Induced Cancer (IAEA, Vienna, 1969), edited by A. Ericson.

17. L. T. Dillman, "Scattered Energy Spectrum for a Monoenergetic Gamma Emitter Uniformly Distributed in an Infinite Cloud," Health Physics Division Annual Progress Report for Period Ending July 31, 1970, ORNL-4584, p. 216.
18. L. T. Dillman, "Depth-Dose Rates in Reference Man from Electrons and Beta Particles in a Radioactive Cloud," Health Physics Division Annual Progress Report for Period Ending July 31, 1971, ORNL-4720, p. 125.

STATISTICAL APPROACH IN DEVELOPING MATHEMATICAL MODELS  
FOR EVALUATING INTERNAL IRRADIATION

V. I. Bad'in, S. A. Andronov, A. P. Ermilov, V. T. Khrushch,  
U. Ja. Margulis

Institute of Biophysics, Ministry of Public Health  
Moscow, USSR

Abstract

It is established that calculation of radioactive intake (or accumulation) into the body according to the controlled dosimetric parameter requires some statistic data about specific peculiarities of the investigated object.

This paper presents data about statistical distribution functions of the radionuclide concentration ratio at the sampling point to its concentration at the respiration zone, protective efficiency of individual protective means, elimination of the radionuclide from the body and others; it gives an estimate of the errors for determining an inhalation intake of radioactivity into the body.

Various techniques for controlling an internal radiation exposure is analysed.

1. Basic environmental data

In order to estimate accumulation of radioactive substances within the body of people by data on the contaminated air it is necessary to know aerosol characteristics averaged over human locations during the working shift with due account for the efficiency of individual protective means. But the necessary qualitative and quantitative data are usually fully lost when stationary samplers are used. This is confirmed by the tabulated data. The data obtained from stationary aerosol radiometers approach those from individual air samplers for rooms which lack local powerful sources of air contamination (Table 1). Otherwise discrepancy of the data will be by the order of several tens.

Table 1\*

Comparative estimate of the data obtained from stationary and individual samplers

Characteristics of the air contaminating sources	The ratio of the data obtained from an individual sampler to those from a stationary one (the log-normal concentration distribution law)			
	mean	maximum (seen de facto)	median	standard geometric deviation
Local sources of initial contamination are available	24	500	12	3.2
No sources of initial contamination	2.5	5	1.8	2.3

\* Both for this Table, and for Table 2-4 there were usual statistical methods to estimate the statistical confidence of data.

Efficiency of individual respiratory protective means is of great importance for calculating the value of inhaled radioactivity.

Table 2 shows the practical decrease rate in contamination of the inhaled air by a respirator of the "Lepestok" type. Both the relatively small efficiency of respirators for low contaminated air and the better efficiency (comparing to the value founded in the laboratory) for the higher level of air contamination are resulted from time of using of respirators during one working day. ("Lepestok" is the respirator of the simple type).

Table 2

Efficiency of the respirator "Lepestok"

The level of air contamination	The decrease rate in contamination of the inhaled air (follows the log-normal law)					
	in real conditions				in laboratory experiments (according to S. M. Gorodinsky)	
	average	maximum	median	standard geometrical deviation	average	median
Below APC	24	300	12	3.2	680	450
Above APC	350	2000	160	3.6		

Therefore both the stationary samplers and individual ones have some positive or negative aspects, but can't used in order to estimate the real individual intakes of airborne radioactive substances. This conclusion will be also confirmed by next discussion.

Dispersity of alpha-active airborne particles deposited at the external nasal orifices and present in the inhaled air and in the air of working premises is estimated in Table 3, where the following designations are accepted:

$r_g$  - geometrical mean radius,

$r_{max}$  - maximum radius of the airborne particle in the sample.

Geometrical mean radius of the log-normal distribution is determined by the ratio

$$\ln r_g = \frac{1}{\sqrt{2\pi} \ln \beta} \int \ln r \cdot \exp \left\{ - \left( \frac{\ln r / r_g}{\sqrt{2} \ln \beta} \right)^2 \right\} d(\ln r) = \frac{1}{n} \sum \ln r_i; \lg r_g = \frac{1}{n} \sum \lg r_i$$

$\ln \beta$  - standard deviation of the radius logarithm;  $\lg \beta = \frac{1}{2.3} (\lg r - \lg r_g)^2$

$r_g$  - geometrical mean activity radius for the non-limited log-normal distribution.

$r_g(\text{lim})$  - geometrical mean activity radius for the limited log-normal distribution

$$r_g(\text{lim}) = r_g \cdot \beta^{-\left( \frac{\ln r_{max} - \ln r_g}{\sqrt{2} \ln \beta} \right)^2} \quad (1), \quad r_{am}(\text{lim}) = r_g \cdot \beta^{\varphi_i^{-1}} \quad (2)$$

$$\xi'_i = \frac{1}{\lg \beta} [\lg r_{max} - (\lg r_g + 2.3) \lg \beta], \quad \varphi_i = 0.5 [\Phi(\xi'_{i2.3}) - 1]$$

$$\Phi(\xi'_i) = \frac{2}{\sqrt{2\pi}} \int_0^{\xi'_i} e^{-\xi'^2/2} d\xi' \quad \text{- Gauss probability integral}$$

$r_{am}(\text{lim})$  - activity median radius with account for the limited log-normal distribution



Estimation of the confident interval indicated that with the level of significance 0.05 the intervals were not overlapped. This result is caused by the difference in the size distribution of airborne particles collected by three various techniques. Neither Fisher relation is followed. Therefore the difference of the mean standard deviations should be considered significant.

Table 3

Dispersivity of airborne particles

Collection technique	Averaged dispersivity of airborne particles					
	$\beta$	$r_{\beta}, \mu m$	$r_{max}, \mu m$	$r_{ag}, \mu m$	$r_{ag}(lim), \mu m$	$r_{am}(lim), \mu m$
Stationary sampler	2.4	3.8	44-120	39	20-32	20-39
Individual sampler	3.2	1.3	21-34	70	11-17	12-19
Smears from the nose	4.3	0.53	15-32	290	17-20	18-20

On the other hand limited aerodynamic activity radiuses with permissible dosimetric error coincide (Table 3). Therefore the activity fraction settled at various parts of the respiratory tract calculated by AMAD<sup>1</sup> will be equal. Individual protective respiratory devices are thought practically not to modify dispersivity of the inhaled dust in the case of coarse-grained particles. Table 4 gives comparative results obtained by three various techniques in the course analysis of the daily inhalation intake for 3 groups of workers.

Table 4

Comparison of various techniques for determining the daily inhalation intake A by the average values in terms of A<sub>11</sub> for the use of individual protective means

No	Technique for determining the intake	The group of workers						The number of analyses for all groups	Note
		1		2		3			
i		A <sub>1i</sub>	B <sub>1i</sub>	A <sub>2i</sub>	B <sub>2i</sub>	A <sub>3i</sub>	B <sub>3i</sub>		
1	Analysis of excretions	1	5.9	0.97	4.8	0.63	5.2	131	With account for solubility of dust determined by experiment
2	Smears from the nose	0.83	2.8	1.1	3.7	6	5.3	77	With account for dispersivity of dust determined by stationary samplers
3	Individual samplers (without account for IPM)	160	2.4	450	4.3	790	3.2	94	Assuming the volume of the inhaled air/10 m <sup>3</sup>

$\beta_i$  - corresponding  $A_{ji}$  distribution standard geometrical deviation.

Therefore it is believed reasonable that the value of individual inhalation intake (or accumulation) into the body should be calculated either by the indications of smears taken from the external nasal orifices or by the excretion dates, whereas activity distribution over the respiratory tract may be estimated with due accuracy by means of stationary or individual samplers (if dispersity is correctly averaged by stationary samplers). The last remark is important for those working rooms where dispersity of airborne particles is changed as a function of working locations and the type of technological operation and consequently the level of dust penetrating the lungs is also changed.

Thus, determination of actual accumulation of activity within the body due to inhalation requires both measurement of individual inhalation intake and one of dispersity of the inhaled aerosols by a direct method. Determination of dispersity can be substituted by the estimation of the relative value of penetrating airborne particle fraction. Practically the most suitable is combination of the method for determining the intake by the smears from the nose and selective individual samplers provided with presettler.

If these requirements are not fulfilled the use of average values may lead to errors in calculating the individual intake by several orders of magnitude:

- up to 20 times due to disparity of stationary and individual sampling;
- up to 10 times due to errors in determining dispersity of the inhaled dust and up to 10 + 100 times due to the differences in real effectivity.

Approximate lognormal space-time distribution statistics for radioactive concentration in the air of working premises is currently given much consideration in literature<sup>2</sup>. Due to logarithmically normal fluctuations in the protective effectivity of individual protective means and due to generally random time schedule of work the above stated factors even after averaging over long time periods (calendar year) lead to the actual radioactive intake to individuals from the homogenous group being described by the lognormal distribution with the significant standard geometrical deviation ( $\beta$ ). We found that  $\beta$  in this case can amount to 2.5-7. It should be emphasized that the groups should be clearly enough classified according to the radiation situation (by profession, by location and time, etc.). Otherwise the standard geometrical deviation will be much increased (up to 10 and more).

## 2. Basic human data

We believe that the lognormal distribution law of radioactivity eliminated from the body with urine and feci is an important half-empirical consideration (Table 4). This law is determined by both the lognormal intake pattern and the statistical character of metabolism within the body.

It will be reasonable to consider the statistical similitude principle as a general assumption for describing metabolism of radioactive substances within the body:

- the ratio  $X_i$  (radiation burden within the body or in some part of the body to a single intake "i" physiological cycles after intake) is a random value not depending on the intake value.

This makes it possible in many cases to describe the result of the  $i$ -th cycle in the form of the proportional effect law:

$$X_i - X_{i-1} \cong \varphi_i X_{i-1} \quad (3)$$

Here  $\varphi_i$  - a random function, if to apply to it reasonably general limitations and to use the central probability concept ultimate theorem it is possible to obtain an expression for the large values of "i" in the form of the lognormal distribution law of the value  $X_i$ . E. g. if to describe elimination of the substance from the body organ one may obtain:

$$P \left\{ \frac{\ln X_i - \ln(x_0 e^{-\lambda i})}{\sqrt{i} \sigma} < y \right\} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^y e^{-\frac{v^2}{2}} dv \quad (4)$$

Where  $P$  - probability of inequality taken in brackets;  
 $x_0$  - the initial radioactive quantity in the organ;

$\lambda = -\ln(1 + \varphi)$  - if to assume that  $\varphi$  is not dependent on  $X$  but is a random value with dispersion  $\sigma^2$ . With other initial conditions one may obtain a different expression for the lognormal distribution median (4) which in the general form is expressed as a function of the number of physiological cycles  $f(i)$ . It is difficult to obtain and consider the function  $f(i)$  in the general form. Therefore currently it is reasonable to use the empirical fact on the lognormal distribution law of  $X_i$  values around  $f(i)$  and a constant value  $\beta$  of the standard geometrical deviation of the same individuum.

For example we found that 5-10 days after removal of the workers from the "hot" laboratory the ratio of the daily elimination of Po-210 for the previous day to the daily elimination for the next day fluctuates around the median value  $\approx 1.0$  following the lognormal distribution with the standard geometrical deviation  $\approx 2.7$ . This agrees with the standard geometrical deviation of the primary value  $X_i$ ;  $\beta = 2.0$ .

Practically the same standard geometrical deviation is probably obtained when individual fluctuations of Pu-239 elimination from the body are estimated. W. S. Snyder<sup>3</sup> found that in 60% of cases of the plutonium daily elimination the difference from the "individual" curve was not more than two-fold and in 80% of cases it was not more than three-fold.

In addition to these metabolic data for polonium and plutonium one cite the experimental results obtained on operation Roller Coaster<sup>4</sup> when the standard geometrical deviation in the aerosol respiratory retention factor was  $\approx 2.0$ .

We found that after a single intravenous or intratracheal injection of Po-210 to rabbits such major body organs as kidneys, liver, spleen and lungs contain various amounts of the radionuclide. This variation may be characterized by  $\approx 2.5$ . With due correction for individual differences of animal species the result obtained may be interpreted as it was stated earlier thus assuming that if similar measurements could be made repeatedly on the some rabbit the lognormal distribution of the results would be obtained  $\approx 2.0$ .

Summarising the cited above experimental data it may be concluded that all the cases being the result of relatively rapid physiological processes can be described (within the same individuum) by the lognormal fluctuation law with the standard geometrical deviation  $\approx 2.0$ . This value  $\beta$  can be considered as basic for all other statistic estimations obtained by monitoring internal radiation exposure.

However it should be noted that the lognormal distribution

cannot be accepted directly as the main probability law for describing most parameters estimated in the course of various methods used for monitoring internal radiation exposure. Thus, we showed that even when the size distribution of radioactive airborne particles was fully suited by the mathematical equations of A. N. Kolmogorov's theory<sup>6</sup> it was necessary to account for limitation of the lognormal distribution from the side of large values. For other radiation parameters the use of the lognormal distribution may be considered valid for the range of 95-99% of cases where it should be treated as a convenient and simple analytical expression.

As a result of the data discussed above we accept the following values of the standard geometrical deviations for primary processes, which are superposing into real situations discussed below:

- A** =2.0 - retention in the respiratory organs;
- A** =2.0 - fraction of the substance entering the organ;
- A** =2.0 - elimination of the substance with urine and feci;
- A** =1.6 - difference in AMAD of the inhaled airborne particles;
- A** =3.2 - difference in the local concentrations;
- A** =3.6 - difference in the protective efficiency of respirators;
- A** =7.0 - difference of the annual individual intake;
- A** =5.0 - difference of the radioactive releases with account for accidental situations.

The choice of the last value is purely subjective assuming that the release which results in the median average for the day radioactive concentration being exceeded by 1000 times corresponds to 0.01% of cases.

### 3. Discussion

Practically any dosimetric control aims at obtaining such result which could provide an unequivocal answer about the degree of individual risk, e. g. internal irradiation of man at a certain time moment. The result obtained by control is compared to the standard values. Basing on the data<sup>7, 8</sup> critical organs of the group (whole body, hemopoietic organs) the following limiting radiation dose values may be given:

- 1) annual permissible radiation dose for individuals from the population - 0.5 rem;
- 2) annual radiation dose for occupational workers who are not subjected to individual control - 1.5 rem;
- 3) annual permissible radiation dose for occupational workers - 5 rem;
- 4) permissible accidental radiation dose - 12 rem;
- 5) accidental radiation dose requiring subsequent medical examination - 25 rem;
- 6) accidental radiation dose which does not result in detectable immediate somatic effects - 75 rem;
- 7) sublethal radiation dose - 200 rem;
- 8) accidental radiation dose with a possible death in the absence of medical aid - 400 rad;
- 9) accidental radiation dose with survival of people only in case of intensive and immediate medical aid - 800 rad;
- 10) accidental radiation dose which permit survival in case of intensive and immediate medical aid - 1200-1500 rad.

Basing on these limiting dose values we established 11 ranges, i. e. ranks of radiation hazard. The philosophy for interpreting each of these ranks is different but now we are most in-

terested in the practical aspect of the problem, i. e. how effective the monitoring itself will be from the point of view of possible errors. As one of the specific methods for monitoring we may consider calculated prediction of radiation situation for design objectives.

Currently existing methods for monitoring external gamma-neutron radiation are usually fairly precise to provide unequivocal identification of the hazard rank by the result obtained. In the worst instance one may overestimate or underestimate the actual hazard rank not more than by 1. As for the results obtained by monitoring internal irradiation the situation is quite different even if to consider the optimum result equal to the average geometrical value of the lower and upper limits of the corresponding radiation dose range.

Table 5 gives conventional classification for the situations which occur at monitoring internal radiation exposure. The situations considered are characterized: by the corresponding standard geometrical deviation of the monitoring result from the possible real value; by the probability in per cent corresponding to the boundary values of a hazard rank; by the number of ranks comprising 99% of cases. The adjacent limiting radiation dose values listed above differ from each other not more than threefold. This maximum value is accepted for estimating the probability which corresponds to one hazard rank, i. e. the monitoring result differs from the corresponding range limits by  $\sqrt{3}$  times. The number of ranks comprising 99% of cases was estimated relative to the result which corresponded to the 6th hazard rank. The resulting standard geometrical deviation was determined by the equation:

$$\beta = \alpha p / \sqrt{\sum (\ln \beta_k)^2}$$

where  $\beta_k$  - standard geometrical deviations characterizing the primary processes cited at the beginning. The third column of Table 5 lists those  $\beta_k$  which were considered for the given situation.

Situation 11 (designed calculation) was estimated somewhat differently. It was supposed that the calculation was based on the average annual permissible concentration with the safety margin factor of 10. This calculated value evidently cannot be assigned to the 6th hazard rank but will be at the 1st one of the average annual value is taken for occupational workers. Therefore the probability in the last but one column was calculated for the ranks 1 and 2.

Of course, Table 5 is relatively limited. In addition the parameters for situation 11 were taken on the basis of subjective choice. However these data do not overestimate the values of the resulting difference.

The estimation of the hazard rank may be more complicated in a number of practical situations which were not considered here. For instance, there are other routes of intake in addition to inhalation. The errors associated with measuring techniques and a limited accuracy in description of metabolic processes were not accounted for also.

In this connection the estimates given in two last columns of Table 5 are very important. In neither of the situations considered the actual individual hazard of internal irradiation can be assigned unequivocally to one hazard rank. In a number of cases (situations 1, 2, 4, 7b, 10) the 99% range comprises 10-11 hazard ranks, i. e. the whole hazard scale. Thus, it is clear that a statistic estimate is an integral whole of the problem

for providing radiation safety of people. The authors deliberately neglected the fact that the original standard data included their own safety margin factors. These factors do not influence the quantitative estimate of the situation.

Conclusion

We believe that a statistic difference found in monitoring internal irradiation of people should be given a quantitative estimate of a wide usage. It will be possible only after international agreement on the main values. As the first step in this direction the authors suggest to introduce a concept of metabolism fluctuation into the characteristics of a standard man and to accept the standard geometrical deviation 2.0 for estimating radionuclide elimination rate fluctuation.

Table 5

Different situations found in monitoring internal irradiation

No i	Situations	Primary standard geometrical deviations $\beta_1$	Resulting standard geometrical deviation $\beta$	Probability corresponding to the ranges of one hazard rank, %	The number of hazard ranks comprising 99% of cases (at the 6th ranks)
1	Estimate of the average annual content by a single measurement with a whole body counter for rapidly eliminated radionuclides	$\beta_1, \beta_2$	8.0	21	11
2	Monitoring a single intake by an individual sampler when the respirator of the "Lepestok" type is used	$\beta_1, \beta_2, \beta_3, \beta_4$	7.6	21	11
3	Monitoring a single intake by a stationary sampler when the respirator of the "Lepestok" type is used	$\beta_1, \beta_2, \beta_3, \beta_4$	10.0	19	11
4	Monitoring a single intake by a stationary sampler in the absence of individual protective means	$\beta_1, \beta_2, \beta_3, \beta_4, \beta_5$	4.9	29	10 (N2+N11)
5	Monitoring a single intake by an individual sampler in the absence of individual protective means	$\beta_1, \beta_2, \beta_3$	3.0	38	7 (N3+N9)
6	Estimate of the single inhalation intake of radionuclide by several results of the complex bioassay	$\beta_2, \beta_3$	2.7	42	7

No i	Situations	Primary standard geomet- rical devia- tions $\beta_{\Sigma}$	Resulting standard geometri- cal devi- ation $\beta$	Probabili- ty corres- ponding to the ranges of one ha- zard rank, %	The number of hazard ranks com- prising 99% of ca- ses (at the 6th ranks)
7	Estimate of the radio- nuclide content in the lungs by a timely sin- gle analysis of urine and feci	$\beta_2, \beta_3$ $\beta_4$	3.0	38	7 (N3+N9)
8	Estimate of the radio- nuclide content in the body (except the lungs) by a single analysis of urine and feci: a) at the time of me- asurement b) on the average for a year	$\beta_1, \beta_2$ $\beta_2, \beta_3, \beta_4$	2.7 9.0	42 20	7 (N3+N9) 11
9	Monitoring a single in- take by smears from the nasal orifices	$\beta_2, \beta_4$	2.3	49	5 (N4+N8)
10	Monitoring a single in- take by smears from the nasal orifices combined with a selective indi- vidual sampler	$\beta_2$	2.0	57	5 (N4+N8)
<u>For example</u>					
	Project of stationary aerosol protection ba- sed on the average an- nual permissible con- centration (with aver- aged 10-fold safety margin)	$\beta_1, \beta_2$ $\beta_3$	8.3	99	2 (N1, 2)

#### Literature

1. Health Phys. v. 12, N 2, pp. 173-207 (1966).
2. Radiation dose measurements. Proc. Symp. Stockholm, 1967. ENEA, Paris, 1967. A. I. Broslin, pp. 511-525; W. A. Langmed pp. 475-494; H. F. Schulte, pp. 495-510.
3. W. S. Snyder. A method of interpreting excretion data which allows for statistical fluctuation of data IAEA-SM-150/35, pp. 485-493.
4. K. Stewart, D. M. K. Thomas, I. L. Terry, R. H. Wilson, A preliminary evaluation of the biological measurements on operation Roller Coaster. AWRE. Report No 0-29/65.
5. Саяпина Р.Я. и др. в кн. "Материалы II научно-практич. конф. по радиац. безопасности". М., изд. ВЦНИИОТ ВЦСПС, 1970, стр.28.
6. Колмогоров А.Н. ДАН СССР, 1941 г. т.31, № 2.
7. Нормы радиационной безопасности НРБ-69, М., Атомиздат, 1970 г.
8. Москвлев В.И. и др. Концепция биологического риска воздействия ионизирующего излучения. М., Атомиздат, 1973 г.
9. Горюдинский С.А. Средства индивидуальной защиты для работ с радиоактивными веществами. Атомиздат, 1973 г.

## A TABULATION OF SPECIFIC EFFECTIVE ENERGIES FOR DOSE CALCULATIONS\*

M. R. Ford and W. S. Snyder  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee

### Abstract

A tabulation is presented of the energy absorbed per gram in various target organs per disintegration of a radionuclide in a source organ. For each radionuclide and each source and target organ, the energy absorbed due to photons, beta-like radiation, alpha particles, and recoil nuclei is given, each weighted with the appropriate quality factor recommended by the ICRP and the NCRP. The total specific effective energy is listed also. The source-target organs include bone and active bone marrow and bone and endosteal cells as well as the organs usually considered in radiation protection programs. The absorbed fractions of photon energy have been recomputed in a more realistic phantom using the methods developed in MIRD Pamphlet 5. When the Monte Carlo calculation did not give a sufficiently accurate result, various approximate procedures were used, and these are described. The tabulation is convenient to use because it avoids separate consideration of each product of the decay scheme and does not imply any restriction on the metabolic model to be used. However, the sources are considered to be uniformly distributed in the source organ, and only a mean energy deposition in the target organ is estimated.

### Introduction

The energy expression used in computing dose and/or MPC values recommended at the present time by the NCRP and the ICRP is referred to as the effective energy. This effective energy is the energy absorbed per disintegration of the radionuclide weighted by factors related to linear energy transfer and to the biological effectiveness of the radiation. The specific effective energy (SEE) values presented here correspond, in principle, to these effective energy values, but they are updated with more recent and extensive decay data and are divided by the mass of the organ receiving the dose. Because of this division by mass, the energy expression is in units of MeV/gram of the organ and thus, to the extent that energy absorbed is proportional to mass, allows for individual variation of the organ mass. Also, the SEE values provide the capability for computing cross irradiation, i.e., irradiation from energy absorbed in a target organ T which originates in a source present in another or source organ S. The effective energies now in use cannot be used in this way, because they are tabulated only for absorption in the source organ.

---

\* Research sponsored by the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.



## Decay Data

Estimation of the energy absorbed by an organ implies knowledge of all the energy emitted, but the usual decay schemes found in compilations of decay data, for example, the compilation by Lederer *et al.*<sup>1</sup>, do not include radiations such as x-rays and Auger electrons resulting from rearrangements in the electron shells, and these contributions are quite tedious to compute. Fortunately, Dillman<sup>2</sup> has published detailed decay data of this kind together with average beta energies for many radionuclides of interest, and he has designed a computer code which produces these data. It is this code which is the source of the decay data used in this compilation.

## Absorbed Fractions

Apart from the difficulty involved in arriving at a complete listing of all radiations emitted per decay, considerable tedious calculations may be required to compute the total energy absorbed in body organs, particularly for photons. The authors have published in MIRD Pamphlet No. 5<sup>3</sup> absorbed fractions of photon energies for 12 monoenergetic photons ranging in energy from 0.01 to 4 MeV and distributed uniformly in 16 source organs of a mathematical anthropomorphic phantom whose organs approximate those of their prototypes in size, shape, composition, and density. A Monte-Carlo-type computer code was used in estimating the absorbed fraction for some 20 target organs. Thus absorbed fractions for various organs can be estimated for all photons emitted in the decay of a particular radionuclide by interpolating on energy. However, many photons may be emitted per disintegration and dose may be required for a variety of source organs to many target organs. For example, if a radionuclide emits 10 x-rays and 4 gammas--a not unusual case--and is present in 6 source organs, 84 simple multiplications and interpolations are required in the table of absorbed fractions (MIRD Pamphlet No. 5) to determine the total photon contribution for just one target organ. If there are a number of target organs, these calculations become quite lengthy. In this event, the tendency could be to neglect the low-energy, low-intensity emissions. While any such photon alone might make a negligible contribution to the overall dose, taken together all such emissions in a decay may be quite significant.

## Specific Effective Energy Tables

To assist health physicists and others involved in making estimates of dose from internal emitters, tabulations of energy absorbed in target organs for particular radionuclides are in preparation. In these tables the above-mentioned interpolations, multiplications, and additions have already been made for all the photons emitted per decay. Also, the energy contributions from all other types of emissions are weighted by the proper quality and/or modifying factors as recommended by the ICRP and the NCRP and are included. These tabulations of specific effective energy (SEE) will be published for a large number of radionuclides including many of interest to the medical profession.

The specific effective energy for a source organ S and target organ T is given by

$$SEE(T-S) = \sum_i \frac{\bar{E}_i f_i (AF)_i(T-S) Q_i^T M_i^T}{m_T} \quad (1)$$



where  $\bar{E}_i$  (MeV) is the average or unique energy of the particle or photon  $i$ ,  $f_i$  is the yield per disintegration, and  $(AF)_i(T-S)$  is the fraction of energy absorbed in target organ T per emission in S.  $Q_i^T$  and  $M_i^T$  are quality factors or modifying factors generally assumed in internal dose estimations, i.e.,  $Q_i$  is taken as 1 for betas, monoenergetic electrons, gammas,

and x-rays; 10 for alpha particles; and 20 for recoil nuclei associated with alpha emission.  $M_T$  is used for alpha emitters in bone, other than radium, and is taken equal to 5. For alpha particles emitted in the contents of the gastrointestinal tract,  $M_T$  is taken as 0.01. This latter factor is recommended by the ICRP and the NCRP because of the demonstrated inefficiency of alpha particles in irradiating the mitosing cells of the tract.<sup>4</sup>

A portion of a SEE table for  $^{212}\text{Bi}$  is shown in Table 1. Each table of SEE values has the source organs listed across the top and the target organs shown vertically. Source organs include the contents of the bladder and of the sections of the GI tract, kidneys, liver, lungs, muscle, ovaries, pancreas, cortical bone, trabecular bone, skin, spleen, testes, thyroid, the total body, and a uniform infinite cloud (external to the body). Target organs include bladder wall, the walls of the sections of the GI tract, skeleton, total marrow, red bone marrow, endosteal cells on cortical bone surfaces, endosteal cells on trabecular bone surfaces, total endosteal cells, thymus, and uterus in addition to all the listed source organs except those labelled as "contents" and "cloud."

ORNL-DWG 73-5728

Table 1. Specific Effective Energy, SEE, for  $^{212}\text{Bi}$  (MeV per disint)/gram.

TARGET ORGANS	TYPE OF DECAY	SOURCE ORGANS		
		TRABECULAR BONE	LUNGS	 (23 SOURCE ORGANS)
TRABECULAR BONE	G	$1.3 \times 10^{-6}$	$3.5 \times 10^{-7}$	
	B	$4.9 \times 10^{-4}$	0	
	A	$1.1 \times 10^{-1}$	0	
	R	$4.2 \times 10^{-3}$	0	
	T	$1.1 \times 10^{-1}$	$3.5 \times 10^{-7}$	
RED MARROW	G	$1.3 \times 10^{-6}$	$4.4 \times 10^{-7}$	
	B	$1.4 \times 10^{-4}$	0	
	A	0	0	
	R	0	0	
	T	$1.5 \times 10^{-4}$	$4.4 \times 10^{-7}$	
LUNGS  (31 TARGET ORGANS)	G	$3.1 \times 10^{-7}$	$5.6 \times 10^{-6}$	
	B	0	$5.1 \times 10^{-4}$	
	A	0	$2.2 \times 10^{-2}$	
	R	0	$8.4 \times 10^{-4}$	
	T	$3.1 \times 10^{-7}$	$2.3 \times 10^{-2}$	

For each source-target combination, the SEE values are tabulated for four types of radiations and for the total of all radiations. The four types of radiations represent contributions due to energy absorbed from (1) gammas and x-rays, (2) betas and monoenergetic electrons, (3) alphas, and (4) recoil nuclei associated with alpha emission. These radiations are designated in the tables by the letters G, B, A, and R, respectively, with T indicating total values.

A general discussion of the computational method used in forming these estimates follows. A detailed account is given in the text accompanying the tabulations.

### Photons

The absorbed fraction of photons was calculated principally by the Monte Carlo method described in MIRD Pamphlet No. 5,<sup>3</sup> but new computer runs were made using a revised phantom and twice as many (60,000) source photons for each source organ and photon energy as were used previously.

Revisions in the phantom include (1) changes of the GI tract and bladder to include walls and contents separately except for the small intestine which is still defined as a wall plus contents, since, being essentially a tube free to move except at the two ends, it has no fixed position; (2) modification of the skeleton to provide a more realistic estimate of dose to red bone marrow and to endosteal cells, including addition of the clavicles and scapulae, rounding of the tissues covering the skull, and designation of areas corresponding to red bone marrow and yellow bone marrow; and (3) division of the combined configuration of the legs into two separate regions to provide a more realistic location for the testes.

Although the increase in sample size did improve the statistics somewhat, the accuracy was not sufficient to warrant use of the Monte Carlo estimates in every case. The method of estimating absorbed fractions of photon energies by use of Berger's<sup>5</sup> buildup factor for a point source of monoenergetic photons in an infinite medium of soft tissue was explored. Integrating the corresponding dose formula over regions representing the source and target organs yielded statistically reliable estimates which were plausibly an overestimate since buildup is maximal in the infinite space. However, compared to reliable Monte Carlo estimates, they differed by no more than 30% in most cases and were rarely high by more than a factor of 2. The justification of this method is discussed more fully by Snyder *et al.*<sup>6</sup>

Thus absorbed fractions estimated by the Berger buildup factor were used in cases where the Monte Carlo estimates were not statistically reliable. However, Berger's tabulations only extend to 15 keV. At lower energies, values have been obtained by extrapolation based on the concept that as the photon energy approaches 0, the absorbed fraction approaches 1 or 0, depending on whether the source and target organs coincide or are distinct.

Certain other absorbed fractions were obtained by use of the reciprocity theorem. This occurs in a few cases where an organ is programmed as a target organ but not as a source organ. For example, the values for muscle with kidney as the source organ were used for kidney with muscle as the source organ. This approach is discussed in more detail by Snyder *et al.*<sup>7</sup>

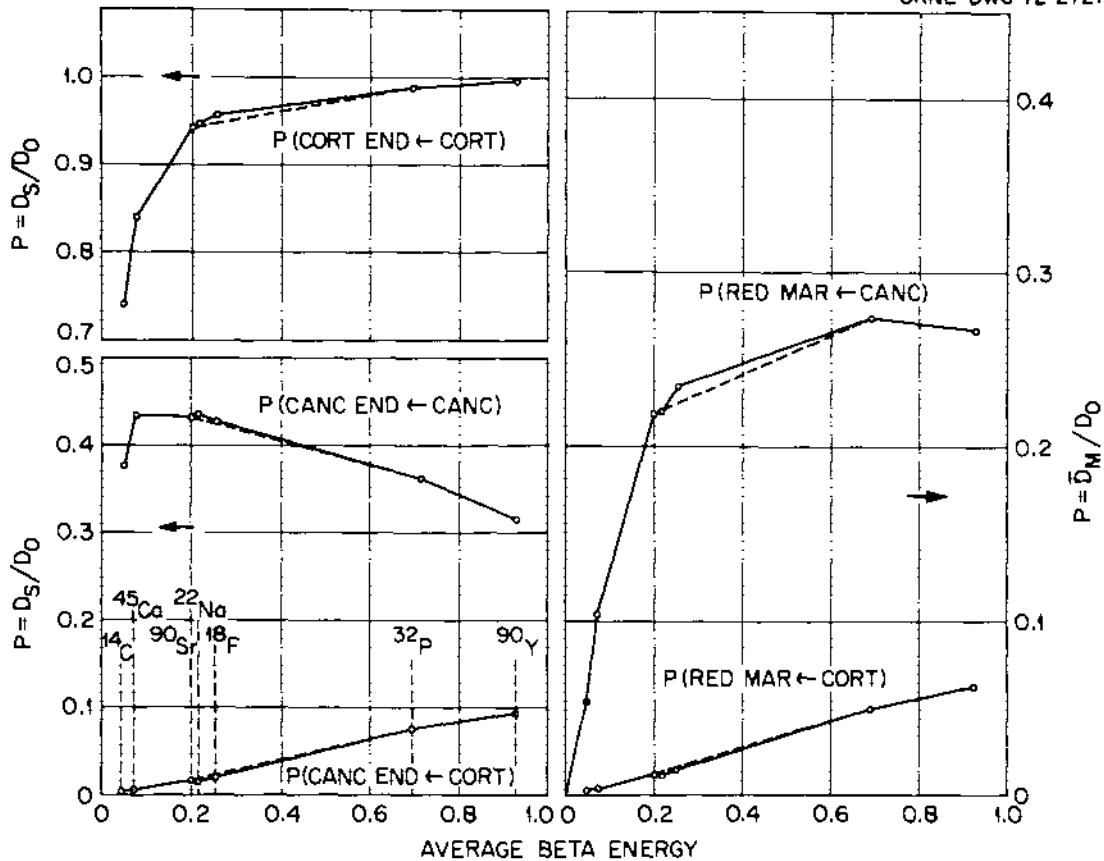
### Beta-Like Radiations

For beta-like radiations, the energy contributions of this short-range radiation are generally taken equal to 0 or to  $\bar{E}$  according as source and target organs are different or coincide. Among exceptions to this general rule are the organs with walls when the source

is in the contents. This occurs in the four sections of the GI tract as well as in the case of the bladder. In all such cases, the energy contribution is taken as  $\bar{E}/2$  where  $\bar{E}$  is the average energy of the beta or positron or equals the energy in the case of a monoenergetic electron. This amounts to using the dose on the surface of a half-space containing the source at a uniform concentration equal to that in the source tissue. Certainly this appears to be conservative in that the critical cells for the GI tract are frequently taken to be the mitosing cells which lie at some depth in the tissue.

The contributions of beta-like radiation to the active bone marrow and to the endosteal cells are based on calculations of Spiers.<sup>8</sup> The ICRP has recommended that endosteal cells be considered the radiosensitive cells for exposure to bone. The estimates of Spiers are shown in Fig. 1. They are expressed as the ratio of the desired doses,  $D_S$  (endosteal cells) and  $D_M$  (active bone marrow), to a dose  $D_0$  based on complete absorption of energy. These ratios,  $D_S/D_0$  and  $D_M/D_0$ , averaged over cortical and trabecular bone, have been given by Spiers for the following radionuclides:  $^{90}\text{Y}$  ( $\bar{E} = 0.927$ ),  $^{90}\text{Sr}$  ( $\bar{E} = 0.200$ ),  $^{14}\text{C}$  ( $\bar{E} = 0.049$ ),  $^{45}\text{Ca}$  ( $\bar{E} = 0.076$ ),  $^{22}\text{Na}$  ( $\bar{E} = 0.218$ ),  $^{18}\text{F}$  ( $\bar{E} = 0.253$ ), and  $^{32}\text{P}$  ( $\bar{E} = 0.695$ ). The values for  $^{18}\text{F}$  and  $^{22}\text{Na}$  were not considered to differ significantly from the values for  $^{90}\text{Sr}$ , and, hence, the dashed line was used for this portion of the graph. Estimates for other beta emitters are interpolated or extrapolated on  $\bar{E}$ .

ORNL-DWG 72-2721R



Ratios of Average Endosteal Dose and of Average Red Marrow Dose to the Equilibrium Dose in Bone from Beta Emitters. (Data from Spiers) Fig. 1.

## Alpha Particles and Recoil Nuclei

For alpha particles and recoil nuclei, the energy is assumed to be absorbed in the source tissue except for organs with walls where, again, the energy contribution is estimated for a surface dose, i.e.,  $E/2$ , and this is weighted by the appropriate quality factors and modifying factors mentioned earlier. However, recoil nuclei are assumed not to penetrate the mucus and irradiate the mitosing cells, and thus the dose from these nuclei is considered to be zero.

## Limitations

It should be noted that the use of the absorbed fractions, as calculated by the Monte Carlo method, implies that the corresponding doses are estimated for a uniform distribution of activity in the source organ and only an average dose to the target organ is obtained. Dose contributions from electrons, alpha particles, and recoil nuclei are also estimated only in the sense of an average dose. This is no inherent limitation of the method, as it would be feasible to estimate dose for other distributions of activity the biological data might indicate. However, these are the assumptions generally used by health physicists, and the paucity of data on the distribution of activity in the source organs is so prevalent that it does not appear worthwhile to provide data for other distributions which would largely be based on little more than guesses.

## References

1. C. M. Lederer et al., Table of Isotopes (John Wiley and Sons, Inc., New York, 1967), 6th edition.
2. L. T. Dillman, "Radionuclide Decay Schemes and Nuclear Parameters for Use in Radiation-Dose Estimation," J. Nucl. Med. Suppl. No. 2: 10 (March, 1969) and Suppl. No. 4: 11 (March, 1970).
3. W. S. Snyder et al., "Estimates of Absorbed Fractions for Monoenergetic Photon Sources Uniformly Distributed in Various Organs of a Heterogeneous Phantom," J. Nucl. Med. Suppl. No. 3: 10 (August, 1969).
4. M. F. Sullivan and R. C. Thompson, "Absence of Lethal Effects following Massive Oral Administration of Plutonium," Nature (London) 180, 651-652 (1957).
5. Martin J. Berger, "Energy Deposition in Water by Photons from Point Isotropic Sources," J. Nucl. Med. Suppl. No. 1: 15 (February, 1968).
6. W. S. Snyder, M. R. Ford, and G. G. Warner, "Estimates of Absorbed Fractions for Photon Emitters within the Body," Health Physics Problems of Internal Contamination, Second European Congress on Radiation Protection, 3-5 May 1972, Budapest, Hungary. Also Health Phys. Div. Annual Progress Report for Period Ending July 31, 1972, ORNL-4822, p. 86.
7. W. S. Snyder, "Estimation of Absorbed Fraction of Energy from Photon Sources in Body Organs," Medical Radionuclides: Radiation Dose and Effects (USAEC, DTI, Oak Ridge, 1970), pp. 33-49.
8. F. W. Spiers, Radioisotopes in the Human Body, Physical and Biological Aspects (Academic Press, New York, 1968).

ESTIMATES OF DOSE RATE TO GONADS OF INFANTS AND CHILDREN  
FROM A PHOTON EMITTER IN VARIOUS ORGANS OF THE BODY\*

W. S. Snyder  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee

Abstract

Estimates of genetic dose are needed for infants and children for realistic appraisal of environmental exposure of the population and also for medical use of radionuclide. Estimates of the dose rate due to a photon emitter from a source in ovaries or in testes are given in this paper for photons of 12 energies between 10 keV and 4 MeV in five phantoms corresponding to the newborn and children of ages 1, 5, 10, and 15 years. These estimates are obtained by use of the Monte Carlo technique applied to a phantom which is a transformation by similitude of a modification of the adult phantom reported in MIRD Pamphlet No. 5. Thus these estimates reflect the relative size, shapes, densities, and compositions of the various organs. By use of the reciprocity theorem, it should be possible to infer gonad dose from a radionuclide deposited in any source organ where the estimate is statistically reliable. It would be expected that this use of the reciprocity theorem would give estimates of gonad dose which are high but not by a large factor, perhaps by a factor of two or three at most.

---

This paper provides some estimates of dose rates to gonads from sources of a photon emitter distributed uniformly in various organs of the body. The results are obtained by application of the reciprocity theorem to the Monte Carlo estimates of dose rate from gonads to large organs. Although only results for photon energies of 0.02, 0.05, 0.1, 0.5, 1, and 2 MeV are reported here, it is planned to complete the series so the 12 monoenergetic sources used in MIRD Pamphlet No. 5<sup>1</sup> will be available for interpolation of other photon energies.

The basic phantoms used are essentially modifications of that reported in reference 1. The modifications include specification of regions where active bone marrow is deposited in the adult, addition of clavicles and scapulae to the skeleton, rounding of the top of the head, separate legs for the phantom, and relocation of the testes. These modifications will be presented in detail in a separate publication.<sup>2</sup>

For these calculations, the head, trunk, and leg sections of the adult phantom were transformed by similitudes to form the new phantoms. Although all organs and regions in each of these sections were shrunk by the same factors, these factors were not identical for

---

\* Research sponsored by the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.

the three sections which were given over-all dimensions typical for a given age. These factors are given in reference 3. The phantoms produced by use of these transformations are referred to below as being those of ages 0 (newborn), 1, 5, 10, 15, and 20 (adult) years. The corresponding masses for the total body are approximately 3.4, 10, 19.5, 31.8, 54, and 70 kg.

Since the dose rates given below are based entirely on use of the reciprocity theorem, it is appropriate to consider the validity of this theorem in phantoms corresponding to the various ages. The validity in the adult phantom has been studied extensively and is reported in reference 4. The theorem has been tested by programming sources of 60,000 photons at three widely separated regions of these phantoms and at several energies. The three regions used were the central volumes (approximately elliptical cylinders of semi-axes 16 cm and 8 cm and height 14 cm) of the top, middle, and bottom fifths of the trunk of the phantom. The regions contain rather different amounts of bone and lung tissue so that the inhomogeneity of the phantom is well represented. Three source energies were chosen--0.03, 0.1, and 0.5 MeV--and in all cases reciprocity held within 12%, generally being within a few percent in the corresponding cases. While far from the ideal solution of the problem, it is believed the values obtained for genetic dose by this means are sufficiently accurate for practical use since the actual distances of the gonads from the various source organs will vary with the individual and will only be approximately specified.

In Table 1 are displayed the estimates of dose rate from various source organs to the ovaries and testes of the newborn and the 1-year-old phantoms. As explained above, these values were obtained by Monte Carlo calculation and represent the dose rate to these organs from sources placed in the ovaries or testes, and thus the estimates given here are based on the use of reciprocity. It is expected that more complete results will be published when the results for the 12 monoenergetic sources of photons become available. When the indicated value is 0, the estimate is not considered to be reliable.

#### References

1. W. S. Snyder et al., "Estimates of Absorbed Fractions for Monoenergetic Photon Sources Uniformly Distributed in Various Organs of a Heterogeneous Phantom," J. Nucl. Med. Suppl. No. 3, 5 (1969).
2. W. S. Snyder and M. R. Fard, ORNL report (to be published).
3. M. J. Hilyer, G. S. Hill, and G. G. Warner, "Dose from Photon Emitters Distributed Uniformly in the Total Body as a Function of Age," These proceedings.
4. Walter S. Snyder, "Estimation of Absorbed Fraction of Energy from Photon Sources in Body Organs," Medical Radionuclides: Radiation Dose and Effects (USAEC/DTI, Oak Ridge, Tennessee, 1970), p. 33.

DOSES (RADS/PHOTON) AND COEFFICIENTS OF VARIATION (PER CENT)

SOURCE IN OVARIES OF NEWBORN PHANTOM

ENERGY (MEV)

	0.020		0.050		0.100		0.500		1.000		2.000	
	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.
ADRENALS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
BLADDER WALL	2.1E-13	8.1E 00	2.9E-13	6.2E 00	3.8E-13	7.7E 00	1.8E-12	1.2E 01	3.5E-12	1.4E 01	4.6E-12	1.8E 01
STOMACH WALL	7.9E-15	2.1E 01	6.1E-14	7.4E 00	7.5E-14	9.3E 00	3.3E-13	1.5E 01	5.7E-13	1.9E 01	9.5E-13	2.2E 01
SMALL INTESTINE	7.2E-13	1.0E 00	5.2E-13	1.2E 00	6.0E-13	1.4E 00	3.1E-12	2.1E 00	5.6E-12	2.4E 00	9.2E-12	2.8E 00
U.L.I. WALL	7.2E-13	2.2E 00	4.5E-13	2.5E 00	5.0E-13	3.3E 00	2.8E-12	4.7E 00	5.0E-12	5.6E 00	7.9E-12	6.7E 00
L.L.I. WALL	1.2E-12	2.0E 00	6.3E-13	2.4E 00	7.3E-13	3.1E 00	3.9E-12	4.6E 00	7.4E-12	5.3E 00	1.2E-11	6.4E 00
HEART	0.0	0.0	1.4E-14	8.7E 00	2.7E-14	8.6E 00	1.4E-13	1.2E 01	3.7E-13	1.2E 01	4.9E-13	1.5E 01
KIDNEYS	5.2E-15	2.1E 01	6.0E-14	6.1E 00	7.6E-14	7.3E 00	4.0E-13	1.1E 01	6.2E-13	1.3E 01	1.0E-12	1.5E 01
LIVER	4.2E-15	9.9E 00	4.4E-14	3.1E 00	6.2E-14	3.5E 00	2.8E-13	5.0E 00	5.4E-13	5.7E 00	8.5E-13	6.8E 00
LUNGS	0.0	0.0	1.2E-14	6.7E 00	2.0E-14	7.0E 00	1.2E-13	9.9E 00	2.5E-13	1.1E 01	4.8E-13	1.2E 01
RED MARROW	1.9E-13	1.1E 00	2.8E-13	1.1E 00	1.8E-13	1.5E 00	4.8E-13	2.6E 00	9.3E-13	3.0E 00	1.6E-12	3.5E 00
YELLOW MARROW	9.3E-14	1.1E 00	1.9E-13	1.0E 00	1.2E-13	1.4E 00	3.4E-13	2.4E 00	6.7E-13	2.8E 00	1.1E-12	3.3E 00
OVARIES	9.4E-11	9.8E-01	2.2E-11	1.8E 00	2.8E-11	2.3E 00	1.7E-10	3.1E 00	3.2E-10	3.6E 00	5.2E-10	4.2E 00
PANCREAS	0.0	0.0	3.8E-14	1.5E 01	7.0E-14	1.6E 01	3.4E-13	2.5E 01	8.3E-13	2.5E 01	0.0	0.0
SKELETON	5.4E-14	1.1E 00	1.1E-13	9.3E-01	7.8E-14	1.3E 00	2.1E-13	2.1E 00	4.1E-13	2.4E 00	6.9E-13	2.9E 00
TOTAL SKIN	2.4E-15	8.6E 00	2.1E-14	2.8E 00	2.8E-14	3.3E 00	1.5E-13	5.1E 00	2.9E-13	5.8E 00	5.1E-13	6.8E 00
SPLEEN	0.0	0.0	5.0E-14	8.4E 00	5.0E-14	1.1E 01	2.2E-13	1.7E 01	5.1E-13	1.8E 01	1.1E-12	2.0E 01
THYROID	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
UTERUS	1.5E-12	2.7E 00	1.0E-12	3.1E 00	1.1E-12	4.0E 00	5.9E-12	5.8E 00	1.1E-11	6.9E 00	1.8E-11	8.0E 00
TOTAL BODY	9.1E-14	7.0E-02	9.2E-14	3.6E-01	1.0E-13	4.6E-01	5.0E-13	5.5E-01	9.2E-13	6.3E-01	1.5E-12	7.6E-01



DOSES (RADS/PHOTON) AND COEFFICIENTS OF VARIATION (PER CENT)

SOURCE IN TESTES OF NEWBORN PHANTOM

ENERGY (MEV)

628

	0.020		0.050		0.100		0.500		1.000		2.000	
	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.
ADRENALS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
BLADDER WALL	1.9E-13	8.4E 00	2.3E-13	7.2E 00	3.4E-13	8.5E 00	1.7E-12	1.3E 01	2.9E-12	1.5E 01	5.0E-12	1.8E 01
STOMACH WALL	0.0	0.0	9.3E-15	2.0E 01	1.7E-14	2.1E 01	1.4E-13	2.5E 01	0.0	0.0	0.0	0.0
SMALL INTESTINE	1.3E-15	2.1E 01	3.3E-14	4.8E 00	4.2E-14	5.3E 00	2.2E-13	7.2E 00	5.0E-13	7.9E 00	6.4E-13	1.0E 01
U.L.I. WALL	0.0	0.0	3.2E-14	8.6E 00	4.1E-14	1.0E 01	2.3E-13	1.5E 01	4.9E-13	1.7E 01	7.8E-13	2.0E 01
L.L.I. WALL	2.1E-14	1.3E 01	9.9E-14	6.1E 00	1.3E-13	7.4E 00	6.1E-13	1.1E 01	1.0E-12	1.4E 01	1.9E-12	1.6E 01
HEART	0.0	0.0	3.0E-15	2.1E 01	5.4E-15	2.1E 01	4.0E-14	2.0E 01	1.0E-13	2.2E 01	2.4E-13	2.3E 01
KIDNEYS	0.0	0.0	5.5E-15	1.8E 01	1.0E-14	1.9E 01	6.7E-14	2.4E 01	1.7E-13	2.5E 01	2.8E-13	2.9E 01
LIVER	0.0	0.0	6.7E-15	8.4E 00	1.0E-14	8.4E 00	6.9E-14	9.5E 00	1.4E-13	1.1E 01	2.1E-13	1.3E 01
LUNGS	0.0	0.0	1.9E-15	1.7E 01	3.3E-15	1.7E 01	2.7E-14	1.9E 01	9.2E-14	1.7E 01	1.3E-13	2.0E 01
RED MARROW	4.5E-15	6.2E 00	4.4E-14	2.6E 00	3.5E-14	3.3E 00	1.1E-13	5.1E 00	1.7E-13	6.2E 00	3.1E-13	7.1E 00
YELLOW MARROW	3.8E-15	5.2E 00	4.7E-14	2.1E 00	3.6E-14	2.6E 00	1.2E-13	3.9E 00	2.3E-13	4.7E 00	4.2E-13	5.5E 00
PANCREAS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
SKELTON	6.7E-15	3.0E 00	5.6E-14	1.4E 00	4.2E-14	1.9E 00	1.3E-13	2.7E 00	2.5E-13	3.1E 00	4.2E-13	3.7E 00
TOTAL SKIN	6.1E-14	1.9E 00	3.8E-14	2.2E 00	4.7E-14	2.8E 00	2.7E-13	3.9E 00	5.2E-13	4.5E 00	8.0E-13	5.5E 00
SPLEEN	0.0	0.0	8.5E-15	2.3E 01	1.4E-14	2.1E 01	0.0	0.0	0.0	0.0	0.0	0.0
TESTES	5.8E-11	7.2E-01	1.4E-11	1.4E 00	1.7E-11	1.8E 00	1.1E-10	2.4E 00	2.0E-10	2.8E 00	3.3E-10	3.2E 00
THYRUS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
UTERUS	1.1E-14	2.8E 01	7.3E-14	1.1E 01	1.1E-13	1.2E 01	4.8E-13	2.0E 01	7.7E-13	2.5E 01	1.8E-12	2.6E 01
TOTAL BODY	6.7E-14	2.3E-01	5.4E-14	5.3E-01	6.2E-14	6.2E-01	3.3E-13	7.1E-01	6.3E-13	8.0E-01	1.0E-12	9.4E-01

DOSES (RADS/PHOTON) AND COEFFICIENTS OF VARIATION (PER CENT)

SOURCE IN OVARIES OF ONE YEAR OLD PHANTOM

E N E R G Y (E V)

	0.020		0.050		0.100		0.500		1.000		2.000	
	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.
	ADRENALS	0.0	0.0	1.2E-14	2.9E 01	1.9E-14	2.9E 01	0.0	0.0	0.0	0.0	0.0
BLADDER WALL	5.0E-14	9.6E 00	1.6E-13	5.0E 00	1.7E-13	6.5E 00	8.3E-13	1.0E 01	1.4E-12	1.3E 01	2.3E-12	1.5E 01
STOMACH WALL	0.0	0.0	2.5E-14	7.1E 00	3.2E-14	7.9E 00	1.6E-13	1.3E 01	3.2E-13	1.4E 01	6.7E-13	1.6E 01
SMALL INTESTINE	2.4E-13	1.0E 00	2.7E-13	1.0E 00	3.1E-13	1.2E 00	1.5E-12	1.7E 00	2.6E-12	2.0E 00	4.3E-12	2.4E 00
U.L.I. WALL	2.7E-13	2.1E 00	2.4E-13	2.1E 00	2.7E-13	2.6E 00	1.3E-12	4.0E 00	2.4E-12	4.6E 00	3.6E-12	5.8E 00
L.L.I. WALL	4.6E-13	1.9E 00	3.3E-13	2.0E 00	3.5E-13	2.6E 00	1.9E-12	3.9E 00	3.5E-12	4.4E 00	5.2E-12	5.5E 00
HEART	0.0	0.0	4.0E-15	1.0E 01	9.5E-15	8.6E 00	5.0E-14	1.1E 01	1.0E-13	1.3E 01	2.2E-13	1.3E 01
KIDNEYS	0.0	0.0	2.4E-14	5.5E 00	3.9E-14	5.6E 00	1.7E-13	8.9E 00	3.4E-13	1.0E 01	5.0E-13	1.3E 01
LIVER	3.3E-16	2.1E 01	1.8E-14	3.0E 00	2.7E-14	3.0E 00	1.2E-13	4.4E 00	2.2E-13	5.1E 00	4.2E-13	5.6E 00
LUNGS	0.0	0.0	3.3E-15	7.1E 00	6.6E-15	7.0E 00	4.3E-14	9.2E 00	1.0E-13	9.9E 00	1.8E-13	1.1E 01
RED MARROW	4.7E-14	1.3E 00	1.5E-13	9.1E-01	1.1E-13	1.2E 00	2.6E-13	2.1E 00	4.4E-13	2.6E 00	7.4E-13	3.0E 00
YELLOW MARROW	2.1E-14	1.3E 00	8.3E-14	8.8E-01	6.4E-14	1.2E 00	1.7E-13	2.0E 00	2.9E-13	2.4E 00	4.7E-13	2.9E 00
OVARIES	4.6E-11	7.9E-01	1.1E-11	1.5E 00	1.3E-11	1.9E 00	8.4E-11	2.6E 00	1.6E-10	3.0E 00	2.4E-10	3.6E 00
PANCREAS	0.0	0.0	2.0E-14	1.2E 01	3.6E-14	1.2E 01	1.0E-13	2.4E 01	3.7E-13	2.3E 01	0.0	0.0
SKELTON	1.3E-14	1.3E 00	5.1E-14	7.9E-01	4.1E-14	1.0E 00	1.1E-13	1.7E 00	1.8E-13	2.1E 00	3.1E-13	2.5E 00
TOTAL SKIN	3.9E-16	1.3E 01	7.9E-15	2.6E 00	1.2E-14	2.8E 00	7.2E-14	4.1E 00	1.4E-13	4.7E 00	2.5E-13	5.6E 00
SPLEEN	0.0	0.0	1.7E-14	8.3E 00	2.5E-14	9.2E 00	1.3E-13	1.3E 01	2.1E-13	1.7E 01	4.1E-13	1.9E 01
THYMUS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
UTERUS	5.0E-13	2.8E 00	5.0E-13	2.8E 00	5.5E-13	3.3E 00	2.7E-12	5.0E 00	4.9E-12	5.9E 00	7.8E-12	7.2E 00
TOTAL BODY	3.1E-14	5.4E-02	4.3E-14	2.8E-01	5.1E-14	3.9E-01	2.3E-13	4.5E-01	4.3E-13	5.1E-01	7.1E-13	6.2E-01

DOSSES (PADS/PHOTON) AND COEFFICIENTS OF VARIATION (PER CENT)

SCIENCE IN TESTES OF ONE YEAR OLD PHANTOM

E N E R G Y (MEV)

639

	0.020		0.050		0.100		0.500		1.000		2.000	
	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.	DOSE	C.V.
ADRENALS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
BLADDER WALL	4.2E-14	1.1E 01	1.3E-13	5.7E 00	1.4E-13	7.4E 00	6.2E-13	1.2E 01	1.4E-12	1.3E 01	2.5E-12	1.5E 01
STOMACH WALL	0.0	0.0	2.1E-15	2.2E 01	8.3E-15	1.7E 01	3.7E-14	2.8E 01	0.0	0.0	0.0	0.0
SMALL INTESTINE	0.0	0.0	1.3E-14	4.4E 00	2.0E-14	4.6E 00	9.0E-14	6.6E 00	1.9E-13	7.3E 00	3.0E-13	8.7E 00
O.L.I. WALL	0.0	0.0	1.2E-14	8.9E 00	2.1E-14	8.4E 00	8.8E-14	1.4E 01	1.9E-13	1.7E 01	3.5E-13	1.8E 01
L.L.I. WALL	3.7E-15	1.8E 01	4.9E-14	5.1E 00	6.0E-14	6.0E 00	2.7E-13	9.6E 00	5.1E-13	1.1E 01	8.2E-13	1.3E 01
HEART	0.0	0.0	0.0	0.0	1.1E-15	2.3E 01	1.3E-14	2.2E 01	3.7E-14	2.0E 01	7.3E-14	2.2E 01
KIDNEYS	0.0	0.0	1.2E-15	2.4E 01	3.9E-15	1.6E 01	3.0E-14	2.1E 01	7.8E-14	2.2E 01	1.7E-13	2.3E 01
LIVER	0.0	0.0	1.7E-15	9.9E 00	4.0E-15	7.9E 00	2.3E-14	1.0E 01	5.9E-14	9.6E 00	1.1E-13	1.1E 01
LUNGS	0.0	0.0	2.8E-16	2.5E 01	1.6E-15	1.5E 01	1.2E-14	1.8E 01	3.0E-14	1.8E 01	7.5E-14	1.8E 01
RED MARROW	4.7E-16	1.0E 01	1.9E-14	2.4E 00	1.8E-14	2.7E 00	4.4E-14	4.5E 00	1.1E-13	4.9E 00	1.6E-13	5.9E 00
YELLOW MARROW	2.7E-16	9.5E 00	1.6E-14	2.1E 00	1.7E-14	2.3E 00	4.9E-14	3.6E 00	1.0E-13	4.0E 00	1.7E-13	4.7E 00
PANCREAS	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
SKELETON	8.6E-16	4.8E 00	2.4E-14	1.3E 00	2.4E-14	1.5E 00	6.4E-14	2.3E 00	1.2E-13	2.6E 00	2.1E-13	3.0E 00
TOTAL SKIN	2.1E-14	1.9E 00	1.7E-14	1.9E 00	2.3E-14	2.2E 00	1.3E-13	3.2E 00	2.5E-13	3.7E 00	4.5E-13	4.2E 00
SPLEEN	0.0	0.0	1.2E-15	2.8E 01	2.6E-15	2.6E 01	0.0	0.0	0.0	0.0	0.0	0.0
TESTES	2.3E-11	5.7E-01	6.2E-12	1.1E 00	7.6E-12	1.4E 00	4.6E-11	1.9E 00	8.1E-11	2.2E 00	1.4E-10	2.6E 00
THYROID	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
UTERUS	0.0	0.0	3.6E-14	8.9E 00	4.9E-14	1.1E 01	2.4E-13	1.7E 01	4.0E-13	1.9E 01	6.3E-13	2.4E 01
TOTAL BODY	2.5E-14	1.9E-01	2.6E-14	4.5E-01	3.2E-14	5.3E-01	1.6E-13	5.9E-01	3.0E-13	6.6E-01	5.1E-13	7.7E-01

DOSE FROM PHOTON EMITTERS DISTRIBUTED UNIFORMLY  
IN THE TOTAL BODY AS A FUNCTION OF AGE\*

M. J. Hilyer, G. S. Hill, and G. G. Warner\*\*  
Health Physics Division, Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830

Abstract

The dose rate from photon emitters distributed uniformly in the total body has been estimated for phantoms corresponding to ages 0 (newborn), 1, 5, 10, 15, and 20 (adult) years. Estimation is by the Monte Carlo technique, and the computer code uses phantoms obtained by transformations from the anthropomorphic phantom essentially as described in MIRD Pamphlet No. 5. Interest here is centered on the distribution of dose in the various organs which include active bone marrow and gonads. The dose rates near the surface and near the central axis are found to differ by approximately a factor of 2 for most energies. The calculation is for 6 monoenergetic sources of photons ranging from 20 keV to 2 MeV. The absorbed fraction of energy is found to vary approximately as  $(\text{mass})^{1/3}$  power for photon energies above 100 keV.

---

In this paper we give specific absorbed fractions for photon emitters distributed uniformly in the total body, i.e., the distribution is directly proportional to the density of the parts of the body. The total body becomes the source S, and any organ, including the total body, is a target organ T. The dose rate in the target organ is proportional to the specific absorbed fraction (SAF) or  $\Phi$ , which is defined as that fraction of the energy emitted which is absorbed per gram of the target organ. Thus

$$\Phi(T - S) = \frac{\text{Energy absorbed in T}}{(\text{Energy released in S}) (\text{Mass of T in g})}$$

Estimates of the specific absorbed fractions were obtained by the Monte Carlo technique for a modified phantom basically similar to the anthropomorphic phantom described in MIRD Pamphlet No. 5.<sup>1</sup> These modifications are mentioned in the report ORNL-4903.<sup>2</sup> The calculation was for six monoenergetic sources of photons ranging from 20 keV to 2 MeV with a sample of 60,000 photons used for each monoenergetic source. The SAF and the dose rate were estimated for phantoms corresponding to ages 0 (newborn), 1, 5, 10, 15, and 20 (adult) years. The phantoms representing the various ages were obtained by transformation from the adult anthropomorphic phantom as described by Snyder and Cook.<sup>3</sup> The adult phantom was reduced by scale factors selected separately for the head, trunk, and leg sections of the

---

\* Research sponsored by the U.S. Atomic Energy Commission under contract with Union Carbide Corporation.

\*\* Mathematics Division

phantom (Fig. 1). All organs and tissues within these sections were reduced by the scale factors for that particular section. Thus the loci of the organs which are nonintersecting in one phantom are nonintersecting in the other phantom.

When the source is the total body, the same general relationship for the SAF of the target organs seems to hold as when the source organ and the target organ are the same.<sup>4</sup> That is,

- (1) The SAF decreases as age increases. The newborn has the highest SAF, apparently due to the effect of the inverse square law.
- (2) The ratios of the SAF at a specific age to SAF for the adult decrease as the age increases. For instance, the SAF ratio for red bone marrow of the newborn to the adult for 0.020 MeV differed by a factor of 20, while the same factor for the 5-year-old to the adult was 4.
- (3) The SAF decreases as energy increases.

For a photon source distributed uniformly over the total body, the SAF (and hence the dose rate) to a target organ near the surface of the phantom is found to differ by approximately a factor of 2 from the SAF to a target organ near the central axis of the phantom. The trunk skin was selected as the surface organ, and the five subregions located along the central axis of the five tiers in the trunk of the appropriate age phantom were selected to simulate a central axis organ and the results are shown in Fig. 2. The ratio of  $\Phi$  averaged over the central axis subregions to the surface organ (trunk skin) is approximately 2 : 1. This ratio of a central axis organ SAF to the SAF of other organs will vary with the position of the organ relative to the central axis. In Fig. 2 this correlation is shown for the ovaries and testes for the 10-year-old phantom. The ovaries, which are positioned near the central axis, show a nearly 1 : 1 ratio with the central axis subregions of the trunk; while the testes, a more nearly surface organ, show a 1 : 1.5 ratio. Moreover, for larger organs, such as the liver, which extend from the center out toward the periphery of the body, the dose to that portion of the organ which is nearest the central axis will be somewhat higher than that part which lies near the surface.

Only data with a coefficient of variation < 30% were used in the statistical evaluations. In some instances, such as the ovaries and testes, the coefficient of variation for the calculated SAF estimates was > 30%. In the case of the testes, for more reliable data, the SAF estimates for the genitalia were used in Fig. 2. For the ovaries, a technique described by Poston and Snyder,<sup>5</sup> in which the SAF estimates to the appropriate subregion or subregions of the phantom in which the organs lie were used; and it is believed these give a more reliable estimate of the SAF for these organs than do the Monte Carlo results.

The specific absorbed fraction (SAF) and coefficient of variation for some of the organs within the heterogeneous phantoms for ages 0 (newborn) and 5 years are shown in Tables 1 and 2 of this report. The data for the phantoms corresponding to other ages are given in ORNL-4903 but are omitted here because of limitations on space. In cases where the coefficients of variation were consistently > 30%, the corresponding data from the appropriate geometrical subregion or subregions in the phantoms in which the organ was located were substituted. The values are distinguished by an asterisk in the table.

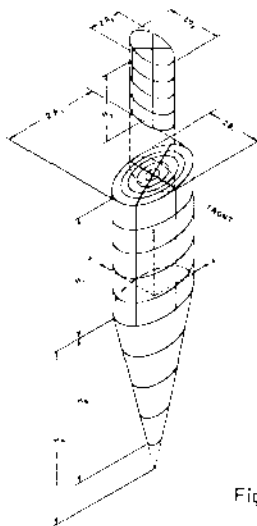
As previously reported,<sup>7</sup> when the source is distributed uniformly within an organ, the absorbed fraction (AF) for the organ might be expected to vary with the cube root of the mass, i.e.,  $(AF)_{x \rightarrow y}$  is proportional to  $\mu_{ab}^3/M$ . In Figs. 3 and 4 the ratios of the AF to  $\mu_{ab} M^{1/3}$  are plotted for various photon energies and organs. For energies of 0.100 MeV and higher, the ratio is found to be approximately a constant for all ages in the case of the liver

as well as for the total body. This behavior appears to hold also for other organs for which the Monte Carlo results are statistically significant, and thus the principle seems to be supported fairly well regardless of age. For lower energies, the rule does not appear to hold as might be expected from its derivation.

### References

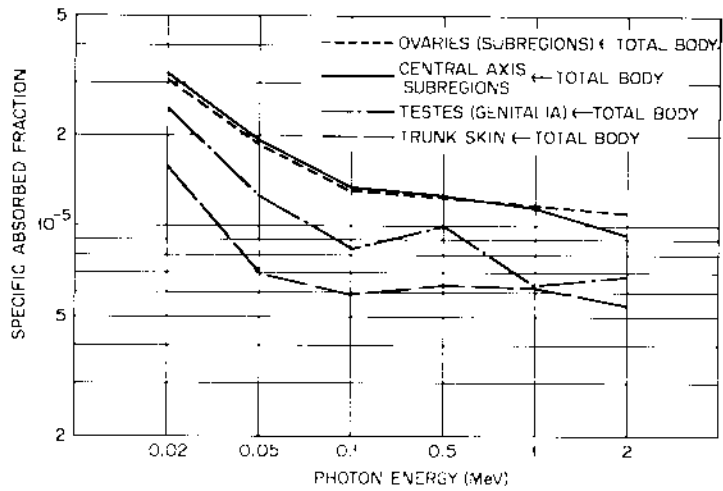
1. W. S. Snyder, Mary R. Ford, G. G. Warner, and H. L. Fisher, Jr., "Estimates of Absorbed Fractions for Monoenergetic Photon Sources Uniformly Distributed in Various Organs of a Heterogeneous Phantom," MIRD Pamphlet No. 5, J. Nucl. Med. Suppl. No. 3: 5 (1969).
2. W. S. Snyder and Mary R. Ford, "Estimates of Dose Rate to Gonads of Infants and Children from a Photon Emitter in Various Organs of the Body," Health Physics Division Annual Progress Report for Period Ending July 31, 1973, ORNL-4903.
3. W. S. Snyder and M. J. Cook, "Preliminary Indications of the Age Variation of the Specific Absorbed Fraction for Photons," Health Physics Division Ann. Prog. Report, ORNL-4720 (1971), p. 116.
4. M. J. C. Hilyer, W. S. Snyder, and G. G. Warner, "Estimates of Doses to Infants and Children from a Photon Emitter in the Lungs," Health Phys. Div. Ann. Prog. Report, ORNL-4811 (1972), p. 91.
5. J. W. Poston and W. S. Snyder, "A Model for Exposure to a Semi-Infinite Cloud of a Photon Emitter," In publication.
6. M. J. Hilyer, G. S. Hill, and G. G. Warner, "Dose from Photon Emitters Distributed Uniformly in the Total Body as a Function of Age," Health Phys. Div. Annual Progress Report for Period Ending July 31, 1973, ORNL-4903.
7. Walter S. Snyder, "Estimation of Absorbed Fraction of Energy from Photon Sources in Body Organs," Medical Radionuclides: Radiation Dose and Effects (U.S. Atomic Energy Commission Division of Technical Information, Oak Ridge, Tennessee, 1970), p. 33.

ORN: -DWG 73-5726



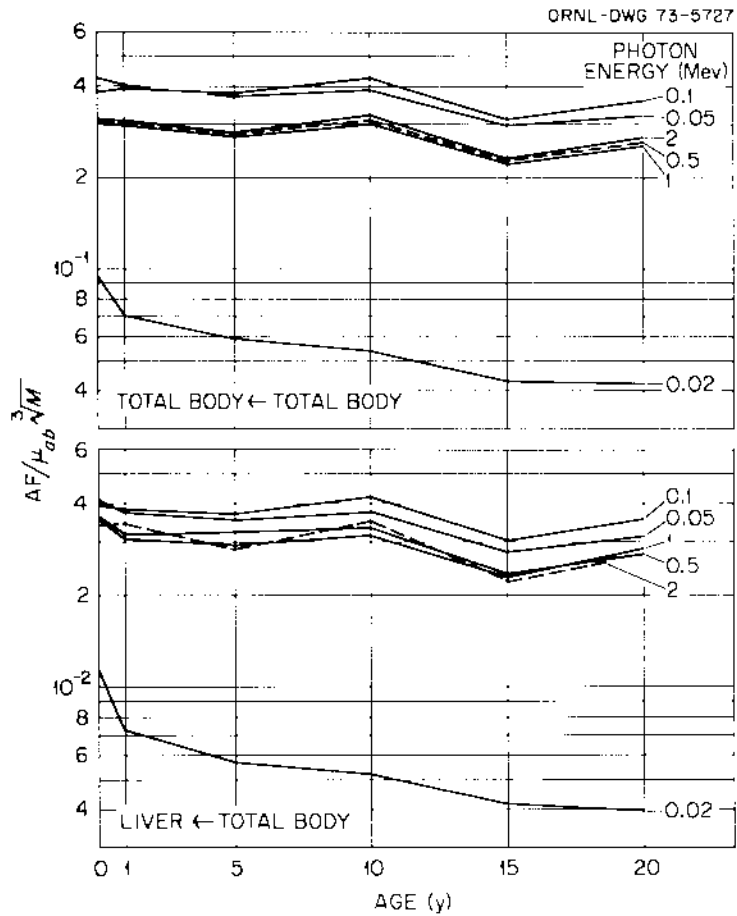
Age (yr)	Mass (kg)	H <sub>1</sub> (cm)	H <sub>2</sub> (cm)	H <sub>3</sub> (cm)	H <sub>4</sub> (cm)	A <sub>1</sub> (cm)	B <sub>1</sub> (cm)	B <sub>2</sub> (cm)	B <sub>3</sub> (cm)
0	3.475	23	10	13	20	5.5	5	4.5	5
1	10.171	33	16	22	36	8	7	6.5	7
5	19.654	45	20	46	57.5	11	7.5	6.5	7.5
10	31.902	54	22	64	80	14	8	6.5	8
15	34.041	65	23	76	97.5	18	9	7	9
20	70.037	70	24	80	100	20	10	7	10

Fig. 4. Masses and Dimensions for Phantoms Corresponding to Various Ages.



SPECIFIC ABSORBED FRACTION AS A FUNCTION OF PHOTON ENERGY AND ORGAN LOCATION FOR 10-yr OLD.

FIG. 2.



Figs. 3. and 4. Absorbed Fractions in Total Body and Liver as Related to Mass as a Function of Photon Energy and Age.

Table I

## SPECIFIC ABSORBED FRACTIONS AND COEFFICIENTS OF VARIATION (PER CENT)

	SOURCE IN TOTAL BODY OF NEWBORN PHANTOM											
	E N E R G Y (MEV)											
	0.020		0.050		0.100		0.500		1.000		2.000	
	S.A.F.	C.V.	S.A.F.	C.V.	S.A.F.	C.V.	S.A.F.	C.V.	S.A.F.	C.V.	S.A.F.	C.V.
*ADRENALS	3.1E-04	2.3E 01	7.5E-05	2.6E 01	8.1E-05	2.3E 01	*8.5E-05	6.8E 00	*8.9E-05	7.7E 00	*7.4E-05	9.0E 00
*BLADDER WALL	3.4E-04	1.2E 01	9.8E-05	1.2E 01	5.6E-05	1.6E 01	7.6E-05	2.2E 01	5.2E-05	2.9E 01	*8.8E-05	1.1E 01
STOMACH WALL	2.6E-04	7.7E 00	9.3E-05	7.3E 00	5.6E-05	9.0E 00	5.9E-05	1.4E 01	5.3E-05	1.6E 01	6.5E-05	1.6E 01
SMALL INTESTINE	3.0E-04	3.0E 00	1.0E-04	3.1E 00	6.5E-05	3.5E 00	6.9E-05	4.8E 00	6.5E-05	5.5E 00	5.1E-05	6.5E 00
U.L.I. WALL	3.0E-04	6.2E 00	1.1E-04	5.8E 00	6.5E-05	6.9E 00	6.5E-05	1.1E 01	5.6E-05	1.3E 01	7.1E-05	1.3E 01
L.L.I. WALL	2.7E-04	7.4E 00	1.1E-04	6.5E 00	5.9E-05	8.5E 00	6.8E-05	1.2E 01	6.3E-05	1.4E 01	4.4E-05	1.8E 01
HEART	2.7E-04	4.2E 00	1.2E-04	3.8E 00	7.4E-05	4.3E 00	8.5E-05	5.7E 00	8.5E-05	6.4E 00	6.5E-05	7.7E 00
KIDNEYS	2.1E-04	6.8E 00	8.2E-05	5.9E 00	5.6E-05	6.8E 00	6.3E-05	9.4E 00	5.6E-05	1.1E 01	5.2E-05	1.3E 01
LIVER	2.9E-04	2.4E 00	9.3E-05	2.6E 00	5.4E-05	3.0E 00	6.2E-05	3.9E 00	5.8E-05	4.5E 00	4.6E-05	5.3E 00
LUNGS	3.0E-04	2.9E 00	9.5E-05	3.0E 00	5.2E-05	3.7E 00	6.3E-05	5.0E 00	5.2E-05	6.3E 00	4.5E-05	7.3E 00
RED MARROW	3.1E-04	1.4E 00	1.5E-04	1.5E 00	5.4E-05	2.0E 00	3.3E-05	3.1E 00	3.1E-05	3.6E 00	2.6E-05	4.1E 00
YELLOW MARROW	4.3E-04	1.0E 00	1.9E-04	1.2E 00	6.4E-05	1.6E 00	4.3E-05	2.5E 00	4.0E-05	2.8E 00	3.4E-05	3.3E 00
*OVARIES	*2.3E-04	5.7E 00	1.6E-04	2.4E 01	1.1E-04	2.7E 01	*5.2E-05	9.2E 00	*4.5E-05	1.1E 01	*3.7E-05	1.3E 01
PANCREAS	3.2E-04	1.2E 01	1.1E-04	1.0E 01	5.1E-05	1.5E 01	7.4E-05	1.8E 01	5.5E-05	2.5E 01	6.7E-05	2.5E 01
SKELTON	3.2E-04	8.0E-01	1.5E-04	9.6E-01	5.1E-05	1.3E 00	3.5E-05	1.9E 00	3.2E-05	2.2E 00	2.6E-05	2.6E 00
TOTAL SKIN	1.1E-04	2.6E 00	3.6E-05	2.5E 00	2.4E-05	3.1E 00	2.9E-05	4.2E 00	3.0E-05	4.6E 00	2.3E-05	5.7E 00
SPLEEN	2.7E-04	7.5E 00	9.5E-05	7.5E 00	5.1E-05	9.5E 00	5.9E-05	1.3E 01	4.1E-05	1.7E 01	3.4E-05	2.0E 01
*TESTES	3.4E-04	1.8E 01	7.2E-05	2.0E 01	4.3E-05	2.9E 01	*6.3E-05	1.5E 01	1.3E-04	2.7E 01	*4.8E-05	2.0E 01
*THYMUS	2.6E-04	2.0E 01	7.8E-05	2.0E 01	5.1E-05	2.4E 01	*6.5E-05	8.6E 00	*5.8E-05	9.9E 00	*5.6E-05	1.1E 01
*THYROID	*1.3E-05	7.9E 00	*1.3E-05	5.4E 00	*7.2E-06	6.1E 00	*1.0E-05	7.2E 00	*8.8E-06	8.5E 00	*6.9E-06	1.0E 01
UTERUS	2.4E-04	1.3E 01	9.3E-05	1.1E 01	6.9E-05	1.3E 01	8.1E-05	1.8E 01	6.1E-05	2.1E 01	4.1E-05	2.8E 01
TOTAL BODY	2.2E-04	2.2E-01	8.0E-05	4.9E-01	4.2E-05	6.0E-01	4.4E-05	6.9E-01	4.1E-05	7.8E-01	3.4E-05	9.2E-01

\* S.A.F. to an appropriate subregion(s) is used instead of S.A.F. to the organ, because the coefficient of variation of the latter exceeded 30%.

Note: The digit following the symbol E indicates the power of ten by which each number is to be multiplied.



Table 2  
SPECIFIC ABSORBED FRACTIONS AND COEFFICIENTS OF VARIATION (PER CENT)

	SOURCE IN TOTAL BODY OF FIVE YEAR OLD PHANTOM											
	E N E R G Y (M E V)											
	0.020		0.050		0.100		0.500		1.000		2.000	
	S.A.F.	C.V.	S.A.F.	C.V.	S.A.F.	C.V.	S.A.F.	C.V.	S.A.F.	C.V.	S.A.F.	C.V.
* ADRENALS	4.7E-05	2.3E 01	2.0E-05	1.8E 01	1.5E-05	2.2E 01	2.2E-05	2.8E 01	*1.7E-05	7.0E 00	*1.6E-05	7.9E 00
BLADDER WALL	4.7E-05	1.3E 01	2.8E-05	9.4E 00	1.6E-05	1.1E 01	1.5E-05	1.9E 01	2.2E-05	1.9E 01	1.3E-05	2.6E 01
STOMACH WALL	4.8E-05	7.7E 00	2.8E-05	5.9E 00	1.6E-05	6.9E 00	1.7E-05	1.0E 01	1.4E-05	1.2E 01	1.3E-05	1.4E 01
SMALL INTESTINE	5.2E-05	3.1E 00	3.0E-05	2.6E 00	1.9E-05	2.8E 00	1.8E-05	3.8E 00	1.8E-05	4.3E 00	1.4E-05	5.3E 00
U.L.I. WALL	4.7E-05	6.6E 00	2.7E-05	4.7E 00	1.8E-05	5.5E 00	1.8E-05	8.5E 00	2.1E-05	8.9E 00	1.4E-05	1.2E 01
L.L.I. WALL	5.2E-05	7.4E 00	2.8E-05	5.5E 00	1.7E-05	6.2E 00	1.8E-05	9.5E 00	2.0E-05	1.1E 01	1.4E-05	1.3E 01
HEART	5.2E-05	4.2E 00	2.4E-05	3.7E 00	1.7E-05	3.9E 00	1.6E-05	5.6E 00	1.5E-05	6.3E 00	1.3E-05	7.3E 00
KIDNEYS	4.9E-05	6.0E 00	2.1E-05	5.1E 00	1.6E-05	5.5E 00	1.7E-05	7.6E 00	1.4E-05	9.1E 00	1.1E-05	1.1E 01
LIVER	5.0E-05	2.4E 00	2.5E-05	2.4E 00	1.5E-05	2.6E 00	1.7E-05	3.1E 00	1.5E-05	3.6E 00	1.2E-05	4.4E 00
LUNGS	5.7E-05	2.8E 00	2.6E-05	2.4E 00	1.7E-05	2.7E 00	1.7E-05	4.0E 00	1.5E-05	4.8E 00	1.2E-05	5.5E 00
RED MARROW	5.9E-05	1.4E 00	4.8E-05	1.3E 00	2.1E-05	1.6E 00	1.2E-05	2.3E 00	1.1E-05	2.7E 00	9.2E-06	3.2E 00
YELLOW MARROW	6.9E-05	1.1E 00	4.8E-05	1.0E 00	1.9E-05	1.3E 00	1.2E-05	1.9E 00	1.1E-05	2.2E 00	9.2E-06	2.6E 00
* OVARIES	6.4E-05	2.8E 01	2.4E-05	2.2E 01	2.2E-05	2.2E 01	*1.7E-05	6.7E 00	*1.5E-05	7.9E 00	*1.3E-05	9.2E 00
PANCREAS	4.3E-05	1.3E 01	3.0E-05	8.6E 00	2.0E-05	1.0E 01	2.1E-05	1.4E 01	1.4E-05	2.0E 01	9.3E-06	2.6E 01
SKELFTON	5.7E-05	8.4E-01	4.3E-05	7.9E-01	1.8E-05	1.0E 00	1.1E-05	1.5E 00	1.0E-05	1.7E 00	8.5E-06	2.0E 00
TOTAL SKTN	2.3E-05	2.5E 00	9.1E-06	2.0E 00	6.7E-06	2.3E 00	8.5E-06	3.2E 00	7.7E-06	3.7E 00	6.7E-06	4.3E 00
SPLEEN	4.6E-05	8.0E 00	2.4E-05	6.2E 00	1.5E-05	6.9E 00	1.2E-05	1.1E 01	1.2E-05	1.3E 01	1.3E-05	1.3E 01
* TESTES	4.4E-05	1.8E 01	1.9E-05	1.5E 01	1.5E-05	1.6E 01	1.1E-05	2.6E 01	1.5E-05	2.5E 01	*1.2E-05	1.4E 01
THYMUS	5.9E-05	1.7E 01	3.2E-05	1.3E 01	1.4E-05	1.5E 01	1.6E-05	2.7E 01	2.1E-05	2.5E 01	3.1E-05	2.5E 01
* THYROID	1.4E-05	2.8E 01	9.9E-06	1.9E 01	4.3E-06	2.6E 01	*3.2E-06	7.0E 00	*2.6E-06	8.2E 00	*2.2E-06	9.9E 00
UTERUS	6.3E-05	1.1E 01	3.2E-05	8.4E 00	2.0E-05	8.9E 00	1.6E-05	1.5E 01	1.8E-05	1.7E 01	1.3E-05	2.1E 01
TOTAL BODY	4.3E-05	1.7E-01	2.2E-05	3.7E-01	1.3E-05	4.7E-01	1.3E-05	5.2E-01	1.2E-05	5.8E-01	1.0E-05	6.9E-01

\* S.A.F. to an appropriate subregion(s) is used instead of S.A.F. to the organ, because the coefficient of variation of the latter exceeded 30%.

Note: The digit following the symbol E indicates the power of ten by which each number is to be multiplied.

DOSIMETRIC STUDY OF RESIDUAL BRAIN CONTAMINATION  
AFTER THE INJECTION OF A SOLUTION OF YTTERBIUM-169-DTPA  
IN THE CEREBROSPINAL FLUID

ZERBIB J.C. - Mme IMBERT L. - BONLEU M. - JOFFRE H.  
Centre d'Etudes Nucléaires de Saclay - France  
Service de Protection contre les Rayonnements

Dr DE TOVAR G. - Dr BEAUFILS F. - Mme COLLONGE M.  
Service des Radioisotopes - Professeur TRIAL R.  
Centre Médico-Chirurgical Foch - SURESNES - France

Abstract

Using radionuclides for medical purposes raises the question of the dosimetry of the radiation emitted by the radioactive products which are taken up in the system, whether preferentially or not.

The authors deal with a concrete case, namely ; the retention of ytterbium-169-DTPA after sub-occipital injection of the product. This kind of injection is made for an examination with a view to diagnosis.

A spectrometric analysis enabled the activity of the ytterbium taken up in the patient's brain mass to be known. A simulation carried out on a phantom was made using as guide :

- the space distribution of the radionuclides observed by scintiscanning
- the quantitative results of gamma spectrometry.

The measurements made on the patient as well as on the phantom enabled the observed dose due to the photons to be determined.

Along a horizontal plane passing through the eyes of the patient, the average absorbed dose is of several tens of rads in the central area of the brain.

In two other patients, the scintiscanning results showed a 20 to 30 % retention of the ytterbium-169 injected, after a rapid initial elimination phase.

The authors also made autoradiography of a frontal section of the brain of a patient on whom an isotopic study was made five weeks before death. This autoradiography showed that the radioactive product had spread throughout the brain and was taken up preferentially near the ventricular walls.

## Introduction

In the case of a sub-occipital injection of ytterbium-169-DTPA a retention greater than expected was observed by hasard on one patient (a retention also found later on other patients for lumbar injections).

The residual ytterbium-169 activity was determined a few weeks after sub-occipital injection of a know quantity of this radionuclide into the cerebrospinal fluid.

Knowing the activity, the absorbed dose is estimated for several points on the brain-pan.

### I - Measurement methods

To determine the activity fixed by the brain the internal contamination is simulated using a "RANDO" type phantom head.

This phantom (human skeleton covered with a "tissue - equivalent" material) is designed for external irradiation measurements.

#### 1.1. Simulation of internal contamination

Seven sealed cylindrical sources of ytterbium-169 roughly simulate the actual contamination.

Figure 1 shows the radiography of the phantom superimposed on the scintigraphy of the patient.

Figure 2 defines the location of the sources in an element of the phantom.

Figure 3 shows the radiography of the phantom superimposed on the scintigraphy obtained with the seven cylindrical sources in place.

Assuming that this simulation is a good enough approximation of the contamination, the activities measured by a spectrometric device, for two identical measurement geometries (patient and phantom), are in the ratio of the total absorption peak surfaces associated with the photon fluences emitted by ytterbium-169.

Figure 4 shows the four geometries for which measurements were made on the patient (once) and on the phantom (twice).

#### 1.2. Activity estimation

For each measurement geometry the total absorption peak surfaces taken into consideration relate to photons of 110 keV (18 %), 131 keV (11 %), 177 keV (22 %), 198 keV (35 %), 261 keV (1,7 %) and 308 keV (10 %). The emission percentages are those given by C.M. LEDERER <sup>1</sup>.

Two experiments have been made which gived the account following :

Experiment No 1 : mean activity =  $98 \pm 15 \mu\text{Ci}$

Experiment No 2 : mean activity =  $105 \pm 19 \mu\text{Ci}$

The two sets of measurements give the activity values calculated on the date of the measurement carried out on the patient (40 days after injection of 900  $\mu\text{Ci}$  ytterbium-169). These values are obtained from one set of measurements on the patient and two sets of measurements on the phantom. The reproducibility obtained is satisfactory.

The arithmetical mean activity value (all energies and all incidences = 24 values) for the two experiments, for a 95 % probability (student's criterion) is :

$$A_{av} = 101 \pm 12 \mu\text{Ci}$$

### 1.3. Elimination of ytterbium-DTPA by the organism

Several studies have been carried out with ytterbium-169-DTPA<sup>2,3,4,5,6</sup>. Recent measurements by KIRCHNER, KUSICH and WAGNER<sup>7</sup> showed a two-component exponential elimination of the product. The fast component involves 95 to 97 % of the injected product, which is eliminated with a half-life between 6 and 36 hours. The slow component covers the remaining 5 to 3 % which disappears with a half-life of 30 days, close to the half-life of ytterbium (32 days), and is thus due to radioactive decay alone.

In the case under investigation, if the activity value measured 40 days after the injection is extrapolated to time  $t = 0$ , we obtain :

$$A_0 = 254 \pm 30 \mu\text{Ci}$$

This means that slowly eliminated fraction represents about 30 % of the initial activity, which can lead to high absorbed doses.

### II - Measurement of absorbed dose

From the absorbed dose due to photons, measured by setting lithium fluoride dosimeters into the phantom in the presence of contamination-simulating sources, the absorbed dose to which the patient is exposed after injection of ytterbium-169-DTPA can be calculated.

The absorbed dose  $D$  delivered between times  $t_1$  and  $t_2$  is equal to :

$$D_{\text{rad}} = \int_{t_1}^{t_2} d(t)dt$$

The mean absorbed dose rate, per unit activity, at a point  $i$  on the phantom is :

$$d_i = \frac{D}{(t_2 - t_1) \cdot A_{\text{mean}}} \text{ rad. } \mu\text{Ci}^{-1} \cdot \text{d}^{-1}$$

$A_{av}$  = mean activity of sources in the phantom, between times  $t_2$  and  $t_1$  expressed in  $\mu\text{Ci}$

$t_2 - t_1$  = irradiation time in days.

The mean absorbed dose rate, per unit activity, delivered to the patients is the same as that measured in the phantom if for both geometries :

- 1) the contamination is identically distributed
- 2) the absorbing media are comparable in every way.

If these conditions are respected, the absorbed dose rate measured, at a point  $i$ , can be used to calculate the absorbed dose received at this point after a time  $t$  :

$$D_i = d_i \int_0^t A(t)dt = d_i \int_0^t A_0 \cdot \exp(-0.693 \frac{t}{T})dt$$

$$D_i = \frac{A_0 \cdot D \cdot T}{(t_2 - t_1) \cdot A_{\text{mean}} \cdot 0.693} \left| 1 - \exp(-0.693 \frac{t}{T}) \right|$$

$A_0$  is the extrapolated activity calculated above (254  $\mu\text{Ci}$ ).

Two distinct exposures were carried out and gave very similar results. For example, the conditions of the first experiment ( $A_{\text{av}} = 381 \mu\text{Ci}$ ,  $t_2 - t_1 = 3.71 \text{ d}$ ) lead to :

$$D_i = 8.29 D \left| 1 - \exp(-0.693 \frac{t}{T}) \right|$$

Figure 5 shows the isodoses for a horizontal plane passing through the patient's eyes. The values are calculated for zero exponential and hence represent the absorbed doses after complete elimination of the radionuclide.

The small circles represent the lithium fluoride detectors.

It must be stressed that the dosimetric measurements only refer to the photons emitted by ytterbium-169. The doses due to electron emission are much higher in the immediate vicinity (about a tenth of a millimeter) of the contaminated areas.

### III - Clinical studies

#### 3.1. Decays observed during gamma-camera measurements

Figure 6 shows the variation with time of the number of impulses measured on the skull by gamma-camera for three cases.

Case number one is that studied by dosimetry above. These results are expressed in percentages on figure 7 and the curves obtained lie close together, but do not comply with the decay scheme described by KIRCHNER, KUSICH and WAGNER<sup>7</sup>.

#### 3.2. Autoradiography of a brain section

This observation concerns a patient for whom the diagnosis of hydrocephalus at normal pressure was debated. The isotopic examination was carried out five weeks before death. A frontal brain section

passing through the third ventricle was autoradiographed two months after autopsy (figure 8) revealing the preferential fixation of ytterbium-169, free or bound, by the ependyma tissue, combined with an appreciable diffusion through out the brain.

#### IV - Conclusions

Gamma-camera measurements show that in the three cases investigated only 70 to 80 % of the activity injected is eliminated during the first few days, whereas according to other work<sup>7</sup> this decrease involves 93 to 97 % of the initial activity.

The fixed ytterbium-169 activity, estimated by simulating the contamination in a phantom, corroborates the results obtained by gamma-camera counting. This absolute measurement of the activity shows beyond doubt the high retention level of the radioactive product.

The mean absorbed dose due to photons is several tens of rads in the central region of the brain throughout the contamination period and for an injection of 900  $\mu$ Ci ytterbium-169-DTPA.

The ytterbium migrates than is fixed in the brain mass in an undetermined chemical form. Two hypotheses are possible :

- a) the complex is destroyed in vivo or in vitro and the radionuclide is fixed on the tissues,
- b) after destruction of the complex, new labelled molecules are formed in the tissues with the freed ytterbium.

If the complex is in fact destroyed as it migrates the fixation of ytterbium-169 is increased. Until these fixation phenomena are better understood it is preferable to reduce the amount of product injected.

It is also important to use a product containing very little non-complexed ytterbium (<1 %). In the event of long-term storage (3 to 4 weeks) it is wise to check the substance by chromatography before injection.

ooo

We wish to thank Madame PARMENTIER (CEA, Health Physics Department, Fontenay-aux-Roses, Nuclear Research Center) and Monsieur BOURDOISEAU (CEA, Radioelements Department, Saclay Nuclear Research Center) for their suggestions, criticisms and material help.

ooo

- 1 LEDERER C.M., HOLLANDER J.M., PERLMAN I.  
Table of isotopes - Sixth Edition (1967) - Edit. John Wiley and Sons, inc.
- 2 DELAND F.H., JAMES A.E., WAGNER H.N., HOSAIN F.  
Journal of Nuclear Medicine, vol 12, n° 10, pp. 683-688 (1971).
- 3 HOSAIN F., REBA R.C., WAGNER H.N.  
Radiology, vol 91, pp. 1199-1203 (december 1968).
- 4 HOSAIN F., REBA R.C., WAGNER H.N.  
Radiology, vol 93, pp. 1135-1138 (november 1969).
- 5 DELISLE M.J., BOUVY F. et VALEYRE J.  
AED-Conf- 118-009 (1971).
- 6 WAGNER H.N., HOSAIN F., DELAND F.H. and SOM P.  
Radiology, vol 95, pp. 121-125 (april 1970).
- 7 KIRCHNER P.T., KUSICK K.M. and WAGNER H.N.  
Journal of Nuclear Medicine, vol 13, n° 6, p. 442 (1972).

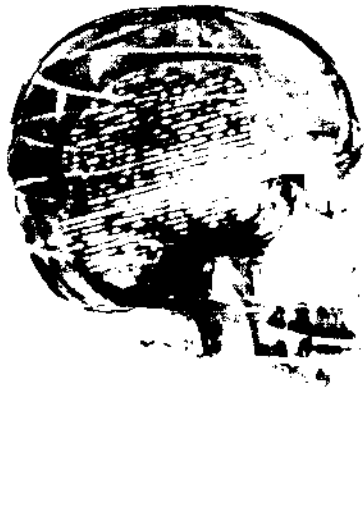


Fig. 1. Scintigraphy of the patient

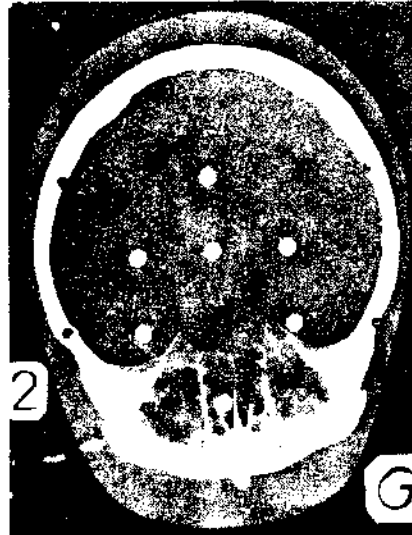


Fig. 2. Location of Ytterbium-109 sources

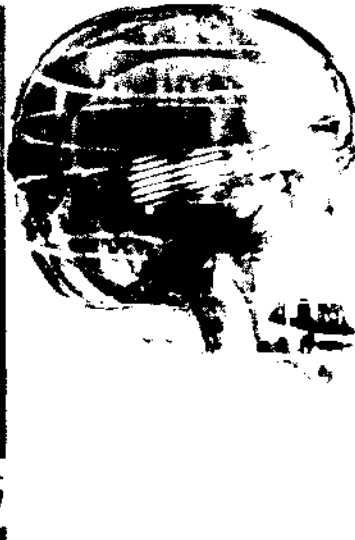


Fig. 3. Scintigraphy of the phantom

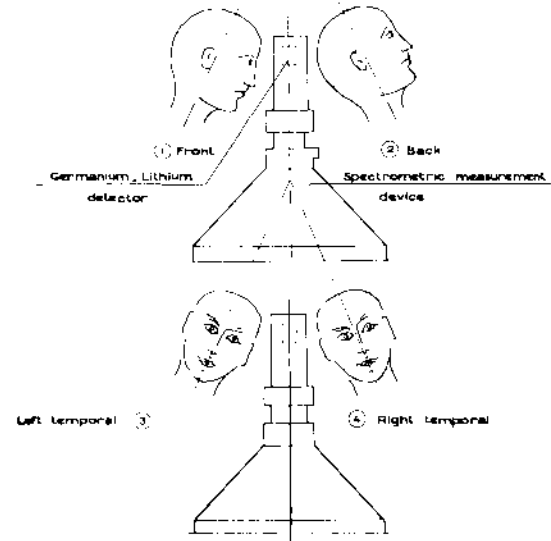
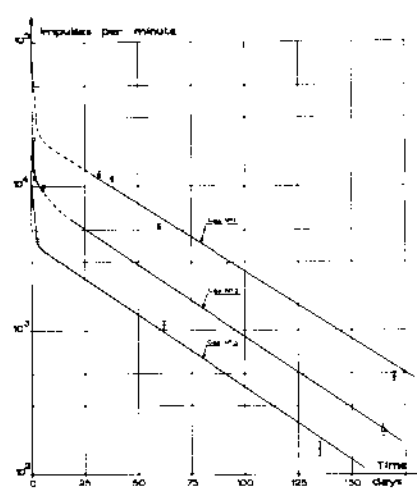


Fig. 4. Measurement geometries



Fig. 5. Sources for a horizontal plane passing through the eyes of the patient



NOTE: The activity injected is identical in all three cases: 900  $\mu$ Ci

Fig. 6. Variation with time in the number of impulses measured in the skull

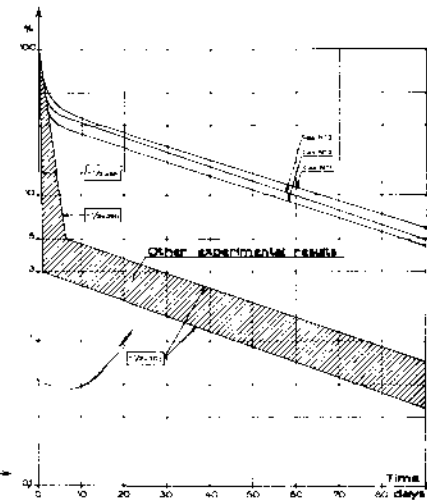
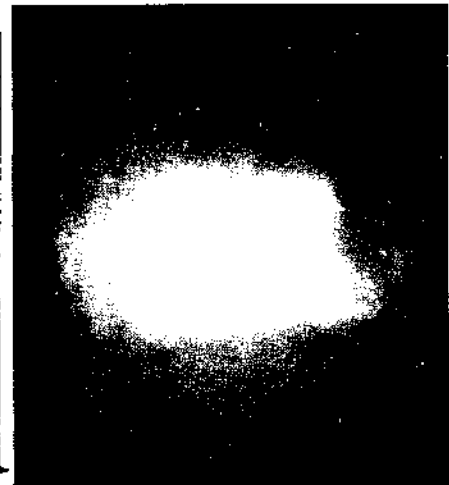


Fig. 7. Change with time in the count rate of impulses measured on the skull by gamma camera



Autoradiograph of a frontal section of the brain

Fig. 8

A TECHNIQUE FOR ESTIMATING THYROID DOSES  
FROM THE INTAKE OF IODINE-129  
FOR THE  
PROCEEDINGS OF THE THIRD INTERNATIONAL CONGRESS OF IRPA

Eric E. Oscarson  
Office of Radiation Programs  
U. S. Environmental Protection Agency  
Washington, D.C. 20460 U.S.A.

Abstract

Recent concern for the environmental buildup of long-lived radionuclides has suggested that dose estimation techniques should be developed for such nuclides. Iodine-129 is a long-lived radionuclide which has recently been found in low concentrations in milk in the environs around a nuclear fuel reprocessing facility. Thus, there was a need to develop a method for estimating the public health impact, in terms of thyroid dose, resulting from the consumption of this milk.

In developing the technique various approaches were considered including specific activity and dietary modeling. It was concluded the most acceptable approach was that using standard man information. The method developed here indicates that for the same level of intake thyroid dose would be only slightly higher from iodine-129 as compared to iodine-131.

Introduction

The projected proliferation of nuclear power reactors implies significantly greater production of radioactive waste products. Currently, the environmental buildup of reactor-produced long-lived radionuclides is causing concern over the health hazards to the world population. One isotope of iodine,  $^{129}\text{I}$  (with a 17 million year half-life), represents an essentially permanent contaminant when released to the biosphere. A normally operating reactor will release very little iodine; however, when the spent reactor fuel is dissolved at a chemical reprocessing plant a portion of the iodine contained in the fuel may be released to the environment.

The critical organ for radioiodine uptake by the general population is the thyroid<sup>1</sup> and the critical pathway considered is ingestion of cow's milk. This pathway is well defined for iodine-131, with its 8-day half life, and should act similarly for iodine-129 when the difference in half lives is considered.



Methods available for thyroid dose calculations include analysis of specific activity, of direct foliar contamination, and dietary and standard man modeling. The first two methods have been discussed by Russell<sup>2</sup> and Bryant<sup>3</sup>. Specific activity analysis may be a very good method for evaluating the impact from the long-term environmental build-up of iodine-129; however, standard man modeling is probably better suited for arriving at action levels. This analysis utilized standard man data and suggests guidance for monitoring operations.

#### Iodine-129 Releases from Reprocessing Plants

The reactor-produced radioactive fission products, including iodine-129, are contained, for the most part, by the fuel cladding. Dissolving the spent reactor fuel at a reprocessing plant removes this cladding barrier and reduces the fission products to a more easily dispersible form. Iodine is particularly susceptible to volatilization and must be removed from the offgas streams. Present technology, using caustic scrubbers in conjunction with silver zeolite, is expected to remove at least 99.8% of the iodine<sup>4</sup>. A typical reprocessing plant will handle 5 metric tons (MT) of fuel a day which contains about  $4 \times 10^{-2}$  Ci <sup>129</sup>I/MT<sup>5</sup>. Expected releases could therefore average  $4 \times 10^{-4}$  Ci <sup>129</sup>I/day.

The first commercial fuel reprocessing plant in the U.S. was operated by Nuclear Fuel Services, Inc. (NFS) at West Valley, New York from 1966 to 1972, when it was shut down for expansion. The State of New York, Department of Environmental Conservation has maintained an extensive environmental sampling program throughout the state. Their milk sampling program produced positive iodine-129 results, ranging from 0.4 to 2.1 pCi/l, in the vicinity of NFS during the first quarter of 1972<sup>6</sup>. These results were higher than previously measured and indicated a need for the more effective iodine control. NFS is currently installing additional cleanup systems, including silver zeolite, to reduce their iodine emissions.

#### Hazard Calculation for Iodine-129 Using Standard Man Data

When considering the public health hazards from iodine-129 the following characteristics are important:

1. The extremely long half-life of  $1.7 \times 10^7$  years, which makes it essentially a permanent environmental contaminant;
2. The physiological concentration of iodine by the human thyroid;
3. The food chain concentration of iodine in cow's milk; and
4. The low energy of the beta and gamma emitters, which make detection and measurement difficult.

The techniques used for calculating thyroid dose from iodine-129 are an adaptation of the methods developed by the International Commission on Radiological Protection (ICRP) in their Publication 2 on "Permissible Dose for Internal Radiation"<sup>1</sup>.

Although standard man data are used where applicable, it is modified by considering the 6-months-old infant as the critical individual<sup>3</sup>. In addition, the philosophy and methods promulgated by the former Federal Radiation Council (FRC), whose responsibilities were transferred to the U.S. Environmental Protection Agency (EPA), are utilized for these calculations.

The basic equation<sup>1</sup> for determining the radionuclide concentration in the medium of interest, M, in this case  $\mu\text{Ci } ^{129}\text{I}/\text{cm}^3$  milk, is:

$$M = \frac{q f_2 \lambda}{(1 - e^{-\lambda t}) S} \quad (1)$$

where: q = burden of iodine-129 in the whole body ( $\mu\text{Ci}$ )

$f_2$  = fraction of iodine-129 in thyroid to that in the whole body

$\lambda$  = effective decay constant =  $0.693/\tau_{\text{eff}}$

$\tau_{\text{eff}}$  = effective half-life (days)

t = period of exposure

S = product of the average rate of intake ( $\text{cm}^3/\text{day}$ ) of milk and the fraction of the iodine-129 arriving in the critical organ ( $f_w$ ).

The above equation may be solved more conveniently by combining it with the following equation<sup>1</sup> for the maximum permissible body burden, q ( $\mu\text{Ci}$ ), based on a maximum permissible dose rate of R rem/week:

$$q = \frac{2.8 \times 10^{-3} m R}{f_2 \epsilon} \quad (2)$$

where: m = mass of thyroid (grams)

R = dose rate (rem/week)

$f_2$  = defined above

$\epsilon$  = effective absorbed energy per disintegration of a radionuclide in the organ of reference (MeV).

The combined equation is as follows:

$$M = \frac{2.8 \times 10^{-3} m R \lambda}{\epsilon (1 - e^{-\lambda t}) S} \quad (3)$$

The assumptions used in solving this equation come from a number of references and are listed below:

1. Mass of the 6-months-old infant thyroid;  $m = 1.8 \text{ grams}^3$
2. Radiation Protection Guide (RPG) for the thyroid, averaged for the general population, of  $0.5 \text{ rem/year}^2$  yields;  $R = .0096 \text{ rem/week}$

3. A biological half-life for iodine in the infant thyroid of 23 days and, since the radiological half-life of iodine-129 is  $1.7 \times 10^7$  years, the effective half-life would be;  $t_{\text{eff}} = t_{\text{bio}} = 23 \text{ days}$ <sup>3</sup>
4. When the period of exposure,  $t$ , is long compared to  $t_{\text{eff}}$ , then  $(1 - e^{-\lambda t}) = 1$ . This is true after about 120 days
5. Milk intake of one liter per day =  $1000 \text{ cm}^3/\text{day}$ <sup>7</sup>
6. Fraction of iodine-129 ingested reaching the infants thyroid;  $f_w = 0.35$ <sup>3</sup>
7. Fraction of iodine-129 in the thyroid to that in the total body;  $f_2 = 0.2$ <sup>4</sup>
8. The effective energy of iodine-129 in the thyroid has been given as 0.068 MeV by ICRP<sup>1</sup>. For this paper the value has been recalculated using more recent data. Effective energy is assumed to be the sum of the average beta as energy, that portion of the gamma which is converted to directly ionizing particles, and the fraction of emitted x-rays which are absorbed. The average beta energy is 0.40 MeV<sup>8</sup>. The iodine-129 gamma ray (0.038 MeV) is very highly converted, i.e.,  $e_k/\gamma = 22$  and  $K/L \approx 10$ <sup>9</sup>. Therefore, only 4% of the gamma energy remains as unconverted photons. However, the fluorescent yield from the converted electron is large and much of the K x-ray energy escapes from the gland. Summing the net kinetic energy of the converted electrons, the energy given to auger and L electrons and that portion of the K x-rays that is absorbed in the gland yields 17 KeV per disintegration<sup>10,11</sup>. Therefore;  $\epsilon = 0.040 + 0.017 = 0.057$ .

Performing the calculation yields a value of  $0.072 \text{ pCi/cm}^3$  or  $72 \text{ pCi/l}$  for the concentration of iodine-129 in milk which, upon consumption of one liter per day, will deliver an annual thyroid dose of 0.5 rem to the 6-months-old infant.

#### Summary and Conclusions

Using the values calculated above, ranges similar to those defined by the FRC are proposed for iodine-129 concentration in milk. For this purpose 72 picocuries per day was rounded off to 70 picocuries per day. The corresponding ranges of transient rates of daily intake would be:

RANGE I      0 to    7 picocuries per day

RANGE II     7 to    70 picocuries per day

RANGE III    70 to  700 picocuries per day

By applying this reasoning to the New York State data one sees that  $2.1 \text{ pCi/l}$  falls within Range I. Under these conditions it would be appropriate to conduct routine surveillance to assure that levels would not reach Range II without knowledge of the authorities concerned<sup>7</sup>.

From this example it can be seen how this method of calculation might be used to provide guidance for monitoring operations. The problem of the long-term environmental buildup of this radio-nuclide appears to require considerable investigative efforts before any definitive guidance can be developed.

#### References

- (1) International Commission on Radiological Protection. Report of Committee II on Permissible Dose for Internal Radiation, Publication 2 (1959). Pergamon Press Ltd., London, England.
- (2) Russell, J. L. and P. B. Hahn, Public Health Aspects of I-129 from the Nuclear Power Industry. Radiological Health Data Reports 12:189-194 (April 1971).
- (3) Bryant, P. M. Derivation of Working Limits for Continuous Release Rates of  $^{129}\text{I}$  to the Atmosphere. Health Physics 19: 611-616 (1970).
- (4) Division of Radiological and Environmental Protection U.S. Atomic Energy Commission. Draft Detailed Statement of the Environmental Considerations Related to the Proposed Operation of the Midwest Fuel Recovery Plant Morris, Ill., by G.E. Co. Docket No. 50-268 (March 1972).
- (5) Oak Ridge National Laboratory. Siting of Fuel Reprocessing Plants and Waste Management Facilities, ORNL-4451. Oak Ridge National Laboratory, Oak Ridge, Tennessee (July 1970).
- (6) State of New York Department of Environmental Conservation. Environmental Radiation Bulletin 72-1. State of New York Department of Environmental Conservation, Albany, N.Y. (July 1972).
- (7) Federal Radiation Council. Background Materials for the Development of Radiation Protection Standards, Report No. 2 (1961). Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402.
- (8) Hogan, O. H., P. E. Zigman, and J. L. Mackin, Beta Spectra, USNRDL-TR-802. U.S. Naval Radiological Defense Laboratory, San Francisco, California (16 December 1964).
- (9) Lederer, C. M., J. M. Hollander, and I. Perlman, Table of Isotopes Sixth Edition, John Wiley & Sons, Inc., New York, N.Y. (1967).
- (10) Medical Internal Radiation Dose Committee. MIRD, Supplement 3. Society of Nuclear Medicine, Inc. (August 1969).
- (11) Medical Internal Radiation Dose Committee. MIRD, Supplement 5. Society of Nuclear Medicine, Inc. (March 1971).

STUDY OF SPATIAL DISTRIBUTION OF TISSUE DOSES  
WITH THE AID OF A PHANTOM-MANNEQUIN

L.N.Smirenniyi, E.G.Litvinova, A.V.Khortsev  
USSR

Abstract

In order to study in detail the radiation environment in a field of complex configuration, the procedure of computing doses in critical organs that takes into consideration real body shapes has been developed and a phantom-mannequin having the dimensions of a "standard man" has been manufactured from a tissue-equivalent material. The paper presents the results of computing doses in critical organs due to a solar flare with magnetic rigidity of 100 Mv and a flux of  $10^{10}$  protons/cm<sup>2</sup>. It also gives the doses and fluxes measured in the phantom-mannequin aboard the automatic interplanetary station "Zond-7" and the Earth artificial satellite "Kosmos-368".

Introduction

Problems related to the ensurance of radiation safety require detailed measurements of the doses affecting various organs of the human body. This is particularly important in cases of nonuniform shielding or attenuation of incident radiation and accumulation of the secondary radiation. The use of the doses estimated without taking into consideration the self-shielding of the body may give rise to a substantial error in the evaluation of radiation hazard. In the general form the solution of these problems is time- and labour-consuming. Therefore, one can measure depth doses experimentally, using phantoms, or compute dose distributions in simple geometric figures (infinite plate, sphere, cylinder, etc.). However, since the radiation field is appreciably nonuniform, these approximations are insufficient and may result in a significant error.

In order to measure depth doses, we made use of a complex approach, having developed a procedure of computing doses in the body of the "standard man" and having built a suitable phantom-mannequin.

Computation of doses in the body

Attempts of taking into consideration the real shape of the human body as precisely as possible were made elsewhere<sup>1,2,3</sup>.

We devised methods of computing depth doses, taking into account real geometry of the body. The thickness distribution in relation to a given point was estimated with the aid of a computer, using coordinates of points on the phantom-mannequin surface that had dimensions of a standard man. In order to determine different arrangement of individual segments, the body was subdivided into the following parts: (1) trunk (with the head), (2) thigh, (3) shin, (4) shoulder, (5) arm (Fig. 1).

The mutual displacement of individual body parts was achieved by turning and transferring the coordinate system of the corresponding part in relation to the coordinate system of the trunk. The computations yielded the relationship of tissue thicknesses  $h_t$  from angles of  $\vartheta$  and  $\varphi$ . Using the dose dependence on the thickness of the shielding tissue-equivalent substance  $K(h_t)$ , derived from the experimental findings (3), dose values in the critical organ can be obtained with the aid of the equation:

$$D = \int_0^{2\pi} \int_0^\pi \frac{dN(\vartheta, \varphi)}{d\Omega} K[h_t(\vartheta, \varphi)] \sin \vartheta d\vartheta d\varphi \quad (1)$$

where  $\frac{dN(\vartheta, \varphi)}{d\Omega}$  - proton flux, proton/cm<sup>2</sup> steradian;

$K[h_t(\vartheta, \varphi)]$  - dose as a function of the tissue thickness for a given proton spectrum, rad cm<sup>2</sup>/proton;

As an illustration we estimated doses in different body organs induced by protons of the solar flare of the following spectrum:  $N(p) = N_0 e^{-\frac{p}{P_0}}$  (2)

where:  $p$  - magnetic rigidity of protons, Mv, and  $P_0 = 100$  Mv. The computation results are given in Table I. The error of the dose computation for a given flux and spectrum depends on the error of assaying the relationship  $K(h_t)$  and accuracy of measuring  $h_t$ .

In our computations the tissue thickness was estimated to an accuracy of 2mm which yielded an error of the dose in critical organs of approximately 15%.

Table I

Dose values in different organs of the human body  
(sitting) during isotropic irradiation with solar flare protons

$$(P_0 = 100, N_0 = 10^{10} \frac{\text{PROTONS}}{\text{cm}^2})$$

<u>Organ of the body</u>	<u>Dose, rads</u>
Eye lens	3400
Gonads	400
Bone marrow (chest, at a depth of 3cm)	250
Bone marrow (vertebral column, at a depth of 3 cm)	270
Bone marrow (cranium, at a depth of 2.5-3 cm)	440

As it follows from Table I, dose values for various organs differ significantly.

Dose values in different organs may depend on the position of body parts as related to one another. Our computations show that the turn of thigh that accompanies the transition from the sitting to the standing position changes the solid angle with a minimum thickness of  $1.2 \text{ g/cm}^2$  by more than an order for gonads. Accordingly, the gonad-absorbed dose induced by solar flare protons increases 5 times (Fig.2). Thus, the method described allows computations of the dose in ray approximation for any organ of the human body upon different position of its parts as related to one another.

#### Tissue-equivalent phantom-mannequin

The experimental investigations of the dose distribution within the human body were carried out, using a phantom-mannequin manufactured from the tissue-equivalent material. The weight composition of the material is given in Table 2 which indicates also the weight composition of the human muscular tissue.

Table 2

Composition of the tissue-equivalent material (% by weight)

Element	Tissue-equivalent material	Average composition of the biological tissue
Hydrogen	7.0	10.4
Carbon	50.0	18.7
Nitrogen	3.0	3.1
Oxygen	40.0	67.8

Our estimations showed that values of the doses absorbed in the material used and in the biological tissue for protons and  $\gamma$ -radiation did not differ by more than 5-10% in the energy range of 1 to 1000 Mev and 0.1 to 3 Mev, respectively.

The phantom-mannequin was designed so as to resemble dimensionally the parameters of a "standard man" (5). It had movable joints (elbow, shoulder, pelvis, ankle) which allowed simulation of different postural positions of the human body.

The phantom-mannequin was equipped with 20 channels which were uniformly distributed along the whole body and allowed measurements of doses in critical organs by means of radiation detectors.

In our experiments we used as detectors thermoluminescent glasses and packages of nuclear emulsions of various sensitivity (6). The procedures employed in our experiments to estimate fluxes and doses were described elsewhere (6).

#### Experimental

Experimental studies of the dose distribution within the human body were carried out during space flights of the automatic interplanetary station "Zond-7" and the Earth artificial satellite "Kosmos-368". In both cases the phantom-mannequin was exposed to the galactic corpuscular radiation and  $\gamma$ -irradiation from isotope sources that were part of the experimental equipment. The use of nuclear emulsions and thermoluminescent glasses helped to estimate dose contributions of individual radiations. Fig. 1a, b shows the distribution of fluxes and doses measured at a depth of 3 g/cm<sup>2</sup> from the ventral surface along the central axis of the body. Measurements of the dose distribution as a function of the body depth indicated that the ventral-to-dorsal surface gradient was about 20%, thus giving evidence



for a significant rigidity of the radiation inside the modules. The head-to-pelvis gradient was the highest. The data shown in Fig. 1a, b demonstrate that onboard the "Zond-7" the  $\gamma$ -radiation dose increased greatly in the pelvic direction whereas fluxes of corpuscular radiations grew in the head area. "Kosmos-368" experiments also showed substantial variations of the fluxes and doses in the head-to-pelvis direction. The gradients of corpuscular fluxes can be attributed to the nonuniform shielding of the body by the space capsule walls and equipment. The nonuniformity of  $\gamma$ -radiation doses can be accounted for by local isotope sources and radiation attenuation by the phantom and equipment.

These findings clearly indicate that measurements with the aid of a phantom-mannequin are necessary to estimate radiation hazard in the field of a complex configuration.

#### References

1. D.L.Dye, Health Phys., 9, 749-756, 1963.
2. B.Liley a. S.C.Hamilton, Modified elemental volume dose program. (MEVDP), AFWL-TR-69-68, August 1969.
3. W.S.Snyder, H.L.Fisher et al., Journal of Nuclear Medicine, pp.7-12, 1969.
4. O.D.Brille et al., Nuclear Interactions in the Spacecraft Shielding, Atomizdat, Moscow, 1968.
5. O.A.Sidorov, Physiological Factors of the Man Determining the Arrangement of the Control Panel, Oborongiz, Moscow, 1962.
6. Y.A.Akatov et al. in: Physical and Radiobiological Investigations onboard Earth Artificial Satellites, Eds. Y.G.Grigoryev a. E.E.Kovalev, Moscow, 1971, p.25.
7. C.D.Zerby a. W.E.Kinney, Nucl.Instr.Meth., 36, 125-140, 1965.

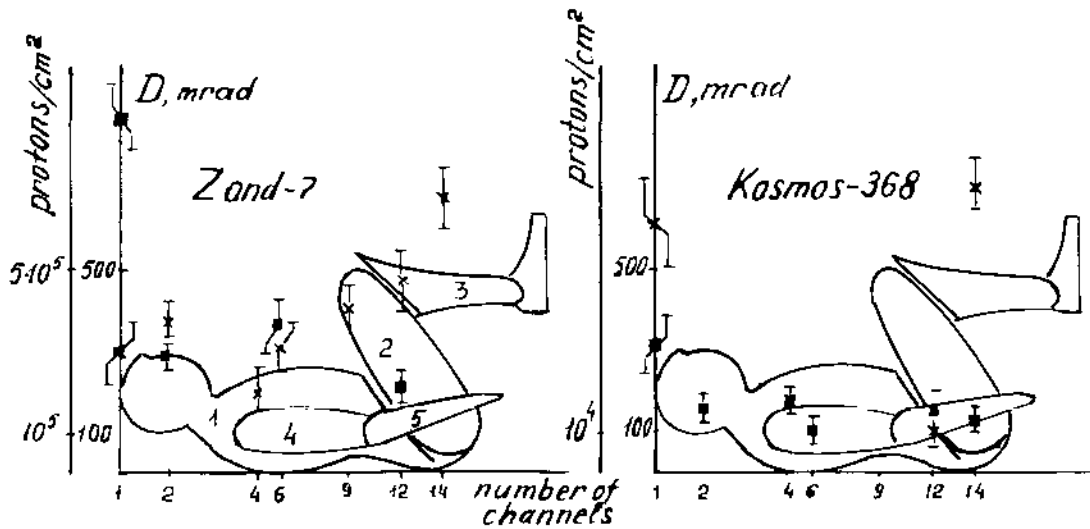


Fig.1. Results of measuring fluxes and doses in the phantom-mannequin during "Zond-7" and "Kosmos-368" flights. ■ - flux, proton/cm<sup>2</sup>, x - dose, mrad.

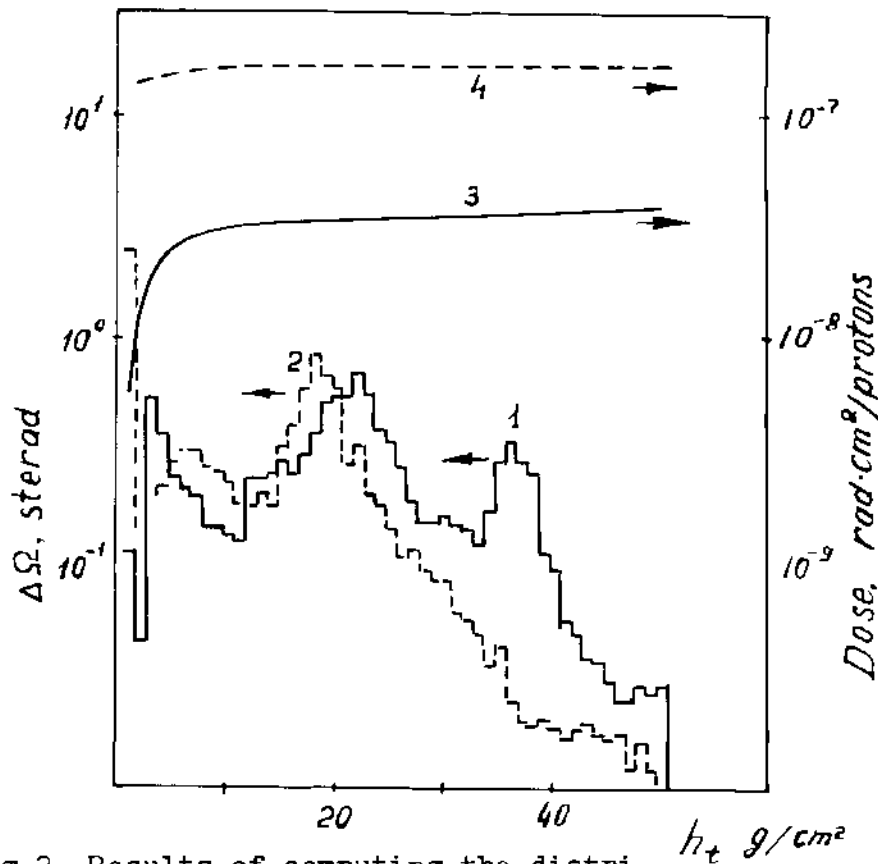


Fig.2. Results of computing the distribution of tissue thicknesses (1,2) as related to gonads and gonad-absorbed doses (3,4),  
 ——— - sitting, - - - - standing.

## THE NEED FOR AN OPERATIONAL RADIATION DOSE INDEX SYSTEM

Saul J. Harris  
Office of Radiation Control  
Department of Health  
New York, N.Y.

### Abstract

The health impact of radiation exposures do not necessarily relate to the dose received by an individual or human population as expressed in standard radiation physical units for dose, such as rad or rem. Nor do such units permit an easy indication of how a change in the design of medical radiographic equipment or in technique affects health.

There exists a need to have developed an operational radiation dose index system which can show, in a single unit, the combined effect upon individual or public health of the following parameters: exposure rate or radioactive material decay characteristics, the duration of the exposure, the effective volume of irradiated tissue, the biological sensitivity of each exposed organ or functioning part of the body, the genetic implications from the exposure (if any), the possibility of repetition of the exposure, the recuperative ability of the exposed volume, and the net "value" of the exposed volume to the individual. Arbitrary decisions for some of these factors would need to be made by expert committees on a scale of 1 to 100. The paper will present some preliminary calculations for several specific exposure conditions, such as a chest radiograph, a G.I. series, whole body occupational-limit exposure, and whole body population-limit exposure.

### Introduction

A need for an operational radiation dose index system is developing to assist regulatory programs at all levels of government in defining priorities for radiation control efforts. Particularly when working with elected officials, non-technical governmental review agencies (such as budget and personnel departments), and groups claiming to provide public representation, estimates of the severity of a radiation problem presented in purely physical units cannot provide a clear indication as to where program attention should be given, assuming a limited resource available in money and manpower. Expressing problems in terms of the genetically significant dose to the total population may be meaningless, both to the health physicist and certainly to the director of a budget agency, when justification for radiation hazard studies must be presented. Cost-benefit formulas have been proposed, through which the public health benefit in dollars is suggested for each rad or rem of dose reduced. Such formulas do indicate the type of programming problem which requires a broader consideration of factors other than the dose received by some organ, tissue or even whole-body, or the dollars saved in total public health from a reduction in such doses to a large segment of the population.

### Examples of Need

"The radiation hazard from the loose Iridium 192 was less than that from a dental x ray."

"The radiation exposure at the fence post would be 100 times less than that from a single chest x ray."

Comments such as these are replete in the press and the literature provided to the public regarding radiation problems. Such comments have been issued by qualified health physicists in efforts to suggest that certain radiation hazards are less than others, and if the general public accepts a radiation risk from certain medical exposures, the risk from other uses of radiation should also be acceptable because the physical radiation dose is less.

Cost-benefit estimates have also been suggested in the recent BEIR report to assist in logical decision-making processes giving due consideration to benefits, costs of reducing the risks, and the risks from alternatives. A national policy of reducing and maintaining radiation exposures to levels "as low as practicable" is also the policy of the City of New York and similar local jurisdictions and states. While such a policy encourages sound practice, estimates of risk and cost-benefit analysis are needed to reflect the public interest. The BEIR report stresses expressing both benefits and risks in a national unit, dollars, and cautions that such an approach does bring inherent problems, but "important advantages." One state has already calculated the health benefits to the citizens from a vigorous radiation control program assuming the program can reduce the state's average radiation exposure by 1 rad per person, or 0.5 rad per person.

UNSCEAR has been studying the same type of question for many years. In 1962 it attempted to summarize the then available data for the purposes of comparison of doses and estimates of risk. The term, "dose commitment," was offered for populations to represent the mean tissue dose to a population during a selected period or from a particular exposure in medical practice. The unit for dose commitment was the rem. In 1962, UNSCEAR concluded that there were insufficient data to make absolute risk estimates at this time.

The BEIR report information has subsequently been utilized by regulatory agencies in attempting to assess the possible dose commitment from radiation sources, such as the nuclear power industry, over the next fifty years, or by the year 2000. Absolute risk estimates for cancer deaths per rad or rem to populations of 1 million persons are set forth, as well as relative risk estimates for the whole body, gonadal, lungs, skin, and thyroid. The data are not repeated here, but it seems as though someone is responding to a real need today by developing and publishing risk estimates for radiation exposures in physical units as well as health effects.

### Proposed Factors in A Dose Index System

To serve the decision-making needs of operational radiation control programs, a dose indexing system should be able to consider the following factors:

- Physical: Dose rate (or effective half-life)
  - Duration of exposure
  - Volume of tissue exposed
  
- Biological: Somatic Effects, including
  - Fractionation
  - Radiation Sensitivity
  - Recuperation (age, health, etc.)
  - Genetic effects (if any)
  
- Risk: Absolute (or relative) risk
  
- Value: Economic
  - Social
  - Political

Physical factors include the rad and rem dose units to a given tissue, organ, or whole body. Biological effects would depend to some extent on dose rate, although for purposes of protection several references assume that the total dose effect can be considered as though it were dose rate independent; the biological sensitivity of the particular volume exposed to radiation; and the ability of the particular tissue, organ or whole body area to repair any damage caused by radiation. The genetic biological effect, if any, would be considered as essentially linear with total dose but dependent upon the age and certain health conditions of the population exposed.

The value judgements would relate to the extent to which the exposed volume is economically valuable to an individual or population in terms of whether employment would be affected, the extent to which an individual could be considered socially deprived from the possible damage from radiation to a particular tissue, and the extent to which it might be politically valuable to control the radiation exposure to portions of the body or to the whole body from particular sources of radiation.

Each of the factors, other than those physical factors which can be estimated with some degree of precision, would be given a range of arbitrary values scaled from 1 to 100. An expert group, presumably international, might provide factors for particular tissues for particular economic, social or political value, as well as for biological sensitivity, recuperation, etc. The value of the complete loss of some tissue, organ, or member of the body might be scaled in terms of the income benefits for disability for permanent bodily loss in weeks of income lost proposed by the Council of State Governments.

### Proposed Dose Index System

For the purpose of encouraging further consideration and study of an operational dose index system by an expert committee, this paper presents a brief list of some typical radiation exposures and the results of a crude calculation of a dose index for each exposure condition. The exposure conditions and dose index numbers are:

	<u>Dose (rad)</u>	<u>Dose Index(rad-M )</u> <sup>3</sup>
<u>Chest x ray</u> (14 x 17 film)	0.05	0.029
<u>G. I. Series</u>	9.0	10.0
<u>Whole Body</u> (occupational)	5.0(per year)	350.0
<u>Whole Body</u> (environmental)	0.005(per year)	3.5
<u>Lung Scan</u> ( <sup>131</sup> I MAA)	1.9	0.037
<u>Thyroid Uptake</u> (Na <sup>131</sup> I)	6.5	0.000036
<u>Thyroid</u> (uncollimated dental x ray)	0.4	0.000024

Ranges of values were assumed from 1 to 100 for biological sensitivity, genetic implications, economic and social value of the portion of the body exposed, and political value of controlling the exposure. Values for absolute risk of the number of cancer deaths based upon the BEIR report and the EPA projections were also included in the calculations. The recuperative value of each exposure was considered a constant for simplification of the calculations. The political value maximum of 100 was applied to the whole body, environmental dose since there has been considerable attention given to the standards for such exposure, while a political value of one was given to the G.I.series and the nuclear medical techniques since these seem to be "acceptable" exposures. The chest x ray, at least the mobile van type of photofluorographic system, was given a political value of ten, since its control has become the subject of more concern for governmental agencies and rates higher than one, but not as high as the maximum.

### Comment

From these very initial calculations, it does seem that a dose index system using arbitrary, consensus-type, "best value judgement" factors may present a formal approach to determining the importance of different radiation exposures. The G.I. series which could present the highest radiation exposure of those listed, and which might also represent the highest absolute risk potential is considerably less in importance as a control problem for health physicists because the risk is considered more acceptable, than the whole body, occupational exposure would be. The whole body, environmental, which is the lowest of any condition in terms of rads (only 5 millirads per year) rates considerably higher in the crude dose index data offered than any of the medical exposures other than the G.I. series; which does seem to be about the relative importance placed upon such exposures by health physicists.

### References

1. B. Lindell and R. L. Dobson, Ionizing Radiation and Health, World Health Organization, Geneva, Switzerland, 1961
2. The Evaluation of Risks from Radiation, ICRP Publication 8, Pergamon Press, London, England, 1966
3. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation, General Assembly Supplement No. 16 (A/5216), New York, 1962
4. The Effects on Populations of Exposure to Low Levels of Ionizing Radiation, Report of the Advisory Committee on the Biological Effects of Ionizing Radiations, National Academy of Sciences - National Research Council, Washington, D.C., November 1972
5. Assessment of the Possible Environmental Dose Commitment Resulting from Release of Long-lived Radionuclides Produced by Operation of the Nuclear Power Industry for the Next Fifty Years, U.S. Environmental Protection Agency, Washington, D.C. (draft of June 1973)
6. C. L. Comar, "Implications of the BEIR Report," remarks presented at the May 1973 meeting of the Conference of Radiation Control Program Directors, Portland, Oregon (to be published by BRH, FDA)

## CALCULS DE DOSE EN IRRADIATION EXTERNE PAR LES GAZ RARES

R. SCHAEFFER  
Electricité de France, Paris.

Abstract - Since the range of beta-rays is limited, it is necessary, for each point P at a depth "h" in the organism, to define the air volume within which the beta rays, which are able to reach the point P, are emitted. In this way the dose rate is calculated for monoenergetic radiations and its variation is plotted against "h" in a graphic form. This graph points out that the attenuation is relatively important for the low energies at the level of the epidermal basal layer and always very strong at the level of the lens; the gonads and the blood-forming organs are never reached. The formula used by the ICRP does not take into account this attenuation, but deals with beta-rays as with gamma-rays. The graph is applied to the beta-rays spectrum of the main inert gases released by nuclear plants, and the dose rates dissipated by the gamma-rays are added in order to get the values of the maximum permissible concentration (MPC) in air. The report concludes that the most MPC values used till now could be strongly increased.

### 1 - Introduction

Les gaz rares constituent la majeure partie des effluents gazeux des centrales à eau légère. En raison du développement de ce type de centrales, il nous a paru intéressant de préciser les débits de dose que les rejets de ces gaz peuvent délivrer aux travailleurs et aux personnes du public.

### 2 - Méthode de calcul

La plupart des gaz rares étant émetteurs de rayonnements bêta et de rayonnements gamma, il convient de distinguer ces deux catégories de rayonnements.

#### 2.1 - Débits de dose délivrés par les rayonnements bêta

Nous considérerons d'abord le cas d'électrons monoénergétiques, qui nous permettra de traiter ensuite le cas pratique des électrons émis par les gaz rares selon un spectre d'énergie.

2.1.1 - Cas des électrons monoénergétiques. Les rayonnements bêta ayant un parcours limité dans l'air et les tissus, il convient de définir, pour chaque point P situé à une profondeur h dans l'organisme, le volume d'air à l'intérieur duquel les rayonnements bêta, qui sont capables d'atteindre le point P, sont émis.

Si le parcours maximal des électrons dans l'air était égal à celui  $R_0$  dans les tissus, ce volume serait la calotte sphérique centrée au point  $P_0$  et de rayon  $R_0$  (fig. 1). Comme en réalité le parcours des électrons est n fois plus élevé dans l'air que dans les tissus, cette calotte sphérique se trouve dilatée selon ce rapport et devient le volume de révolution V de forme ovoïde représenté sur la figure 1.



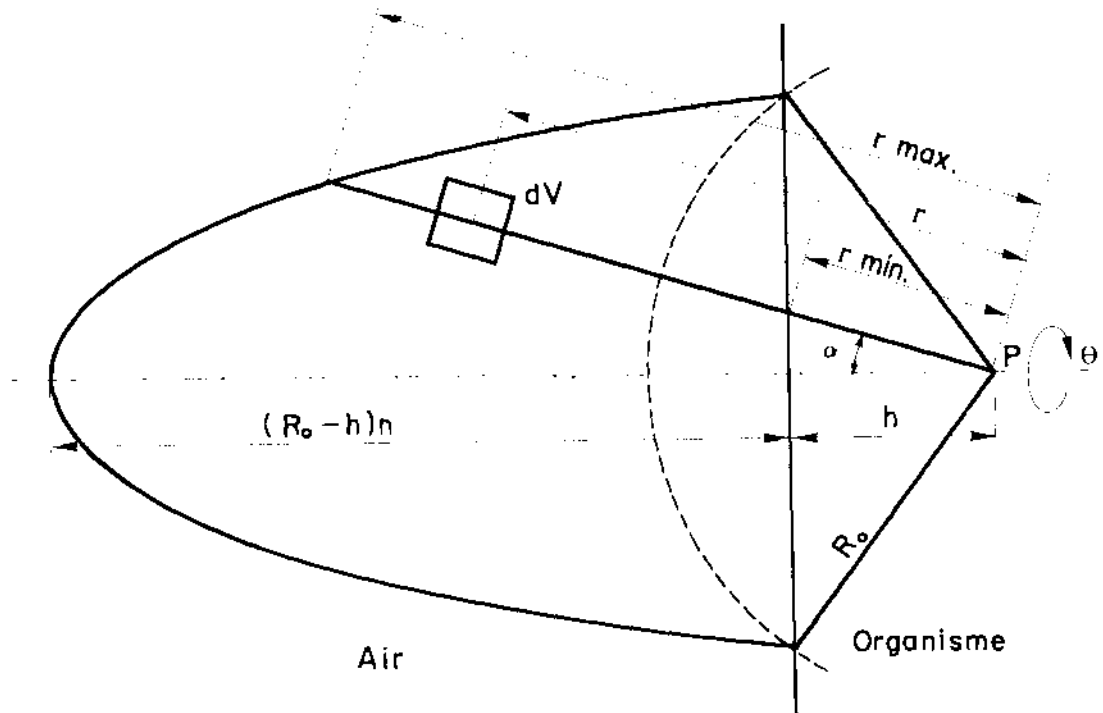


Fig.1 - Calcul du débit de fluence des électrons au point P.

Considérons un volume élémentaire  $dV$  à l'intérieur de ce volume. Le faisceau d'électrons qui en est issu et arrive au point  $P$  peut être considéré comme parallèle. Dans ces conditions, le débit de dose qu'il délivre au point  $P$  est donné, en millirads par heure, par l'expression :

$$d\dot{D} = 0,057 (S/\rho)_p d\psi, \quad (1)$$

où  $(S/\rho)_p$  représente la valeur du pouvoir d'arrêt massique des tissus au point  $P$ , exprimé en mégaelectrons volts centimètres carrés par gramme, et  $d\psi$  le débit de fluence d'électrons en ce même point, par centimètre carré et par seconde.

Le problème revient donc à calculer les valeurs du pouvoir d'arrêt massique et du débit de fluence au point  $P$  qui correspondent à chaque volume élémentaire  $dV$ , puis à intégrer l'équation différentielle (1) sur l'ensemble du volume  $V$ .

a) Valeur du pouvoir d'arrêt massique des tissus.

Nous avons adopté les valeurs indiquées par M. J. BERGER et S. M. SELTZER pour l'eau<sup>1</sup>, dont les caractéristiques en ce domaine sont voisines de celles des tissus mous. Dans ce cas,  $\rho = 1$  et les valeurs de  $S$  sont représentées, en fonction de l'énergie  $E$  de l'électron, sur la figure 2.

Nous constatons que la courbe de cette figure peut être assez bien traduite par l'expression analytique :

$$S = 1,9 + \frac{0,21}{E}, \quad (2)$$

qui constitue ainsi une forme très simplifiée, mais suffisamment approchée, de l'équation de BETHE qui a servi au calcul de  $S$ .

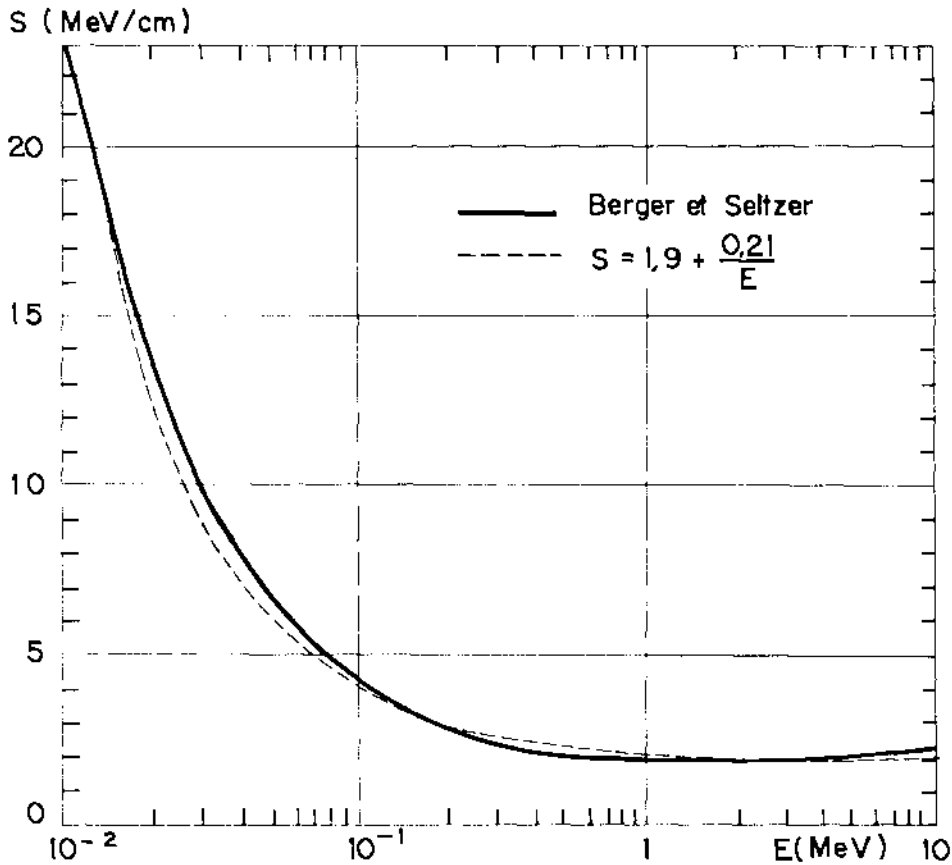


Fig. 2 - Variation du pouvoir d'arrêt de l'eau S pour les électrons en fonction de l'énergie E .

Puisque, par définition, le pouvoir d'arrêt exprime l'énergie dE perdue par unité de longueur de parcours dx :

$$S = - dE/dx ,$$

l'équation (2) peut encore s'écrire :

$$dx = - \frac{E}{1,9 E + 0,21} dE .$$

Son intégration de E à 0 donne la valeur du chemin restant à parcourir par l'électron jusqu'à son arrêt :

$$x = 0,526 E - 0,134 \log_{10} (1,9 E + 0,21) - 0,091 \quad (3)$$

Si l'on considère l'énergie initiale  $E_0$  de l'électron, x représente le parcours maximal  $R_0$  :

$$R_0 = 0,526 E_0 - 0,134 \log_{10} (1,9 E_0 + 0,21) - 0,091 \quad (4)$$

Cette expression présente sur les formules empiriques citées dans la littérature l'avantage d'être valable sur une gamme très étendue d'énergies, de 10 keV à 10 MeV.

Pour intégrer l'équation (1), nous avons besoin de connaître la variation de S le long de la trajectoire de l'électron. Si dans l'équation (3) on remplace E par sa valeur en fonction de S, tirée de (2), on obtient :

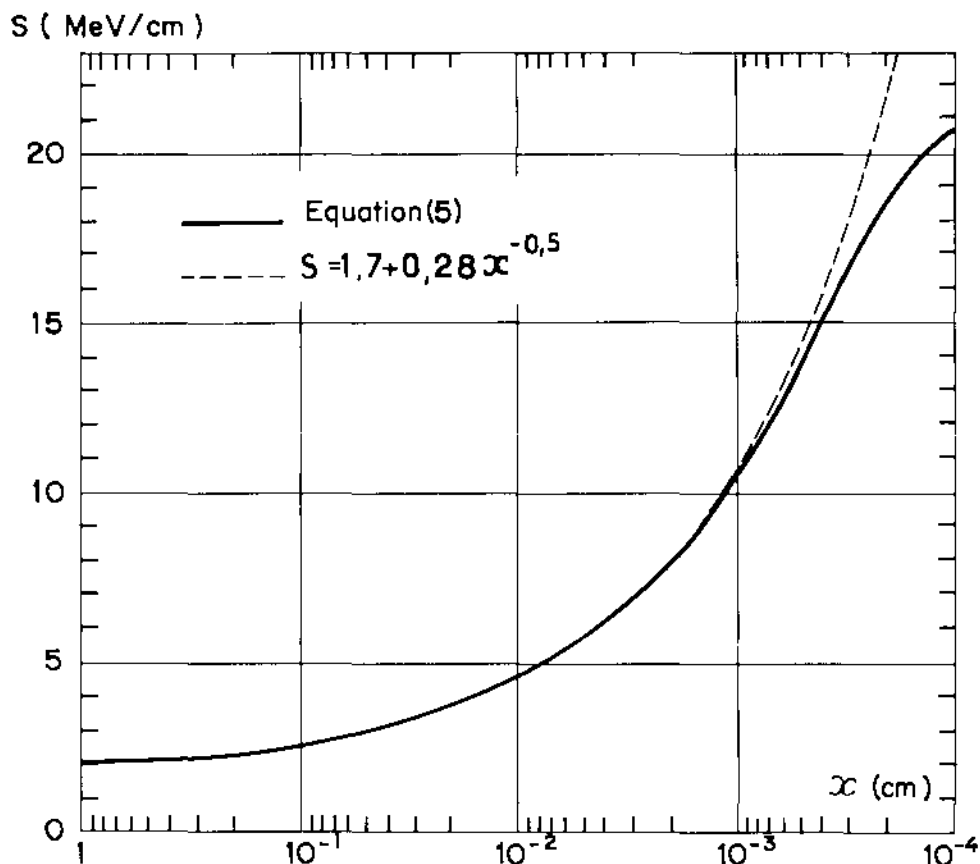


Fig. 3 - Variation du pouvoir d'arrêt de l'eau S pour les électrons en fonction de la longueur  $x$  de trajectoire qui leur reste à parcourir avant arrêt.

$$x = \frac{0,11}{S-1,9} - 0,134 \log_{10} \left( 0,21 + \frac{0,4}{S-1,9} \right) - 0,091 . \quad (5)$$

La courbe représentative de cette équation est représentée sur la figure 3. Nous constatons qu'elle peut être assez bien traduite par l'expression analytique beaucoup plus simple :

$$S = 1,7 + 0,28 x^{-0,5} \quad (6)$$

b)  Valeur du débit de fluence. Désignons par  $q$  ( $\mu\text{Ci}/\text{m}^3$ ) l'activité volumique de l'air, par  $\eta$  le rendement de la désintégration et par  $r$  (cm) la distance qui sépare le volume élémentaire  $dV$  du point P. Le débit de fluence des électrons issus de ce volume et arrivant au point P est :

$$d\varphi = 3,7 \times 10^{-2} q \eta \frac{dV}{4 \pi r^2} .$$

On lit sur la figure 1 :

$$dV = r^2 \sin \alpha \, dr \, d\alpha \, d\theta .$$

Il en résulte :

$$d\varphi = 2,93 \times 10^{-3} q \eta \sin \alpha \, dr \, d\alpha \, d\theta . \quad (7)$$

c) Intégration sur l'ensemble du volume V. Nous nous sommes inspirés de la méthode d'intégration utilisée par D. E. CHARLTON et D. V. CORMACK pour calculer les doses dissipées par les éléments ostéotropes au voisinage des interfaces des os et des tissus mous<sup>2,3</sup>.

La distance  $r$  parcourue par l'électron d'abord dans l'air puis dans les tissus jusqu'au point  $P$  est équivalente à une distance qui serait entièrement parcourue dans les tissus égale à :

$$\frac{r}{n} + \frac{h}{\cos \alpha} \left(1 - \frac{1}{n}\right)$$

La distance qui reste à parcourir au point  $P$  est donc :

$$x = R_0 - \frac{r}{n} - \frac{h}{\cos \alpha} \left(1 - \frac{1}{n}\right) \quad (8)$$

Les expressions (1), (6), (7), (8) donnent :

$$\dot{D} = 1,67 \times 10^{-4} q \eta \sin \alpha \, dr \, d\alpha \, d\theta \left\{ 1,7 + 0,28 \left[ R_0 - \frac{r}{n} - \frac{h}{\cos \alpha} \left(1 - \frac{1}{n}\right) \right]^{-0,5} \right\}$$

$$r \text{ varie de } r_{\min} = \frac{h}{\cos \alpha} \text{ à } r_{\max} = nR_0 + \frac{h}{\cos \alpha} (1 - n)$$

L'intégration conduit à deux intégrales triples :

$$\begin{aligned} \dot{D} = 1,67 \times 10^{-4} q \eta \left\{ 1,7 \int_0^{2\pi} \int_0^{\arccos \frac{h}{R_0}} \int_{r_{\min}}^{r_{\max}} \sin \alpha \, dr \, d\alpha \, d\theta \right. \\ \left. + 0,28 \int_0^{2\pi} \int_0^{\arccos \frac{h}{R_0}} \int_{r_{\min}}^{r_{\max}} \left[ R_0 - \frac{r}{n} - \frac{h}{\cos \alpha} \left(1 - \frac{1}{n}\right) \right]^{-0,5} \sin \alpha \, dr \, d\alpha \, d\theta \right\} \end{aligned}$$

La première de ces intégrales triples est directement résoluble. Sa solution est :

$$10,7 \, n \left( R_0 - h + h \log_e \frac{h}{R_0} \right)$$

La deuxième intégrale triple se réduit à une intégrale simple :

$$3,5 \, n \, R_0^{0,5} \int_0^{\arccos \frac{h}{R_0}} \left( 1 - \frac{h}{R_0 \cos \alpha} \right)^{0,5} \sin \alpha \, d\alpha$$

Si l'on pose  $\cos \alpha = u$ , cette intégrale s'écrit :

$$F(u) = \int_{h/R_0}^1 \left( 1 - \frac{h}{R_0 u} \right)^{0,5} du \quad (9)$$

Nous traçons, en fonction de  $u$  compris entre  $h/R_0$  et 1, les courbes :

$$f(u) = \left( 1 - \frac{h}{R_0 u} \right)^{0,5}$$

pour différentes valeurs du rapport  $h/R_0$  et nous mesurons les surfaces qu'elles limitent.

Les résultats obtenus sont représentés par la courbe de la figure 4, qui peut être assez bien traduite par l'expression analytique simple :

$$\left( 1 - \frac{h}{R_0} \right)^2$$

L'équation du débit de dose est donc :

$$\dot{D} = 10^{-4} q \eta n \left[ 17,8 \left( R_0 - h + h \log_e \frac{h}{R_0} \right) + 5,85 \, R_0^{0,5} \left( 1 - \frac{h}{R_0} \right)^2 \right]$$

Rappelons que  $\dot{D}$  est exprimé en millirads par heure,  $h$  et  $R_0$  en centimètres,  $q$  en microcuries par mètre cube.

$R_0$  est calculé à l'aide de l'équation (4).

$n$ , rapport du parcours maximal de l'électron dans l'air à celui dans les tissus, est égal au pouvoir d'arrêt des tissus relatif à l'air, lequel présente une valeur sensiblement constante pour les électrons relativement peu énergétiques émis par les gaz rares<sup>1</sup>:

$$n \approx 890$$

### 2.1.2 - Cas des électrons émis selon un spectre d'énergie.

La figure (5) représente les spectres des six principaux gaz rares rejetés par les centrales à eau légère. Nous les avons construits selon le procédé établi par J.H. MARSHALL<sup>4</sup> à partir de la théorie de FERMI<sup>5</sup>.

Les spectres partiels 1,2 MeV de  $^{41}\text{A}$  et 1,25 MeV du  $^{87}\text{Kr}$ , ainsi que tous les spectres du  $^{85\text{m}}\text{Kr}$ , du  $^{133}\text{Xe}$  et du  $^{135}\text{Xe}$  appartiennent à la classe des spectres dits de "transition permise". Les autres spectres sont de la classe des spectres dits de "transition interdite au premier ordre".

Les schémas de désintégration adoptés sont ceux qui ont fait l'objet des publications les plus récentes<sup>6</sup>.

Si  $N(E)$  représente les ordonnées de ces spectres,  $E_{\text{max}}$  l'énergie maximale et  $D(E)$  le débit de dose délivré par les électrons monocinétiques d'énergie  $E$  à une profondeur donnée dans l'organisme, le débit de dose moyen produit à ce niveau par le gaz considéré est :

$$\dot{D} = \int_0^{E_{\text{max}}} N(E) \dot{D}(E) dE / \int_0^{E_{\text{max}}} N(E) dE$$

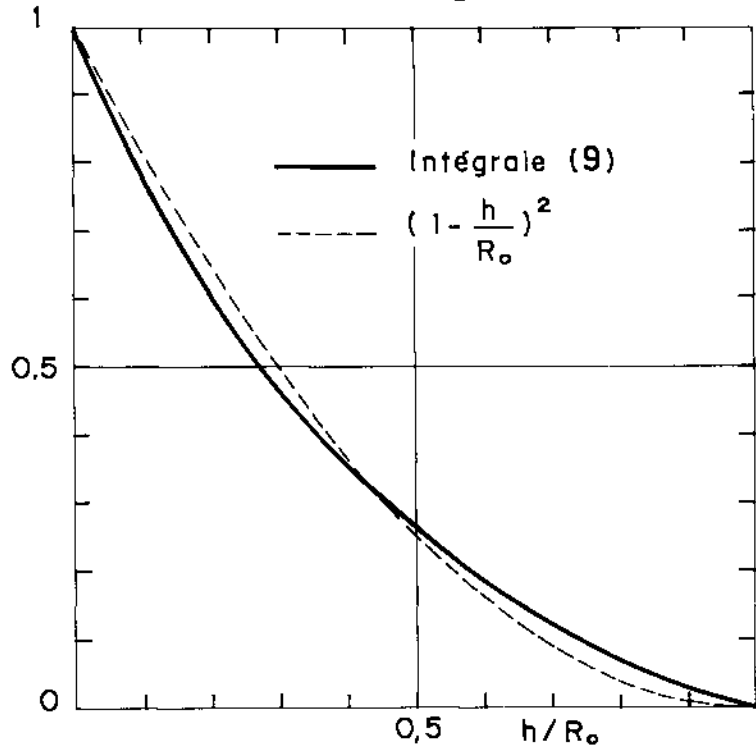


Fig. 4 - Valeurs de l'intégrale (9) en fonction du rapport  $h/R_0$

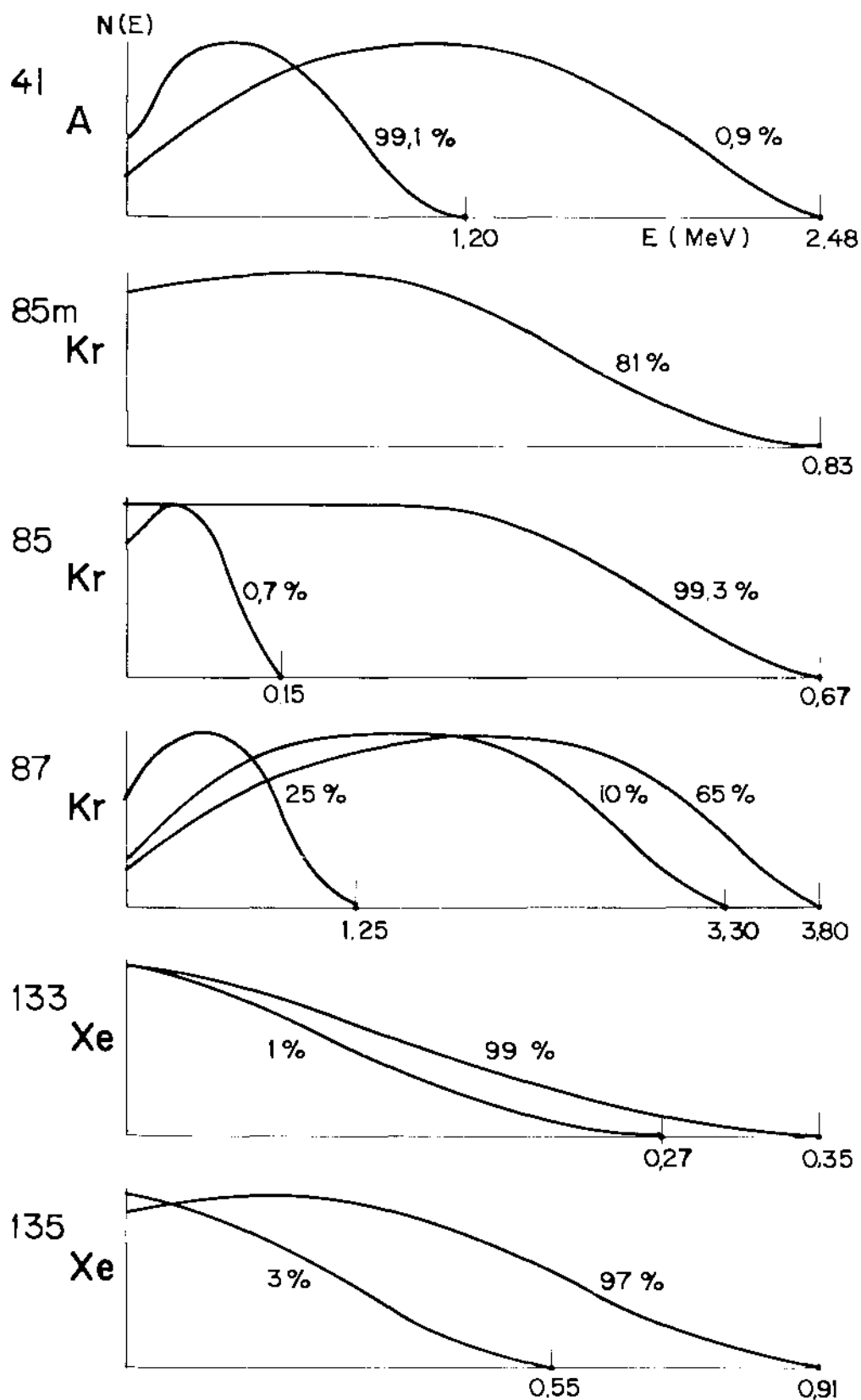


Fig. 5 - Spectres d'énergie bêta

Nous avons effectué ce calcul en mesurant les surfaces limitées par les courbes  $N(E)$  et  $N(E) D(E)$  tracées en fonction de  $E$ .

## 2.2 - Débits de dose délivrés par les rayonnements gamma

Pour les rayonnements gamma nous pouvons adopter la méthode de calcul utilisée par l'I. C. R. P. <sup>7</sup>.

Puisque le volume d'air contaminé est supposé très grand, un état d'équilibre s'établit et l'énergie absorbée par unité de volume d'air est égale à l'énergie émise dans ce même volume. Dans ces conditions, le débit de dose dans l'air, exprimé en millirads par heure, a pour valeur :

$$\dot{D}_a = 1,64 q \eta E,$$

où  $q$  représente l'activité volumique de l'air ( $\mu\text{Ci}/\text{m}^3$ ),  $\eta$  le rendement de la désintégration gamma et  $E$  son énergie (MeV).

Le débit de dose  $\dot{D}$  dans les tissus est égal au précédent divisé par deux (puisque l'organisme est irradié sous un demi-angle solide en raison de la présence du sol) et multiplié par le rapport des kerma dans les deux milieux, égal à 1,11 :

$$\dot{D} = 0,91 q \eta E.$$

Nous ne tiendrons pas compte de l'atténuation linéique du rayonnement gamma dans les premiers centimètres de l'organisme, car elle est en général compensée par un fort "buildup factor" <sup>8</sup>.

## 3 - Résultats

Tous nos résultats correspondent à une activité volumique de l'air de 1 microcurie par mètre cube.

La variation du débit de dose absorbé, en fonction de la profondeur dans l'organisme et de l'énergie, est représentée, pour les électrons mono-énergétiques, sur la figure 6.

Nous avons considéré les trois organes critiques les plus proches de la surface du corps et pour lesquels l'I. C. R. P. donne les profondeurs effectives <sup>9</sup>:

- la peau, dont la partie radiosensible est représentée par la couche basale de l'épiderme,  $h = 70 \mu$ ,
- le cristallin,  $h = 3 \text{ mm}$ ,
- les testicules,  $h = 1 \text{ cm}$ .

Les valeurs des débits de dose délivrés à ces organes par les rayonnements bêta et les rayonnements gamma émis par les six principaux gaz rares rejetés par les centrales sont rapportées dans le tableau 1.

Tableau 1 - Débits de dose (mrad/h) délivrés aux organes par les gaz rares selon le rayonnement émis, pour  $1 \mu\text{Ci}/\text{m}^3$

gaz	$\beta$			organisme entier
	peau	cristallin	testicules	
<sup>41</sup> A	0,37	0,005	0	1,16
<sup>85m</sup> Kr	0,17	0	0	0,16
<sup>85</sup> Kr	0,17	0	0	0,0033
<sup>87</sup> Kr	0,83	0,26	0	0,90
<sup>133</sup> Xe	0,04	0	0	0,074
<sup>135</sup> Xe	0,23	0	0	0,24

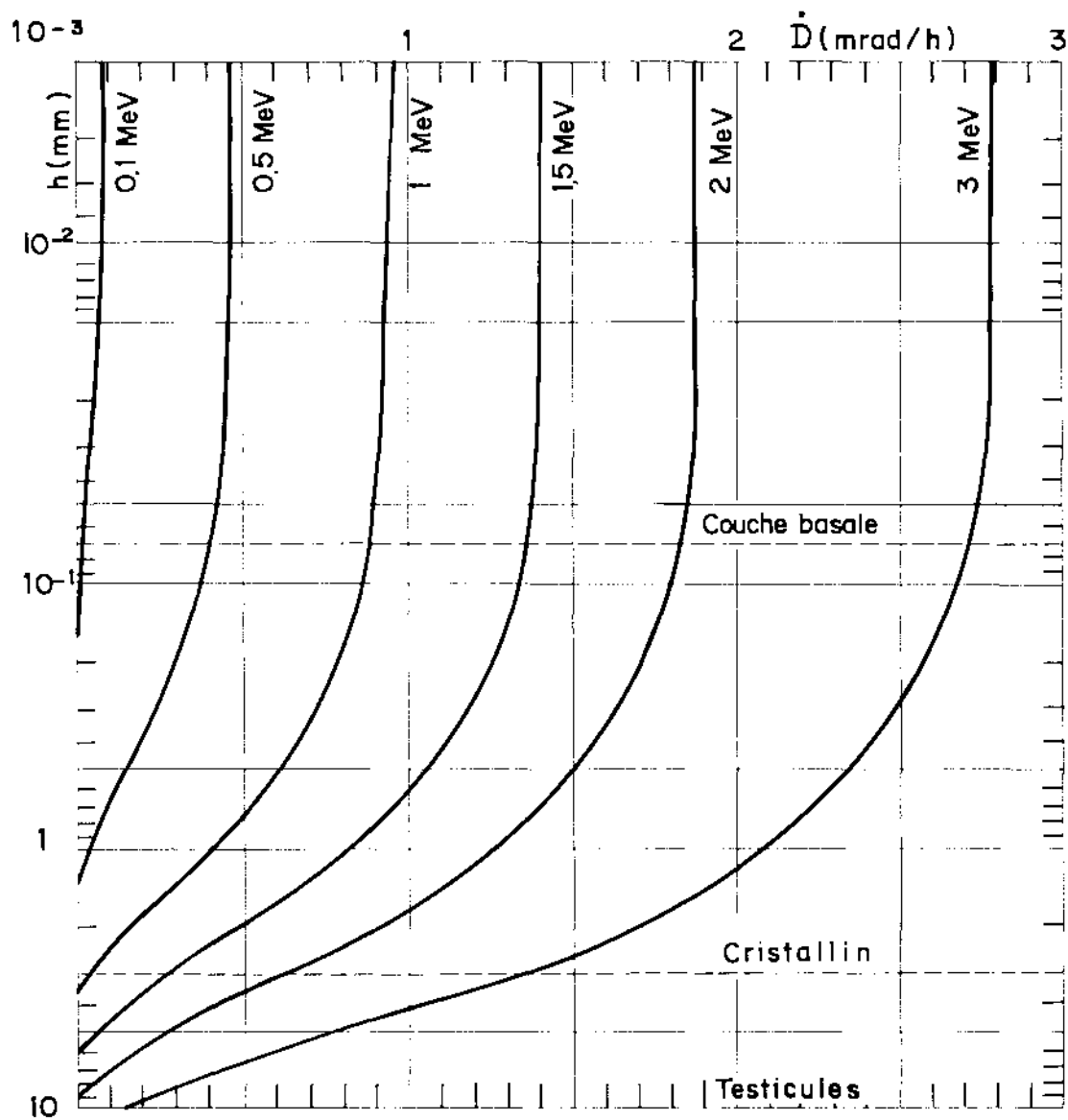


Fig. 6 - Variation du débit de dose  $\dot{D}$  en fonction de la profondeur  $h$  dans l'organisme et de l'énergie initiale de l'électron, pour une activité volumique de l'air de  $1 \mu\text{Ci}/\text{m}^3$ .

#### 4 - Discussion

Nos résultats montrent que l'atténuation des rayonnements bêta est relativement importante pour les basses énergies au niveau de la couche basale de l'épiderme et toujours très forte au niveau des cristallins. Les testicules ne sont jamais atteints par ces rayonnements, même si l'on néglige la protection par les vêtements; il en est évidemment de même pour les deux autres organes les plus radiosensibles, ovaires et organes hématopoïétiques, puisqu'ils sont situés à des profondeurs effectives encore plus grandes<sup>9</sup> (respectivement 7 et 5 centimètres).



La méthode de calcul utilisée par l'I. C. R. P.<sup>7</sup> ne tient pas compte de cette atténuation, mais traite les rayonnements bêta comme les rayonnements gamma. Il est donc intéressant de comparer les valeurs que l'on peut tirer de cette étude pour les concentrations maximales admissibles (CMA) des gaz rares dans l'air à celles recommandées par l'I. C. R. P.<sup>7</sup>.

Une seconde question mérite attention : il faut vérifier que les débits de dose qui résulteraient d'une irradiation interne consécutive à l'inhalation des gaz ne l'emportent pas sur ceux qui sont dus à l'irradiation externe seule.

Nous discuterons successivement ces deux points.

#### 4.1 - Comparaison des valeurs des CMA obtenues ici à celles recommandées par l'I. C. R. P.

Ces valeurs sont facilement calculables à partir du tableau 1, où les débits de dose délivrés par les rayonnements gamma à l'organisme entier doivent toujours être ajoutés à ceux qui sont délivrés par les rayonnements bêta, puisque tout organe particulier fait partie de l'organisme entier. Établies pour une exposition professionnelle de 168 heures par semaine, elles sont rapportées dans le tableau 2.

Tableau 2 - Valeurs des CMA ( $\mu\text{Ci}/\text{m}^3$ ) des gaz rares dans l'air pour une exposition professionnelle de 168 h/semaine.

gaz	valeurs trouvées, selon l'organe de référence			valeurs de l'I. C. R. P. (org. entier)
	peau	cristallin	org. entier	
<sup>41</sup> A	2,2	1,5	0,5	0,4
<sup>85m</sup> Kr	10	10	3,5	1
<sup>85</sup> Kr	20	520	170	3
<sup>87</sup> Kr	1,7	1,2	0,6	0,2
<sup>133</sup> Xe	30	23	7,7	3
<sup>135</sup> Xe	7,3	7,2	2,4	1

Ce tableau montre que la plupart des CMA recommandées jusqu'à présent par l'I. C. R. P. pourraient être fortement augmentées. Pour le krypton 85, l'organe critique est la peau au lieu de l'organisme entier.

#### 4.2 - Irradiation interne consécutive à l'inhalation du gaz

L'inhalation des gaz rares peut entraîner d'abord l'irradiation des poumons, puis, en raison de la solubilité de ces gaz dans les fluides de l'organisme, la contamination de tissus particuliers, tels que les graisses.

4.2.1 - Irradiation des poumons. Le débit de dose délivré au tissu pulmonaire par l'air inhalé est donné, en millirads par heure, par l'expression :

$$\dot{D} = 2,13 \frac{\epsilon}{m} q \quad (10)$$

où  $\epsilon$  représente l'énergie effective (MeV),  $m$  la masse (g) des poumons,  $q$  l'activité ( $\mu\text{Ci}$ ) présente dans les poumons.

L'énergie effective est calculée selon la méthode indiquée par l'I. C. R. P.<sup>7</sup>. La capacité pulmonaire totale étant voisine de 5 litres, à une contamination de l'atmosphère de  $1 \mu\text{Ci}/\text{m}^3$  correspond une valeur de  $q$  égale à  $5 \times 10^{-3} \mu\text{Ci}$ . La valeur de  $m$  est 1000 g.

Le débit de dose total reçu par les poumons est la somme du débit précédemment calculé et de celui délivré à l'organisme entier par les rayonnements gamma du nuage radioactif, dont la valeur est indiquée au tableau 1.

On en déduit les valeurs des CMA relatives aux poumons, rapportées dans le tableau 3 .

Tableau 3 - CMA ( $\mu\text{Ci}/\text{m}^3$ ) des gaz rares dans l'air pour les poumons.

gaz	$^{41}\text{A}$	$^{85\text{m}}\text{Kr}$	$^{85}\text{Kr}$	$^{87}\text{Kr}$	$^{133}\text{Xe}$	$^{135}\text{Xe}$
CMA	1,5	10	294	1,9	22	7

La comparaison des tableaux 2 et 3 montre que les poumons ne constituent jamais l'organe critique.

4.2.2 - Irradiation par solubilisation. Nous prendrons l'exemple du krypton 85. Ce gaz est rapidement distribué dans la plus grande partie du corps en raison de sa solubilité relative dans le sang et plus particulièrement dans les graisses<sup>10</sup>. On admet qu'une contamination de l'air de  $1 \mu\text{Ci}/\text{m}^3$  entraîne une concentration de krypton 85 dans le sang de 0,04 pCi/g et dans les graisses de 0,4 pCi/g. Les débits de dose correspondants peuvent être calculés à l'aide de l'expression (10) où les concentrations précédentes sont représentées par le rapport q/m. On trouve :

$$\begin{aligned} \text{sang : } \dot{D} &= 2 \times 10^{-8} \text{ mrad/h} \\ \text{graisses : } \dot{D} &= 2 \times 10^{-7} \text{ mrad/h} \end{aligned}$$

Ces valeurs sont tout à fait négligeables devant les débits de dose qui sont dus à l'irradiation externe.

#### 5 - Conclusion

Nous nous sommes efforcés de baser nos calculs sur une théorie rationnelle plutôt que de recourir à des formules empiriques<sup>11</sup>, qui supposent que les milieux contenant la source et la cible sont identiques et dont les valeurs à attribuer aux différents paramètres sont souvent incertaines.

Nos résultats montrent que la plupart des CMA recommandées jusqu'à présent par l'I. C. R. P. pourraient être fortement augmentées; nous trouvons notamment que le rayonnement du krypton 85 rend ce gaz noble moins nocif que ne pourraient le faire croire la longueur de sa période et une valeur de CMA de  $3 \mu\text{Ci}/\text{m}^3$ .

#### Références

1. BERGER, M. J. and SELTZER, S. M. (1964) Energy spectra and angular distributions of electrons transmitted through sapphire ( $\text{Al}_2\text{O}_3$ ) foils - NASA (Natl. Aeron. Space Admin.) SP-3008.
2. CHARLTON, D. E. and CORMACK, D. V., Radiation Res., 17 (1962), 34-49.
3. CHARLTON, D. E. and CORMACK, D. V., Brit. J. Radiol., 35 (1962), 473-477.
4. MARSHALL, J. H., Nucleonics, 13 (1955), 34-38.
5. EVANS R. D., The Atomic Nucleus, Mc Graw-Hill, New-york (1955), 536-566.
6. Nuclear Data Sheets, Academic Press Inc., New-York (1966).
7. Report of Committee II on Permissible Dose for Internal Radiation, ICRP publication 2, Pergamon Press, Oxford (1960).
8. DELPLA, M. et SCHAEFFER, R. Les organes critiques et les rayons X mous. Problèmes de radioprotection liés à l'émission de rayons X parasites par des systèmes électroniques, Colloque international, Toulouse,

- 1970, Commission des Communautés européennes, EUR 4640 d-f-e, Luxembourg (1971), 523-544.
9. Recommandations de la Commission Internationale de Protection contre les Radiations, Journal de Radiologie, 36, n°10 bis (1955), 9.
  10. DIETHORN W.S. and STOCKHIO, W.L., Health Phys., 23 (1972), 653-662.
  11. LOEVINGER R., JAPHA E.M., and BROWNELL G.L., Discrete radioisotope sources, Radiation Dosimetry, Academic Press inc. New York (1956), 693-799.

CORRELATION BETWEEN FAST NEUTRON DOSE EQUIVALENTS  
AND IN VIVO ACTIVATION PRODUCTS

T. Schlesinger and Y. Klopfer  
Radiation Safety Department  
Soreq Nuclear Research Centre  
Yavne, Israel

Abstract

The possibility of establishing a quantitative relationship between fast neutron dose and specific activation products in living tissue is discussed. Such a relationship may be useful for fast neutron dose evaluation in routine neutron therapy and following accidental neutron over-exposures.

Tissue equivalent phantoms of different dimensions were exposed to 15 MeV D-T neutrons from a linear ion accelerator. The radioactivity of activation products, such as  $^{13}\text{N}$  from the  $^{14}\text{N}(n,2n)^{13}\text{N}$  reaction,  $^{28}\text{Al}$  from the  $^{31}\text{P}(n,\alpha)^{28}\text{Al}$  reaction,  $^{49}\text{Ca}$  and  $^{38}\text{Cl}$  from  $(n,\gamma)$  reactions, were measured immediately after irradiation by a total body counter with a sensitivity of the order of 1 nanocurie.

The neutron dose is calculated and correlated to the activity of some of the activation products.

Introduction

The use of fast neutron beams for radiotherapy has recently been extended due to the ability of such beams to deliver a more effective radiation dose to tumors containing anoxic cells<sup>1</sup>. The development of convenient quantitative methods for the measurement of absorbed doses and dose equivalents of fast neutrons in the human body is one of the central problems connected with this new trend in radiation therapy<sup>2</sup>. In the present work the quantitative relationship between absorbed doses and dose equivalents of 15 MeV neutrons and the amount of specific radioisotopes produced by these neutrons in tissue has been experimentally examined. Similar methods have already been applied for neutron dose-equivalent estimations following criticality accidents, by the measurement of Na-24 activity in the body and P-32 activity in the human hair and nails<sup>3,4</sup>.

Experimental

In all experiments a ten minute irradiation time was used.

A cylindrical phantom 27 cm in dia.  $\times$  45 cm high filled with tissue equivalent liquid was exposed to the broad neutron beam (Fig.1).

Neutron fluence values were determined using several activation detectors with different threshold energies. Radioisotopes produced in the tissue equivalent liquid e.g. N-13, Cl-38, Ca-49 and Na-24 were identified and measured by a whole body counter and other high sensitivity gamma spectrometers immediately after neutron exposure. The absorbed doses and dose equivalents in the phantom were calculated from the measured neutron fluence values, using theoretical

Monte Carlo calculation results for cylindrical phantoms of similar dimensions<sup>5-8</sup>.

Neutron source

The neutron beam was produced by the D-T reaction using a Texas Nuclear series 9500 model 150 - IH ion accelerator. The neutron yield was of the order of  $10^{10}$  nsec<sup>-1</sup>. Incident neutron fluxes of the order of  $3 \times 10^5$  n cm<sup>-2</sup>sec<sup>-1</sup> could therefore be obtained with source-to-phantom distance 50 cm (Fig.1).

Phantoms

We prepared the tissue equivalent phantom from the following mixture.

<u>Material</u>	<u>% by weight</u>	<u>Element</u>	<u>% by weight</u>
Water	57	O	68
Gelatin	9.5	H	10
Glucose	19.0	C	16
Urea	4.75	N	3.3
Calcium nitrate	9.35	Ca	1.58
Sodium chloride	0.32	P	0.95
		Cl	0.19
		Na	0.12

Neutron fluence measurements

The thermal component of the fluence and the cadmium-ratio were measured by a pair of gold wires, one bare and other covered with a 1 mm thick cadmium tube.

To measure the fast neutron fluence and to get information on the energy distribution of the neutrons in the incident and attenuated beam, we used the different threshold reactions described in Table 1. Some of the prominent activation products are also given in this table.

The sulfur pellets were manufactured and counted according to recommendations of the IAEA reported by W. Kohler (1969).

The five inner bottles illustrated in Fig. 1 were used to sample the specific activity of the tissue equivalent liquid at different depths in the phantom. After stirring, to homogenize the contents, about 15 ml of liquid from each inner bottle were sampled and measured separately in the low level gamma spectrometer within 10-30 minutes after irradiation. The main bottle containing 25 liter of the TE liquid was counted in scanning bed geometry in a Whole Body Counter after homogenization of its content.

The Whole Body Counter consists of a shadow-shield, 9" dia. x 3" high NaI(Tl) crystal made by Quartz & Silice. Its construction permits the use of scanning bed geometry. A 10 minute scan assures a detection limit of about 1 nanocurie.

TABLE 1  
Physical properties of fluence monitors and prominent activation products

<u>Reaction</u>	<u>Threshold</u>	<u>Target is. abundance</u>	<u>Cross Section (14 MeV)</u>	<u>Half-life</u>	<u>E[MeV]</u>	<u>(abundance)</u>
$^{63}\text{Cu}(n,2n)^{62}\text{Cu}$	11.8 MeV	69.2%	0.55b	9.7 min	0.511	(195%)
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	6 MeV	100%	0.12b	14.9 h	1.37	(100%)
$^{32}\text{S}(n,p)^{32}\text{P}$	2 MeV	95%	0.3b	14.3 d	1.71	(100%) <u>beta</u>
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	thermal	100%	96b(th)	2.7 d	0.412	(95%)
$^{14}\text{N}(n,2n)^{13}\text{N}$	11.3 MeV	99%	6mb	10 min	0.511	(200%)
$^{37}\text{Cl}(n,\gamma)^{38}\text{Cl}$	thermal	24.5%	0.56b(th)	37.3 min	2.17	(47%)

## Absorbed dose and dose equivalent determinations

Absorbed doses and dose equivalents were calculated from the measured fluence values. Dose equivalents were also measured with an Anderson-Braun type neutron Rem-counter (corrected for energy response).

## Results and Discussion

Neutron fluence values: Neutron fluence values calculated from the activity of activation products (from the reactions described in Table 1) are given in Figs. 2 and 3 for different depths in the phantom. The expected error in the determination of the exact position of the detectors and in the calculation of the fluence values are also indicated.

The flux of incident fast neutrons (Fig.2) with energies above 6-8 MeV was  $3.5 \times 10^5 \text{ n cm}^{-2} \text{ sec}^{-1}$ . This is the mean value calculated from the activity of Cu-62 and Na-24 (from Al-27). The flux of fast neutrons measured by these detectors decreased monotonically with depth in the phantom to a mean value of  $1.1 \times 10^5 \text{ n cm}^{-2} \text{ sec}^{-1}$  at a depth of 13.5 cm and to a mean value of about  $6 \times 10^4 \text{ n cm}^{-2} \text{ sec}^{-1}$  at a depth of 25 cm. Since apparently most of this flux is due to those neutrons which did not undergo collisions, a significant part of this decrease is attributed to the geometrical attenuation (our source was a point source).

The flux of neutrons with energies above 2 MeV measured with the sulphur pellets decreased with depth at a much slower rate.

Although the threshold energies for the production of Cu-62 from Cu-63 and N-13 from N-14 are very close, the fluence values calculated from the activity of these detectors were different at some depths. This discrepancy can partly be explained by the large statistical errors in counting due to the very low activity of N-13 obtained in our experiments because of the low neutron yield and the low cross section for N-13 production. Because of the relatively long half-life of P-32 and the short irradiation time (10 min) the activities produced in the sulphur-pellets were also very low and therefore the error in the fluence values calculated from the P-32 activity was also high.

The measured thermal-neutron fluxes and cadmium-ratios at different depths in the phantom are shown in Fig.3. A peak for both parameters is observed a few centimeters under the surface of the phantom. This is in agreement with published results<sup>9,10</sup>.

Activation products in the phantom: Identification and measurement of the activation products in the tissue equivalent liquid within the phantom were carried out using the Whole Body Counter and Ge(Li) spectrometer. Four radioisotopes, N-13, Ca-49, Na-24 and Cl-38, were identified but only N-13 and Cl-38 were measured quantitatively. The total activity of N-13 in the main phantom at the end of irradiation was found to be 620 nc, and the specific activity was therefore  $2.55 \times 10^{-5} \text{ } \mu\text{c/g}$  phantom. The specific activities of N-13 in the five inner bottles are given in Table 2.

TABLE 2  
N-13 activity as a function of depth in the phantom

Bottle No.	Depth of bottle center, cm	Specific activity of N-13 $\mu\text{c/g}$ phantom
1	2.5	$2.7 \times 10^{-5}$
2	8.0	$1.3 \times 10^{-5}$
3	13.5	$1.6 \times 10^{-5}$
4	18.5	$1.7 \times 10^{-5}$
5	24.1	$0.8 \times 10^{-5}$

The total activity of Cl-38 at the end of irradiation was 65.2 nc and the specific activity was  $2.6 \times 10^{-6}$   $\mu\text{c/g}$  phantom.

Absorbed doses, dose equivalents and their relation to the activation products: Absorbed dose and dose equivalent values for different depths in the phantom were calculated using fluence-dose conversion factors based on Monte-Carlo calculation results as explained above. According to these calculations<sup>5</sup> the conversion factor for 14 MeV incident neutrons is  $7.0 - 7.5 \times 10^{-9}$  Rad/unit fluence for the first few cm in the phantom, decreasing to  $5 \times 10^{-9}$  at 14 cm,  $3 \times 10^{-9}$  at 22 cm and to  $2 \times 10^{-9}$  at a depth of 27 cm. The contribution of gamma dose to these absorbed dose values are between 5-10%. The effective quality factor for 14 MeV incident neutrons changes only slightly with depth in the phantom from a value of 7.5 at the point of incidence to a value of 7 at a depth of 15 cm, increasing again to above 7.5 at 25 cm<sup>5</sup>.

According to these conversion factors and using the measured incident flux value of  $3.5 \times 10^5$   $\text{n cm}^{-2} \text{sec}^{-1}$  and irradiation time of 600 sec, we obtained an absorbed dose of 1.5 - 1.6 Rad in the first few cm of the phantom, about 1 Rad at a depth of 14 cm and 0.5 Rad at 27 cm. The corresponding dose equivalent values are 11.3 - 12.0 Rem for the first few cm, 7 Rem at 14 cm and 3.8 Rem at a depth of 27 cm.

As mentioned above, the conversion-factors we used were calculated for broad beams. In our case we had a point source and a source-to-phantom distance of 50 cm, leading to appreciable geometrical attenuation. This attenuation amounts to a factor of 1.6 at a depth of 14 cm and to a factor of 2.4 at a depth of 27 cm. We did not correct for these factors which mainly affect the part of the beam which did not undergo collisions.

The gamma-dose measured by photographic film was found to change only slightly with depth, having a mean value of 170 mRad i.e. 15-18% of the mean absorbed dose, but only about 2-3% of the mean dose-equivalent.

The dose equivalent was also measured by the Rem counter which indicated a dose equivalent of 19.2 Rem if neutrons of 14.8 MeV are assumed, 9.6 Rem for neutrons of 12 MeV and 7.7 Rem for neutrons of 10 MeV. Since the Rem counter is calibrated to measure maximal dose equivalent for radiation protection purposes its indications represent the dose equivalent in the first few cm of the phantom. This is in agreement with the dose equivalent of about 11.5 Rem calculated from the incident fluence value.

Assuming a delay of ten minutes between the end of irradiation and the start of counting, our results indicate that an activity of about 40 nanocuries/Rem N-13 and about 60 nanocuries/Rem Cl-38 are available for measurement (activity per mean dose equivalent).

#### Summary

The preliminary experiments reported in this paper demonstrate the possible use of Whole Body Counters or even simpler devices, e.g. shadow shielded detectors, as routine instruments for dosimetry purposes in neutron therapy. In our experiments the delivered doses were of the order of 0.5-2 Rads and the dose equivalents were of the order of 4 - 12 Rems. In actual neutron therapy the delivered doses are at least one order of magnitude higher and this is also true for the amount of activation products. On the other hand the irradiation conditions used in therapy are completely different mainly because collimated beams are used. In further experiments of the kind reported here, using anthropomorphic phantoms, and collimated neutron beams similar to those used in neutron therapy, a quantitative relationship can possibly be established between the absorbed dose, dose equivalents and the activity of specific radioisotopes in the human body, following actual neutron therapy treatment.

### References

1. J.F. FOWLER, D.K. BEWLEY, "Applications of Fast Neutron Beams in Radiation Therapy" p.239, Radiobiological Applications of Neutron Irradiation, Proceedings of a Panel, Vienna, 6-10 December 1971 (IAEA, Vienna, 1972).
2. J.J. BROERSE, J.E. BROERS-CHALLISS and T. MARUYAMA, EURATOM Report EUR 4896 d-f-e, Vol.II, p.627 (1972).
3. M. DOUSSET, A. RICOURT, N. PARMENTIER et al., in Symposium on Neutron Monitorings for Radiation Protection Purposes, Vienna, 1972 (in press).
4. M. TRAJKOVIC, I. MIRIC, Z. UBOVIC, Proceedings of a symposium, Advances in Physical and Biological Radiation Detectors, p.697, Vienna (IAEA, Vienna, 1971).
5. Protection Against Neutron Radiation, NCRP Report No.38 (1971).
6. J.A. AUXIER, W.S. SNYDER and T.D. JONES, Radiation Dosimetry (Academic Press, New York, 1968) Vol.1, p.275.
7. W.S. SNYDER and T.D. JONES, in ref. 2 Vol. II p.597.
8. J.A. AUXIER, ref.3.
9. K. BODDY et al., Proceedings of a symposium on Nuclear Activation Techniques in the Life Sciences (IAEA, Vienna, 1972).
10. J. ANDERSON, C.K. BATTYE, S.B. OSBORN, R.W.S. TOMLINSON, *ibid.* p.571.

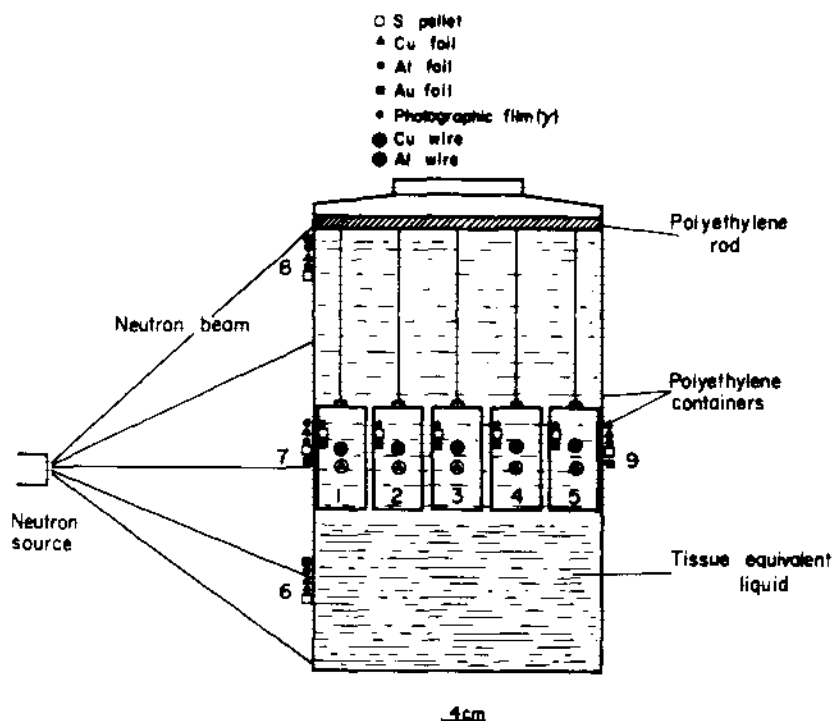


Fig.1. Schematic diagram of irradiation geometry and phantom dimensions. A 27 cm dia. x 43 cm high polyethylene cylinder filled with tissue equivalent liquid is exposed to the neutron beam. Different activation monitors are used to measure neutron fluence. Five inner bottles filled with the same liquid are used to sample and measure activation products in the liquid after irradiation.



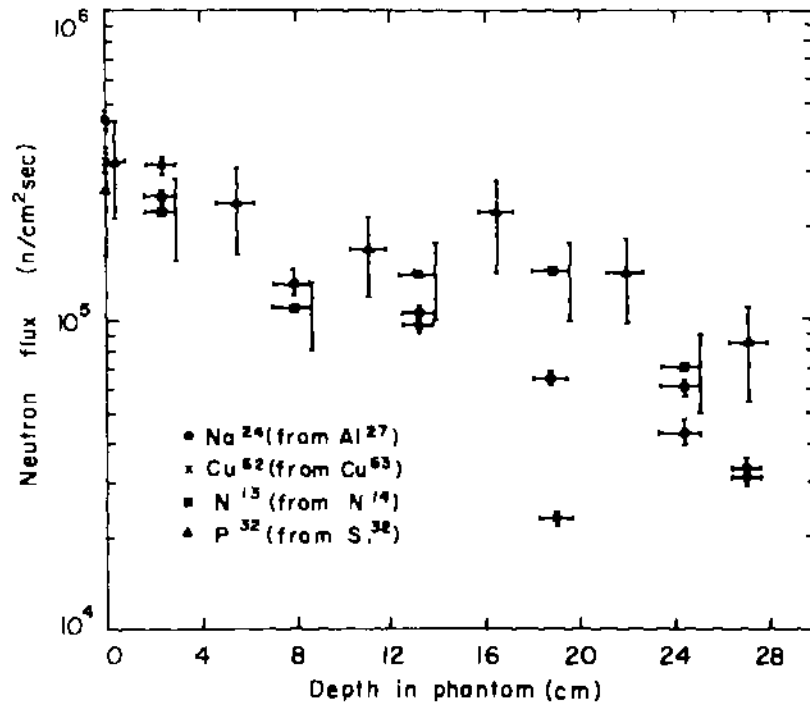


Fig.2. Fast neutron fluence values measured by the activation detectors at different depths in the phantom. (The error bars are slightly shifted at some of the points in order not to interfere with other error bars.)

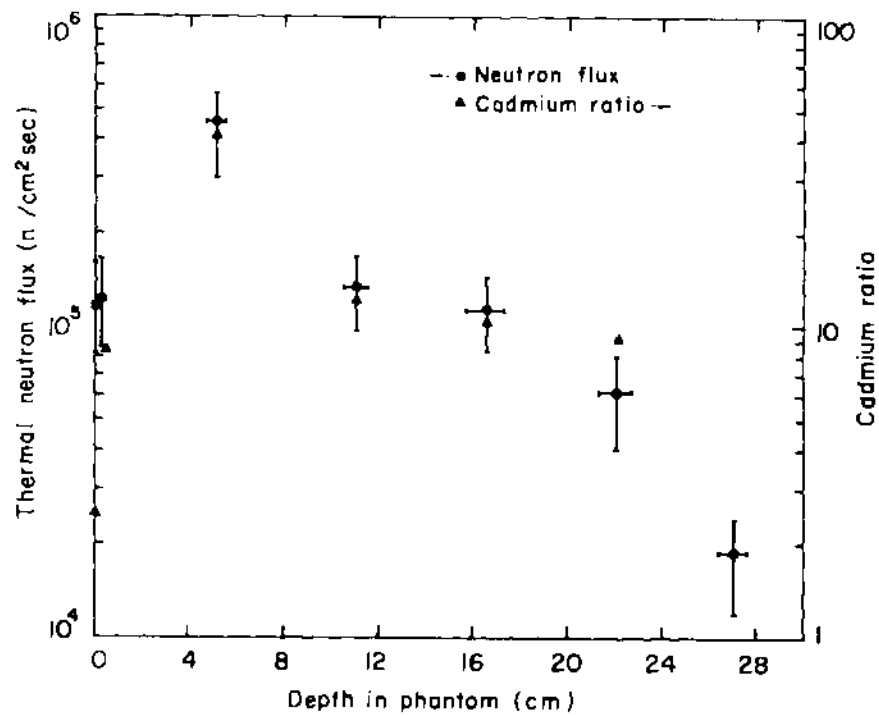


Fig.3. Thermal neutron flux at different depths in the phantom

DETERMINATION OF ABSORBED ENERGY FOR VARIOUS TYPES  
OF MEDICAL IRRADIATIONS

L. Bozóky  
Central Institute for Oncology, Budapest, Hungary

I. Fehér  
Central Research Institute for Physics, Budapest,  
Hungary

Abstract

LiF thermoluminescent powder /TLD/ was suspended in paraffin alkyl chloride /PAC/ paste and this modulable dosimeter was used to simulate tumours of different shapes in order to evaluate the dose absorbed by the tumour in different types of radiotherapeutical treatment under similar to in-vivo conditions. The results for the following types of treatment are reported: X-ray beam therapy with Chaoul-machine, interstitial irradiation with radium needles, gynaecological intracavitary irradiation with cobalt applicator, extracorporal beta-irradiation with strontium-90, gamma ray beam therapy with cobalt-60 source, Bremsstrahlung and electron irradiations with a 25 MeV betatron. In case of homogeneous radiation field the yields of ionization chambers and the TLD-PAC dosimeters are in good agreement. The TLD-PAC technique was found to be particularly useful for the evaluation of the absorbed energy in anatomical formations which are difficult to follow-up by calculation and its application is thought to be of special importance in the case of inhomogeneous radiation fields.

Introduction

In radiotherapy used up-to-day methods partly achieve the nearly homogeneous irradiation of the entire tumour by careful treatment planning, but definitely inhomogeneous irradiation techniques can also be employed. These two essentially different procedures have been compared by direct measurements in some typical radiotherapeutical arrangements. For this purpose a method was needed which permits the direct measurement of the total energy absorbed by the tumour in both cases. The TL powder technique based on physical integration, which is applicable to a wide range of doses, proved to be appropriate for the planned measurements. Tumours of 60 ccm volume, thus of medium size were irradiated by a cobalt-60 source and as well as by a Bremsstrahlung and electron beam of a 25 MeV betatron, using focal skin distances /F.S.D./ from 60 to 100 cm, while small sized tumours were treated by X-ray beam therapy with Chaoul machine, interstitial irradiation with radium needles, gynaecological intracavitary irradiation with radium or cobalt applicator and extracorporal irradiation from little distances. Then measurements with radium needles were carried out in three arrangements, that is the needles were placed into plexiglass capsules having 1 and 2 mm thick walls too in or-

der to suppress the contribution to the absorbed dose from the immediate surroundings of the needles.

### Experimental

The total energy absorbed by the tumour phantom was measured by the method of physical integration using TLD 100 LiF powder suspended in paraffin n-alkyl /C<sub>20-22</sub>/ chloride /PAC/ mixture, varying the LiF concentration from 0.1 to 1.0 %. In our earlier investigations<sup>1</sup> the method proved to be useful for measuring the average absorbed dose. Its advantages are the following:

- LiF powder does not deposit from the PAC,
- TLD-PAC can be easily moulded to simulate the shape of a tumour,
- PAC is tissue-equivalent at energies above 10 keV,
- LiF can be recovered from cold PAC with CCl<sub>4</sub> and after decantation the thermoluminescent effect of the sample can be measured,
- the LiF TLD yield is not affected by the treatment.

The LiF TLD powder was measured by the TLD reader developed in our laboratory. The gamma radiation of the cobalt-60 source was used for calibration. During the calibration of the LiF-PAC sample and the LiF powder were placed between two, 4 mm thick each, plexiglass plates. The doses were measured with condenser ionization chambers without handle, developed at our laboratory<sup>2</sup> and calibrated by IAEA measurements. For the calibration the accuracy of the dose measurements was  $\pm 3$  %. For the irradiation with Chaoul-machine, a Siemens phantom chamber was used for dosimetry.

The results of the TLD measurements were found to be reproducible to  $\pm 2$  %. The standard deviation on the repeated runs was less than  $10$  %. This value obviously includes the contribution from the deviation in the geometrical factors.

Except for Sr-90 + Y-90 radiation the maximum dose was always below 700 rad in both the calibrating and the phantom measurements. Thus the superlinearity of LiF caused no problems in the measurement. The results for Sr-90 + Y-90 were properly corrected for the superlinearity effect.

To simulate the absorption and scattering occurring in the case of in-vivo therapy the PAC phantoms were placed into a larger phantom from rice flour. Other data characterizing the different irradiation arrangements are specified in the figures.

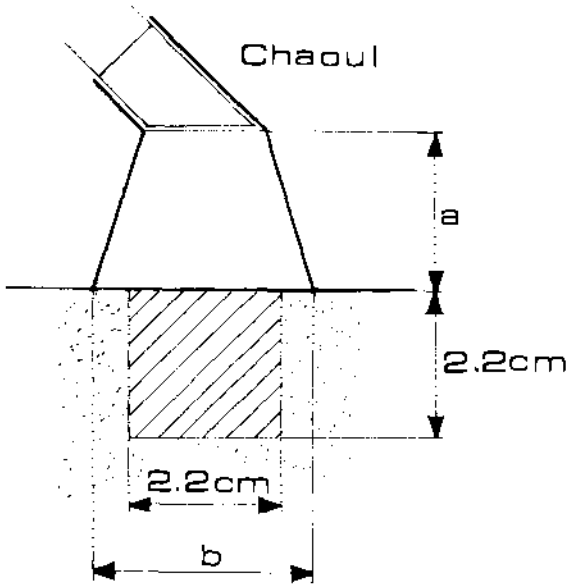
### Results

#### 1./ X-ray irradiation with Chaoul-machine

Phantoms of 10 cm<sup>3</sup> volume were irradiated at 3 and 5 cm F.S.D. using circular tubes, 2.5 and 3.5 cm in diameter, operated at 60 kVp, 4 mA.

#### 2./ Interstitial radium treatment

Phantoms of 6, 12 and 18 ccm were irradiated using 3 needles of 3 mCi activity Ra-226 each. The length of the needles was 2.7 cm, their diameter 0.18 cm and the active length 1.5 cm. The thickness of the Pt + 10 % Ir wall was 0.5 mm. The needles were inserted with a spacing of 1 cm. The run with the 12 ccm phantom



a /cm/	b /cm/	$\overline{\text{rad}}$ Siemens R
3	2	0.81
5	3	0.82

Figure 1.

X-ray irradiation with  
Chaoul-machine, 60 kVp  
and 4 mA

was repeated by placing the  
needles into plexiglass capsules  
having 1 and 2 mm thick walls.  
In this way the contribution  
from the absorption in the imme-  
diate surroundings of the need-  
les could be evaluated. Expo-  
sure times of 40 and 80 minutes  
were used.

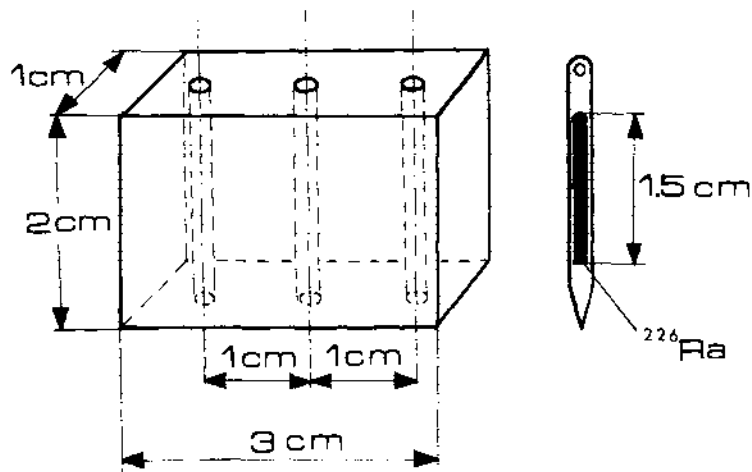


Figure 2. Interstitial radium treatment with 3 needles

volume, ccm	6	12	18
average absorbed dose in rad/mCi.h	11.1	9.1	8.7
absorbed energy in erg/mCi.h	6600	10900	15600
average absorbed dose in rad/9 mCi.day	2400	2000	1900

capsula wall thickness, mm	0	1	2
absorbed energy in 12 ccm, erg/mCi.h	10900	7600	5800

3./ Gynaecological intracavitary treatment with  $^{60}\text{Co}$  applicator

volume	50 ccm
average absorbed dose	1.93 rad/mCi.h= 2300 rad/50 mCi.day

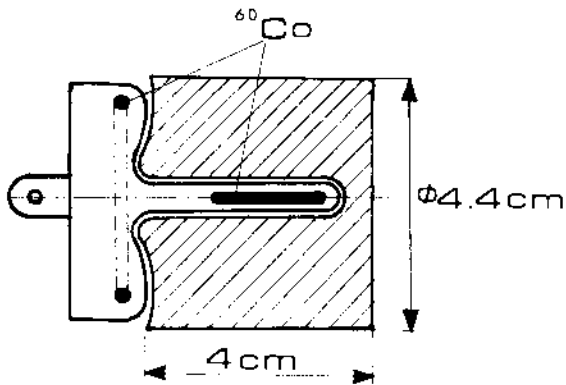
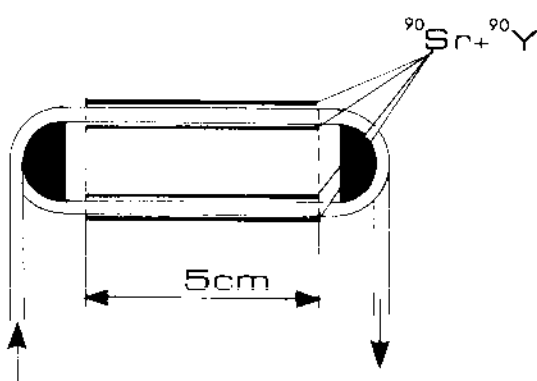


Figure 3. Gynaecological intracavitary treatment with Co-60 applicator. Activity 50 mCi

For the simulation of precarcinoma treatment a Co-60 applicator prepared from epoxy resin which has already been successfully employed, was applied. The applicator has a 3.2 cm diameter ring with an active rod protruding into the uterus. A cylindrical phantom, 4.4 cm in diameter and 4.1 cm in length was irradiated for 10 and 15 minutes with a total activity of 50 mCi. The results are presented in the table.

4./ Extracorporeal blood irradiation



blood volume, ccm	13
Sr-activity, Ci	16
average absorbed dose in rad/min	2300

Figure 4.  
 $^{90}\text{Sr} + ^{90}\text{Y}$  apparatus for extracorporeal blood irradiation

The blood is led from the artery to the vein across a thin-walled polyethylene tube which forms a coil with 5 windings. The blood is thus repeatedly exposed to high intensity beta radiation. The 16 Ci activity Sr-90 source is mounted as a closed source with 4 backing of 2x5 cm. The two pairs of plates are facing each other and form thus 2 intense beta sources spaced at 6 mm. The absorbed dose was measured under statical conditions introducing prior to the measurement warm liquid PAC into the coil of 13 ccm volume. The exposure times were 1 and 4 min.

5./ Cobalt-60 irradiation

Ionization chamber	128 rad/min	100 %
PAC-TL	120 rad/min	94 %

ROTACERT-type equipment<sup>3</sup> loaded with 3000 Ci activity Co-60 was used at 60 cm source-to-skin distance. The irradiation field was 8x10 cm<sup>2</sup>, The ellipsoidal phantom of 50 ccm volume and 1.8 cm in height was lying 5 cm below the surface of the rice flour phantom.

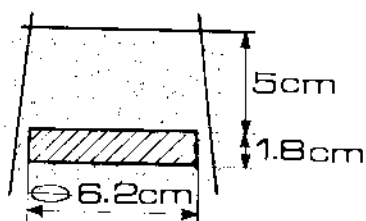


Figure 5.  
 PAC-phantom irradiation in rice flour with Co-60 gamma rays, 25 MeV Bremsstrahlung and 25 MeV electrons.

6./ 25 MeV Bremsstrahlung irradiation

A phantom of the same shape and size as that used for the Co-60 gamma irradiation, located again at 5 cm below the rice flour surface was irradiated with a 25 MeV betatron using 100 cm F.S.D. and an irradiation field of 8x10 cm<sup>2</sup>.

Ionization chamber	61 rad/min	100 %
PAC-TL	63 rad/min	103 %

## 7./ 25 MeV electron irradiation

An ellipsoidal 50 ccm PAC phantom was irradiated again with the 25 MeV betatron, using 100 cm F.S.D. and an irradiation field of  $8 \times 10 \text{ cm}^2$ , but now the surfaces of the PAC and rice flour phantoms were made to coincide.

Ionization chamber	300 rad/min	100 %
PAC-TL	277 rad/min	92.5 %

### Conclusions

1./ For the approximately homogeneous radiation fields /Co-60 gamma ray therapy, Bremsstrahlung and electron beam from 25 MeV betatron/ the PAC-TL method reproduced the dose values measured by ionization chambers to a few percent accuracy. Thus the method can be applied to inhomogeneous radiation fields too, where the ionization chambers are practically useless.

2./ If the Ra-needles are conventionally applied to a rectangular tumour for instance of  $1 \times 2 \times 3 \text{ cm}$ , three needles, 3mCi Ra activity each, yield an average dose of 2400 rad in 24 hours. Now, if a 1 mm thick, anyhow necrotised layer around the needles is left out of consideration in the evaluation of the average dose, the above dose value is reduced to 2/3 value. If a 2 mm thick layer of the tissue around the needles is disregarded, the rest of the tumour is exposed to only half of the above dose, that is to about 1200 rad/day. This is a relatively low applied dose rate even for an irradiation for two days.

3./ For the Co-60 applicator used for the gynaecological intracavitary treatment experiment, the average absorbed dose for 24 hours was measured as 2300 rad. If the deeper, in many cases actually non-existent layers simulated by our phantom are left out of consideration, the average value of the absorbed dose increases. This shows that the method is particularly useful for the accurate measurement of the average dose rate of an asymmetric tumour. One has simply to prepare a similar formation from PAC.

4./ The dose rate to be applied can be precisely evaluated even in such special cases as e.g. the extracorporal irradiation of blood.

Acknowledgements Thanks are due to Mr. Gy. Lancsarics for technical assistance in the measurements.

### References

1. Fehér, I., Koblinger, L., Szabó, P.P., Calculation and measurement of the organ doses due to internal gamma emitters. Proc. of the IRPA II. European Congress on Radiation Protection p.49. Akadémiai Kiadó, Budapest, 1973.
2. Bozóky, L., Production of small condenser ionization chambers, Magyar Radiológia, /in Hungarian/ 3. 1-3, 1951.
3. Bozóky, L., New technique for full radiation protection of personnel in telecobalt treatments, Proc. of the First International Congress of Radiation Protection, Vol.II.pp.1597-1601 Pergamon Press, Oxford 1968.

## BETA DOSE FROM CUTANEOUS CONTAMINATION

K. Unnikrishnan, R.K. Hukkoo, (Miss) Sharda Bhati  
and S. Somasundaram  
Health Physics Division (BRMS)  
Shabha Atomic Research Centre  
Bombay 400 001, India

### Abstract

Determination of beta dose to the basal layer of the skin from surface contaminants has been carried out using the latest available data on the dose distribution around point beta sources. The calculations have been performed for the thin planar source model and taken into account the effect of air tissue interface on the dose rate. As the average depth of the basal layer may vary from 4 to 40 mg/cm<sup>2</sup> depending on the body site, the variation of dose rate in this layer is discussed. Finally, for ready use in the estimation of maximum permissible skin contamination of a mixture of beta emitters, an attempt is made to provide an empirical formula for the dose rate at a depth of 7 mg/cm<sup>2</sup>, (the average value suggested by the ICRP), as a function of beta energy.

### Introduction

In the assessment of the dose to the skin from beta-gamma emitters present on its surface, the contribution from the beta component is likely to be the determining factor since beta particles in general possess enough energy to reach the sensitive cells of the basal layer underlying the epidermis and sufficiently high LET to deliver an appreciable dose. The earliest estimations of dose to the skin from the beta component were made by assuming a value of 10 rad/hr for the dose-rate from a contamination of 1  $\mu$ Ci/cm<sup>2</sup> of the skin.<sup>1</sup> Such estimates are evidently crude since no account is taken of the beta energy. More elaborate calculations were performed by Casnati and Breuer<sup>2</sup>, using the point source dose function of Rossi and Ellis, and of Loevinger and assuming a semi-infinite plane geometry. They compiled for some 10 radionuclides data on the dose-rate to the basal layer from a contamination of 1  $\mu$ Ci/cm<sup>2</sup> accumulated to a thickness of 0.001 cm on the skin. More recently Clarke and Beynon<sup>3</sup> have used Cross's point kernels for beta emitters to estimate the skin dose from fission products by arranging the latter in eight groups such that the average energy for each group is close to the average energy of one of the nuclides studied by Cross. In another recent paper, Henson<sup>4</sup> has computed depth doses in skin from surface contaminants of some medical radionuclides, using a method which is very similar to that employed in the present paper.



### Dose Calculation

In our calculations we have assumed that the penetration of the contaminant into the skin is negligible. In view of the relatively short range of beta particles in tissue, we have adopted the commonly used geometry of semi-infinite plane representing the skin surface on which an infinitely thin layer of the contaminant is present. The dose-rate at any depth in the skin may be calculated by integrating a point source dose function over appropriate limits. For this purpose, we have used the scaled absorbed dose distribution  $F$  tabulated by Berger<sup>5</sup>, since these data are available for a fairly large number of radionuclides in a form convenient for dose calculation in various media. The dose-rate  $J(x)$  from a point source of strength  $1\beta/\text{sec}$  in a medium of density  $\rho \text{ g/cm}^3$  is given by

$$J(x) = 1.6 \times 10^{-8} \frac{\bar{E}_\beta}{\rho} \frac{F(x/x_{90})}{4\pi x^2 \rho x_{90}} \quad \text{rad/sec} \quad (1)$$

where  $x_{90}$  is the 90-percentile distance within which 90% of the emitted energy is absorbed and  $\bar{E}_\beta$ , the average energy of the beta spectrum. The values of  $x_{90}$  in skin may be related to those in water, given by Berger<sup>5</sup>, by

$$x_{90} = \frac{(x_{90})_{\text{water}}}{a_{21} \rho}$$

where  $a_{21}$  is the relative attenuation factor. Assuming skin to be tissue-equivalent insofar as the average atomic number is concerned,  $a_{21} = 0.987$ . The average value of  $\rho$  for skin<sup>6</sup> is  $1.1 \text{ g/cm}^3$ . Thus for a surface activity of  $1\beta/\text{cm}^2\text{-sec}$ , the dose-rate at a depth  $d$  is given by

$$D = \frac{28.8 \bar{E}_\beta}{\rho x_{90}} \int_d^R \frac{F(x/x_{90})}{x} dx \quad \text{urad/hr} \quad (2)$$

where  $R$  is the range of the beta particles.

Since the tabulated values of  $F$  are valid only for a uniform infinite medium, the dose-rates must be corrected for the loss of scattered electrons at the tissue-air interface. The dose reduction factor as a function of the ratio of the distance from the interface to the maximum particle range, given by Berger<sup>7</sup>, is used for this purpose.

The average thickness of the epidermis adopted by ICRP<sup>8</sup> is  $7 \text{ mg/cm}^2$ . However, recent measurements by Whitton<sup>6</sup> suggest that a closer approximation to reality would be to ascribe 3 different thicknesses to three body sites. The suggested values are 4, 8 and  $40 \text{ mg/cm}^2$ . Computations have been performed for values of  $d$  corresponding to the four epidermal thicknesses mentioned above, by integrating eq.(1) numerically using the values of  $F$  and  $x_{90}$  for 24 beta emitters having  $E_\beta$  in the range  $0.0057$  (<sup>2</sup>H) -  $1.4322$  (<sup>42</sup>K) MeV. The value of  $R$  has been taken to be equal to  $2x_{90}$ ,

since  $F$  is equal or very close to zero at  $1.8x_0$ , the maximum distance for which the value of  $F$  is given in ref. 5.

### Results and Discussion

The softest beta emitter considered ( $^3\text{H}$ ) is found not to penetrate even the minimum epidermal thickness of  $4 \text{ mg/cm}^2$ . The hazard from this isotope would appear to be due to its exchange with hydrogen in tissue - an aspect which is outside the scope of this paper. This isotope will not be considered further in the discussion that follows.

The results for other radionuclides indicate that the dose-rate  $D$  at any given depth is not a smooth function of either  $\bar{E}_\beta$  or  $\bar{E}_0$ , the maximum beta energy. The manner in which  $D$  at a depth of  $7 \text{ mg/cm}^2$  varies with  $\bar{E}_\beta$  is illustrated by the inset in Fig. 1 from which it is clear that initially the dose-rate increases steeply but haphazardly with energy, but at higher energies (beyond 0.5 MeV) it tends to remain close to a value of 180 urads/hr. However, the product of  $D$  and  $\bar{E}_0$ , the weighted average of the end-point energies of all the betas emitted by an isotope appears to increase with  $\bar{E}_\beta$  in a predictable manner.  $\bar{E}_0$  was evaluated using the decay schemes given by Dillman<sup>6</sup>. Table 1 lists the values of  $\bar{E}_\beta$  and  $\bar{E}_0$  for the nuclides considered in the present work.

Table 1. The average energy,  $\bar{E}_\beta$  and the weighted average of the end-point energies  $\bar{E}_0$  for the beta spectra of various radionuclides

Nuclide	$\bar{E}_\beta$ (MeV)	$\bar{E}_0$ (MeV)	Nuclide	$\bar{E}_\beta$ (MeV)	$\bar{E}_0$ (MeV)
$^{35}\text{S}$	0.0488	0.1674	$^{22}\text{Na}$	0.216	0.546
$^{14}\text{C}$	0.0495	0.1561	$^{52}\text{Mn}$	0.2409	0.571
$^{203}\text{Hg}$	0.058	0.213	$^{198}\text{Au}$	0.3159	0.955
$^{45}\text{Ca}$	0.0773	0.252	$^{11}\text{C}$	0.385	0.980
$^{60}\text{Co}$	0.0965	0.314	$^{99}\text{Mo}$	0.3917	1.09
$^{59}\text{Fe}$	0.1185	0.392	$^{132}\text{I}$	0.5063	1.263
$^{65}\text{Zn}$	0.1433	0.325	$^{24}\text{Na}$	0.5546	1.362
$^{47}\text{Sc}$	0.1611	0.4824	$^{32}\text{P}$	0.695	1.71
$^{131}\text{I}$	0.1834	0.5818	$^{68}\text{Ga}$	0.826	1.898
$^{137}\text{Cs}$	0.1884	0.557	$^{124}\text{I}$	0.8313	1.81
$^{90}\text{Sr}$	0.1963	0.546	$^{90}\text{Y}$	0.9367	2.273
$^{58}\text{Co}$	0.2007	0.480	$^{42}\text{K}$	1.4322	3.246

Fig. 1 shows the variation of  $(D \cdot \bar{E}_0)$  at a depth of  $7 \text{ mg/cm}^2$  as a function of  $\bar{E}_\beta$ . An equation of the form

$$D \cdot \bar{E}_0 = A \cdot \bar{E}_\beta + B \quad (3)$$

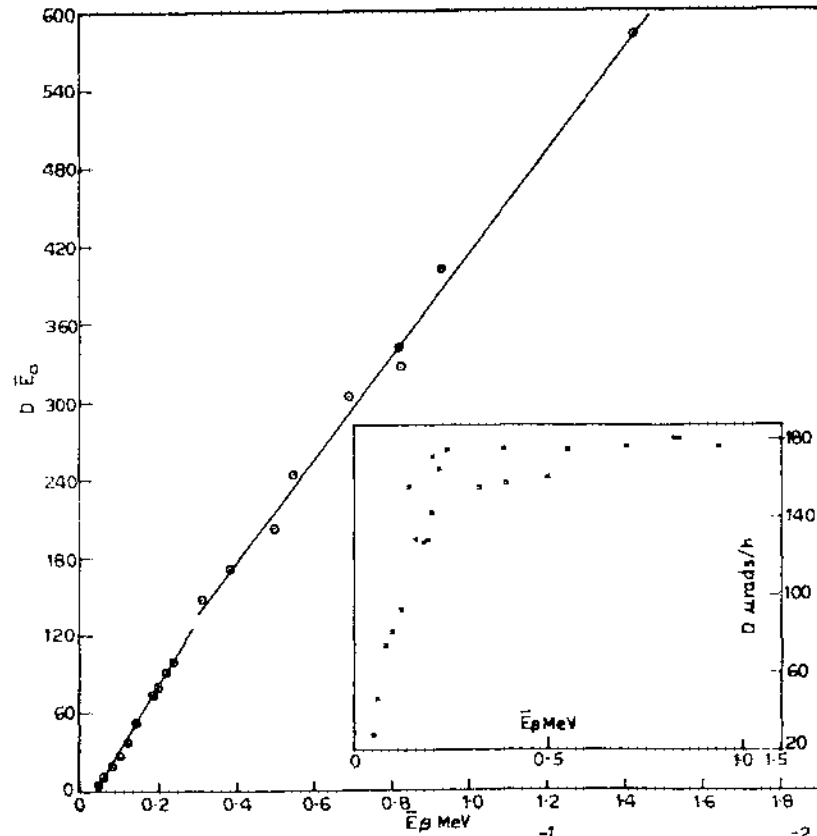


FIG. 1- THE PRODUCT OF DOSE-RATE  $D \mu\text{rad h}^{-1}$  AT A DEPTH OF  $7 \text{ mg cm}^{-2}$  AND THE WEIGHTED AVERAGE OF THE END-POINT ENERGY  $\bar{E}_0$  MeV FOR A SURFACE CONTAMINATION OF  $1 \mu\text{Sec}^{-1} \text{ cm}^{-2}$ . THE INSET SHOWS THE VARIATION OF  $D$  WITH  $\bar{E}_\beta$ .

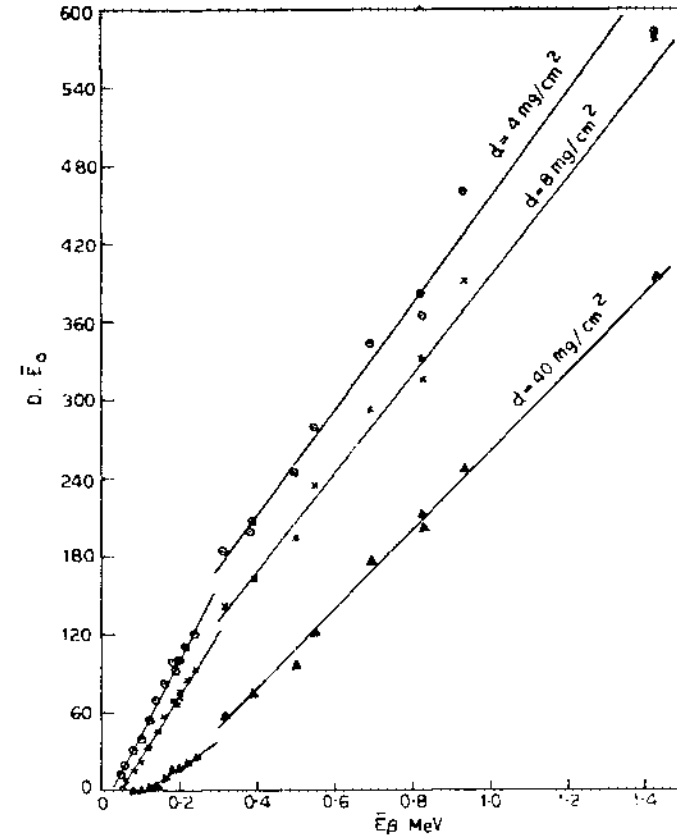


FIG. 2- THE PRODUCT OF DOSE-RATE  $D \mu\text{rad h}^{-1}$  AT DEPTH  $d$  AND THE WEIGHTED AVERAGE OF THE END-POINT ENERGY  $\bar{E}_0$  MeV, FOR A SURFACE CONTAMINATION OF  $1 \mu\text{Sec}^{-1} \text{ cm}^{-2}$ .

fits the data very well, the values of A and B depending on whether  $\bar{E}_\beta$  is less or greater than 0.3 MeV. The coefficients, as determined by a least square fit, are

$$\begin{array}{lll} A = 503.0 & B = -20.7 & (\bar{E}_\beta < 0.3 \text{ MeV}) \\ A = 393.4 & B = 17.7 & (\bar{E}_\beta > 0.3 \text{ MeV}) \end{array}$$

The average percentage deviation is 5.8 for  $\bar{E}_\beta < 0.3$  MeV and 2.9 for  $\bar{E}_\beta > 0.3$  MeV. If for the sake of simplicity a single regression equation is used over the entire range of energy, the deviation is about 30%.

Except for the extremely low energy betas below 60 keV, the values calculated using the above set of coefficients agree with the actual ones within about 10%. It seems plausible that below 60 keV another set of coefficients is required; however, no attempt is made to derive them since sufficient data are not available within this range.

The variation of  $(D \cdot \bar{E}_0)$  at other depths is shown in Fig. 2. It is apparent that the trend is more or less the same as before. However, as these epidermal thicknesses are not yet widely accepted, the corresponding data are not analysed in detail.

It should be mentioned that in our study the contribution to the dose from possible monoenergetic electron emission has not been considered. Henson<sup>4</sup> has shown that this may be appreciable for certain radionuclides. The primary objective of our work was to explore the possibility of determining the beta dose to the skin from any radionuclide for which the point source kernel is not readily available. It is felt that the use of eq.(3) with appropriate values of the coefficients involved might be useful for skin dose calculation in complicated cases such as contamination from mixtures of beta emitters.

#### References

1. Dunster, H.J., Proceedings of Symposium on "Radioactive Contamination of Workers", EURATOM Report No. EUR 2210 (1964) 135.
2. Casnati, E. and Breuer, F., Proceedings of Symposium on "Personnel Dosimetry for Radiation Accidents", IAEA, Vienna (1965) 525.
3. Clarke, R.H. and Beynon, S.M., Health Physics, 23 (1972) 807
4. Henson, P.W., Brit. J. Radiol., 45 (1972) 938
5. Berger, M.J., MIRD Pamphlet No. 7, J. Nucl. Med., 12, Supplement No. 5 (1971)
6. Whitton, J.T., Health Physics, 24 (1973) 1
7. Berger, M.J., in Medical Radionuclides: Dose and Effects, AEC Symposium Series 20, US Atomic Energy Commission, Washington D.C. (1970) 63
8. Report of Committee II on Permissible Dose for Internal Radiation, Health Physics, 3 (1960) 9
9. Dillman, J.T., MIRD Pamphlet Nos. 4 & 6, J. Nucl. Medicine Supplements 2 (1969) and 4 (1970).

CALCULATION OF SURFACE DOSE AND INTERNAL ORGAN DOSE  
FOR HUMAN EXPOSURE TO WEAPON NEUTRONS

Robert A. Facey  
Defence Research Establishment Ottawa  
Ottawa, Canada.

Abstract

A method is described of calculating internal organ dose for humans by computer. Any type of neutron spectrum may be used for input. The method is illustrated by the use of a typical neutron energy angular distribution prepared for civil defense purposes.

Introduction

Dose relationships for fast neutrons are of particular importance in criticality accident dosimetry, and for so-called "disaster dosimetry". The latter includes casualty prediction for civil defense and military applications. After allowance for the large differences in the applicable quality factors, the same relationships also could be useful for fast neutron therapy.

A theoretical model would be concerned with dose to the internal organs of the body. This internal organ dose or doses then would be related by the model to dose on selected areas of the surface of the body where a surface dosimeter might be worn, and also to an external reference such as the midline tissue dose, free-in-air. Dose to the bone marrow system and to the intestines were of interest to this study.

Previous Work

A previous approach to the problem of dose prediction in the body was that of Mechali<sup>1, 2</sup>. His method was to create a theoretical, elliptical cylindrical phantom of homogeneous tissue, and to locate point representations of bone marrow at reasonable depths within it. He then applied the slab calculations of Handbook 63<sup>3</sup> to derive dose and dose-equivalent components at these points.

Mechali made the assumption that the chest dosimeter was on the most exposed region (even for bilaterally incident radiation), and that maximum internal absorbed dose or RBE-dose could be taken as the surface dosimeter reading. The problem of the most suitable location for a surface dosimeter for prediction of absorbed dose is of interest and it is worth noting that experimental phantom irradiations with monoenergetic sources of photons<sup>4</sup> or neutrons<sup>5</sup> have favoured locations on the torso lower than the chest. One aim is to relate internal organ dose in a theoretical model to the readings of various surface dosimeters, as well as to midline tissue dose, free-in-air.

Method

Input spectrum

For the first calculations, for a man exposed to neutrons from a nuclear weapon, the spectrum chosen was that recommended for shielding calculations for civil defense purposes for the Committee on Civil Defense of the U.S. National Academy of Sciences<sup>6</sup>. This spectrum, which will be referred to as the "civil defense spectrum" was for a typical thermonuclear weapon of intermediate yield at a slant range of 1200 m in infinite air. It is recognized that the thermal and blast environments could be quite unsupportable at such a location, but here we are concerned only with the method.

The civil defense spectrum used presents relative neutron fluences as a

function of angle of incidence and of neutron energy. The spectrum is normalized to unit incident dose, although in the original work of Straker and Gritzner<sup>7</sup>, the normalization was per source neutron.

The use of a spectrum, calculated in terms of incident dose seemed appropriate to our purposes. The civil defense spectrum<sup>6</sup> is specifically intended for use with shielding calculations for buildings. There is an analogy between the calculation of dose ratios inside and outside a blockhouse, and the calculation of similar ratios inside and outside the human body. A further advantage of the 1200 m spectrum is that the spectrum from a thermonuclear source varies only slowly with range for ranges greater than 600 m<sup>6</sup>.

The normalization of the results occurs automatically with this form of the spectrum. The use of the ORNL cylindrical phantom depth-dose data<sup>8</sup>, as described later, provides factors which are dimensionally the same as fluence-to-dose conversion factors. The application of these factors to the civil defense spectrum produces quantities that are thus automatically in units of dose per incident dose. This is numerically the same as ratios of internal dose to external free-in-air dose, continuing the analogy between the blockhouse and the human body. However, because for some internal locations the depth-dose calculations of Auxier, Snyder and Jones<sup>9</sup> for the cylindrical phantom lie above the fluence-to-dose conversion factors of Henderson recommended<sup>6</sup> for use with the civil defense spectrum, the analogy starts to break down. At some locations, the internal dose can apparently exceed the exposure by more than 40%, owing to this inconsistency in the two sets of calculations. If the fluence-to-kerma factors of Ritts et al.<sup>9</sup> are used this is reduced in the worst case to less than 30%.

#### Phantoms

The depth dose was determined in two steps, using two theoretical "phantoms". The first of these, called the DREO phantom, consisted of a series of horizontal cross-sectional drawings of the human body at a number of heights in an erect individual. Each drawing showed realistic locations of all bones, and effective centres were chosen for the bone marrow in each bone. The lungs were also included and an intestinal location was shown. Lines were drawn in the horizontal plane through each effective centre parallel to the directions of the incident radiation, and measurements made of the penetration distances from the irradiated surface. Six equally spaced horizontal directions were considered, but only 4 needed to be calculated. Penetration distances were corrected for passage through lung or bone. Two representative sections of the DREO phantom are shown in Figs. 1 and 2.

The second phantom was the ORNL circular cylindrical phantom of 30 cm diameter, used in the calculations of Auxier, Snyder and Jones<sup>6</sup>. The corrected penetration distances were transferred onto the ORNL phantom so that the various effective marrow centres now fell into various volume elements of the middle section (layer number three of Ref. 8) of the cylindrical mass of tissue. Because of the variable radius of curvature of the irradiated surface of a real body, the organs in the DREO phantom were classified as either midline-located (e.g. the vertebrae for frontal irradiation) or side-located (e.g. the ribs). The midline organs fell on the midline of the ORNL phantom, while the side-located organs were plotted at reasonable displacements towards the side of the ORNL phantom. The DREO phantom contained fixed organs, but was freely rotatable to any chosen angle with respect to the incident radiation. The ORNL phantom was rigidly fixed with respect to the direction of incident radiation, but it contained equivalent organs, whose positions formed a changing pattern whenever the DREO phantom changed its orientation in the radiation field.

#### Determination of depth dose

For each of the 20 volume elements of layer three of the ORNL phantom,

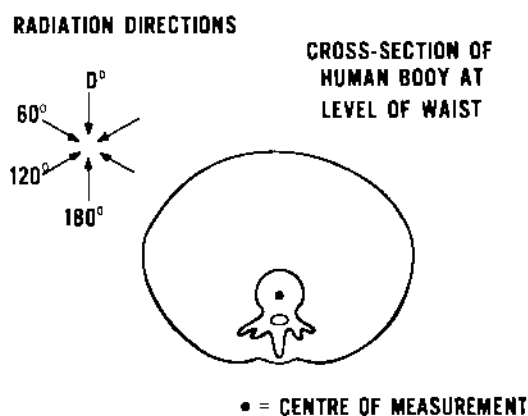


Fig. 1

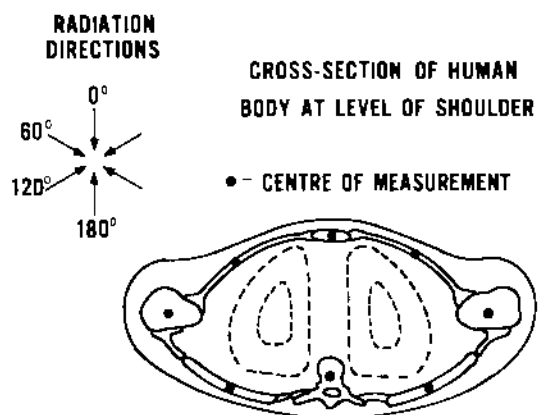


Fig. 2

and two supplementary surface elements, obtained by extrapolation, dose-versus-energy graphs were plotted using the data of Ref. 8. The dose-energy graphs were read off for each volume element at the mid-point of the 22 energy bins of the civil defense spectrum<sup>6</sup>. This provided the input data in units of  $\text{rad cm}^2 \text{neutron}^{-1}$  at 22 energies for 22 volume elements, to be used for the determination of the depth dose.

#### Angular constraint

Ultimately a useful theoretical model should be able to apply various angular spectra to a three-dimensional set of penetration distances to take maximum advantage of the large amount of angular information implicit in such spectra. It is already well demonstrated<sup>4,5</sup> that the case of normally incident radiation, is over-conservative and unrealistic, when it is applied to prediction of the dose to the bone marrow or to points in the intestinal tract for the cases of off-angle (elevated angle of incidence), or semi-isotropic incidence. This is due to the effects of body self-shielding for the organs in question.

In the present program there was a severe angular constraint on the three-dimensional spectral data. The fluences in each angle bin of the input spectrum were regrouped into six wide angle bins, and these six bins were then constrained to lie in the horizontal plane. Thus all the neutrons of the civil defense spectrum were presumed to be incident upon the phantom horizontally. This unavoidably overestimated the internal dose to our organs of interest. It must be emphasized that the concern of the present paper with normally incident radiation in no way implies any belief that this is an adequate approach to what is really a three-dimensional problem. Until we are able to complete our set of three-dimensional measurements, a two-dimensional approach has to suffice.

#### Program

After reading-in the spectrum and the depth dose data, and calculating their product, the program performed the summations over energy to produce an array of partial doses in each volume element of the ORNL phantom for each of our six angles of incidence. This was done by applying the six angular fluences, one at a time, to the front irradiated surface of the ORNL phantom. The rest of the program consisted of a system of keeping track of the internal organs or the surface dosimeters with respect to their constantly changing locations in the ORNL volume elements, and of summing up their accumulated doses with respect to angle. This produced an integrated dose for each location. In the case of the bone-marrow system, the calculated partial dose at each effective centre of a bone, was weighted by the local percentage of red marrow for that bone or group of bones according to the data of Ellis<sup>7,8</sup>. This gave an array of dose contributions for each of the 22 bones or bone groups, and this was then summed to give mean bone marrow dose.

On printout, a man could be considered as facing into the radiation from the weapon, or at angles of  $60^\circ$ ,  $120^\circ$  or  $180^\circ$  from it. Tables are printed of the dose contributions at all the sites due to the fluence reaching him from one of the  $60^\circ$  angle bins in the horizontal plane. This procedure is repeated for the six angles. At each of the six orientations, dose due to total fluence as well as the partial angular fluences is obtained.

A variant of the program was written to perform calculations for monoenergetic, monodirectional neutrons at the mid-points of the upper 13 energy groups used previously. This gave a number of irregularly-spaced energies lying between 330 keV and 13.6 MeV. The program and the types of output obtained were similar to that just described.

### Results

Because of uncertainties due to the imposed angular constraints, it would be unwarranted to attach much significance to the absolute values predicted. However, the relative dose values are less sensitive to error. Figs. 3 to 6 are histograms showing relative dose contributions, in the case of a man facing the weapon, for neutrons arriving from various angles. Fig. 3 shows mean bone marrow dose. Fig. 4 shows intestinal dose at a representative location. As one would expect the intestines are more heavily dosed than the bone marrow, and their dose is much more dependent upon the fraction of the neutrons arriving from the direction of the source. Fig. 5 shows the dose to a front surface dosimeter, located on the midline at the waist. This is even more directionally dependent when the man is facing the source. Fig. 6 shows dose to a similar waist dosimeter worn at the back. In this case, the largest dose contribution is received from neutrons entering the bins at  $120^\circ$  and  $240^\circ$ . In the case of a man facing  $180^\circ$  away from the weapon, Figs. 5 and 6 would simply be reversed for the two waist dosimeters.

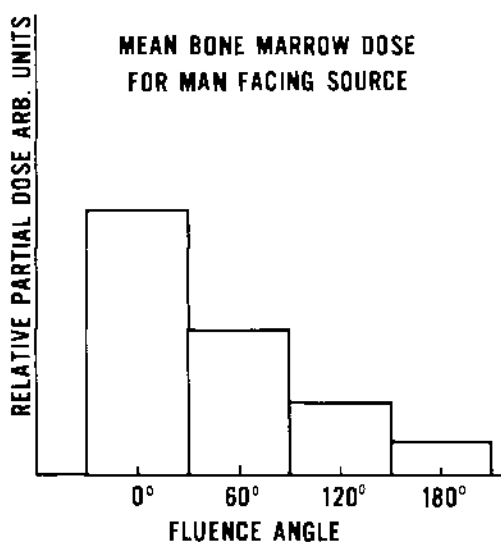


Fig. 3

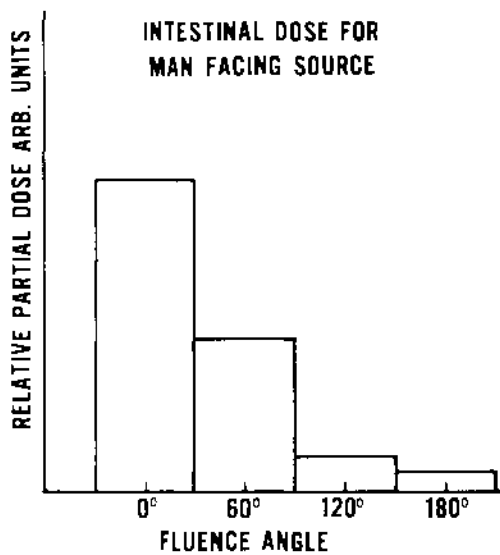


Fig. 4

Fig. 7 shows a table of dose ratios for a man standing in four different orientations in the field, from face-on to the source ( $0^\circ$ ) to back-on ( $180^\circ$ ). The top line shows intestinal dose over the reading of a front waist dosimeter. The ratio is about 0.5 to 0.7, and is not very sensitive to orientation. The second line shows the ratios for intestinal dose over a back waist dosimeter. The body shielding of the dosimeter in this location raises the  $0^\circ$  ratio to 0.8, and the directional dependence now changes the ratio by a factor of about 2 as the man turns to the back-on position. The bottom line shows the ratio of the intestinal dose to the free-in-air tissue dose. There is little directionality,



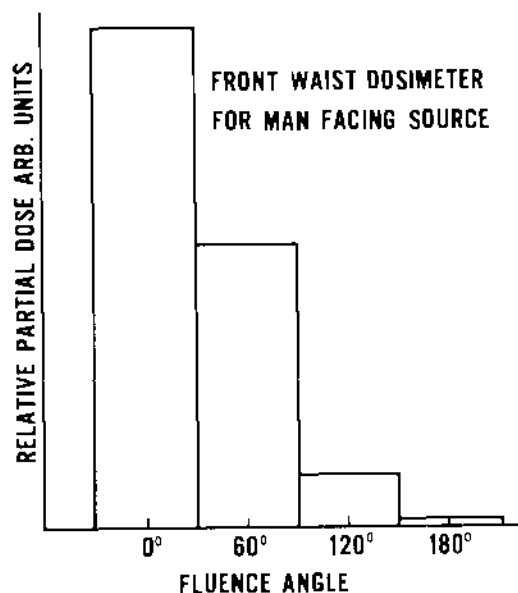


Fig. 5

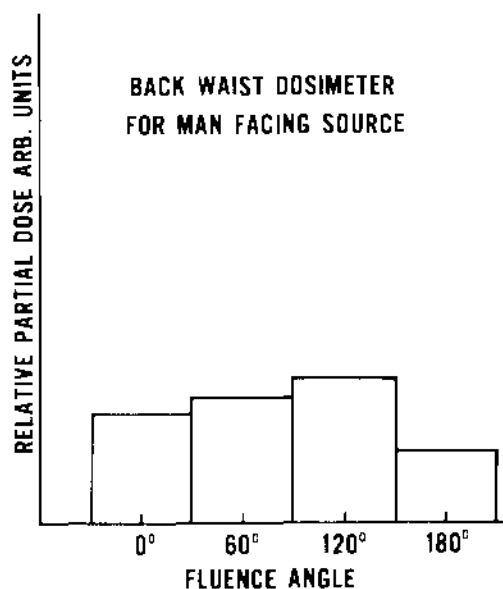


Fig. 6

but the ratio is around 0.45. This supports the view<sup>5</sup> that the free-in-air dose, unless weighted by ratios like this, is a poor indicator of dose to deeply located organs such as the intestines, or the pelvic bone marrow.

Fig. 8 illustrates the results of the monoenergetic calculations for a human exposed face-on to the source. The ordinate gives total mean bone marrow dose (recoil ion plus gamma components), and the abscissa gives neutron energy. The present work is higher in dose than the calculation of Mechali<sup>1</sup>, particularly at 3.5 MeV, where the difference is around 25%. When the monoenergetic calculations at 2.74 MeV were compared with the DREO experimental human phantom measurements at 2.95 MeV<sup>5</sup> the results agreed within 20% for the intestinal location and for the front surface-waist for both face-on and back-on orientations.

		ORIENTATION OF MAN IN FIELD ( DEGREES OFF FACE-ON )			
		0°	60°	120°	180°
SURFACE WAIST	FRONT	.53	.55	.60	.66
	BACK	.81	.67	.49	.43
FREE-IN-AIR		.50	.49	.43	.40

Fig. 7

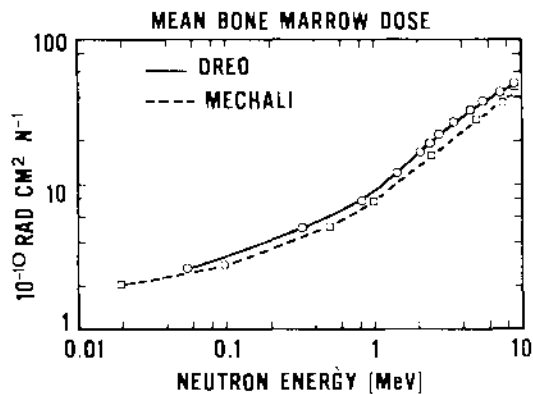


Fig. 8

### Conclusions

This method can provide a rapid and versatile approach to problems of dose prediction. The approach is particularly powerful in the amount of angular information that can be made available, both with respect to neutron angle and to angle of orientation of the man. A complete breakdown of all bone-marrow components is also always available. The demands on computer time and memory are insignificant.

It appears that the calculations support the view that a dosimeter

located on the surface of the body facing the unscattered radiation in a semi-isotropic field overestimates the dose to deep internal organs by a factor of around 2, while a detector on the far side may not. This is in line with the view previously expressed<sup>4, 5</sup> that a partially shielded dosimeter location such as the groin may be an improved location for a detector for disaster or criticality dosimetry.

It is felt that future work should involve a method to handle the three-dimensional case, without any angular constraint on the incident fluence.

#### Acknowledgements

This program was devised and written by Mr. D.A. Collins, to whom I express my sincere gratitude. I also wish to thank Dr. C.E. Clifford and Dr. P.A. Tate (of the Department of National Defence) for much help and direction, and Mr. R.A. Gravelle for assistance.

#### References

1. D. MECHALI, et al., Personnel Dosimetry for Radiation Accidents (p. 483), published by I.A.E.A., Vienna (1965).
2. D. MECHALI, Stockholm Symposium on Radiation Dose Measurements (p. 227), published by E.N.E.A., Paris (1967).
3. NCRP (1957). National Bureau of Standards (US), Handbook 63.
4. C.E. CLIFFORD and R.A. FACEY, Health Phys. 18, 217 (1970).
5. R.A. FACEY and C.E. CLIFFORD, Angular Dependence of the Components of Dose to the Bone Marrow and Abdomen in a Human Phantom from 2.95-MeV Neutrons. To be published in Health Phys. (1973).
6. J.A. AUXIER et al., Nuclear Weapons free-field Environment Recommended for Initial Radiation Shielding Calculations, ORNL-TM-3396 (1972).
7. E.A. STRAKER and M.L. GRITZNER, Neutron and Secondary Gamma-Ray Transport In Infinite Homogeneous Air, ORNL-4464 (Dec. 1969).
8. J.A. AUXIER, W.S. SNYDER and T.D. JONES, Radiation Dosimetry (Edited by F.H. ATTIX and W.C. ROESCH), Ch. 6. Academic Press, New York (1968).
9. J.J. RITTS, E. SOLOMITO and P.N. STEVENS, Calculations of Neutron Fluence-to-Kerma Factors for the Human Body, ORNL-TM-2079 (1968).
10. R.E. ELLIS, Phys. Med. Biol. 5, 255 (1961).

DOSE DUE TO PRACTICAL NEUTRON ENERGY DISTRIBUTIONS  
INCIDENT ON CONCRETE SHIELDING WALLS

by

A. M. Serkoff  
National Bureau of Standards, Washington, D.C. 20234

and

A. H. Chilton  
Nuclear Engineering Laboratory  
University of Illinois, Urbana-Champaign, Illinois 61801

Abstract

In order to calculate the dose equivalent for persons on the far side of an ordinary concrete shielding wall from a plane parallel source of neutrons, published monoenergetic neutron data and practical neutron spectra have been folded together. Some interpolation of the monoenergetic neutron data, both in energy and slab thickness, was required to prepare the data for computation in steps of 50  $\mu\text{cm}^2$ . The result is a set of useful dose-equivalent data for walls as thick as 800  $\text{g/cm}^2$  used to shield particle accelerators producing neutrons up to 100 MeV in energy. A simple, empirically derived, analytical expression for the dose equivalent for walls of 50 to 800  $\text{g/cm}^2$  thickness is given with suitable constants for neutron spectra produced by more than 60 selected examples of the following reactions: slow and fast neutron induced fission,  $(\gamma, n)$ ,  $(p, n)$ ,  $(^3\text{He}, n)$ ,  $(\alpha, n)$  and fission  $(\gamma, n)$ .

Introduction

There is a famous recipe for rabbit stew that begins, "First you catch a rabbit." Many chefs never read beyond that first line. In a similar fashion, this brief paper must begin, "First you measure (or calculate) the total number of neutrons incident on each unit of area of an ordinary concrete shielding wall surrounding a neutron source, such as an accelerator." The material presented here allows a calculation of the tissue dose-equivalent, due to both neutrons and gamma rays induced in the shield at the far side of a wall of thickness  $x$  (in  $\text{g/cm}^2$ ).

This calculation was undertaken to provide data in a readily usable form for shielding designers. The need for such information became evident in the preparation of design guidelines for accelerator facilities by Scientific Committee #22 of the National Council on Radiation Protection and Measurement, and the results will be used in that document. Multi-collision dose-equivalent due to monoenergetic plane-parallel neutrons incident on ordinary concrete walls has been reported by Roussin and Schmidt<sup>(1)</sup> (herein after called R&S), R.G. Alsmiller et al<sup>(2)</sup> (herein after called AMBE) and by Roussin, Alsmiller and Bartish<sup>(3)</sup> (herein after called RAB) for neutron energies from thermal up to 400 MeV. Also, a large number of workers have measured neutron energy spectra and reported their results. A good-sized sample of these spectra, representative of thick target conditions, in the vicinity of 1 to 100 MeV particle accelerators have been taken from the literature. This report gives the results of a straight forward folding of some of these two types of data together.

There are several factors, not discussed in this report, which also influence the work of a neutron shielding designer. For the variation of energy spectra and neutron flux with angle, we refer the reader to many of the articles given as references for the neutron energy spectra. A recent study by one of the authors<sup>(4)</sup> (ABC) has shown the importance of water content and other constituents on shielding for neutrons up to 15 MeV in energy. In this report, only a single silicious type of concrete is considered with a density of 2.31  $\text{g/cm}^3$  and composition as used by R&S and by RAB in units of  $10^{21}$  atoms per cubic centimeter as follows: H(8.5), C(20.50), O(35.50), Mg(1.86), Al(0.60), Si(1.70), Ca(11.30) and Fe(0.19). This concrete has 5.5% water, by weight. The location of any type of ducts or irregularities in the wall has a strong bearing on neutrons shielding characteristics and has been discussed in detail elsewhere<sup>(5)</sup>.

It should be noted that dose-equivalent is a defined quantity without direct physical significance. Neutron energy and gamma-ray energy are physical quantities, but the quality factor which relates these quantities to dose equivalent is the result of biological investigations. The set of fluence-to-dose-equivalent conversion factors used by R&S, AMBE and RAB is given in reference 1 and are consistent with the information given in NCRP Report No. 38<sup>(6)</sup>.

This report is organized in 4 sections. The first discusses the neutron energy spectra selected from the literature for the calculations. The second presents the monoenergetic neutron dose equivalent data derived from R&S, AMBE and RAB. The third section discusses the dose equivalent calculations including an empirical analytical fit to the resulting dose equivalent data. The fourth and final section presents a few comments on the results obtained.

I. The Neutron Energy Spectra

To the extent that beam particle type, incident particle energy, target material and target geometry vary, the data taken from the literature can only be considered to be a fair sample of the neutron spectra to be expected under other circumstances. The spectra used are listed in Table 1 where part (a) includes 3 fission cases<sup>(7)</sup> and 3  $(\alpha, n)$  cases<sup>(8)</sup>; part (b) includes 35  $(n, n)$  spectra and a single  $(^3\text{He}, n)$  case and part (c) includes 20 bremsstrahlung induced  $(\gamma, n)$  spectra for a total of 62 spectra. In some cases neutron energy spectra are integrated over all angles, but most of these data are for neutrons leaving a target in a restricted angular region. The constants A, B, and C listed in these tables relate to the empirical dose equivalent expression to be discussed in section III. As the format and scale of the original

Table 1  
Neutron Sources and Related Dose Equivalent Constants

Table 1a No.	Incident Particle	Particle Source	Particle Target	A	B	C	Ref. No.
1	$\alpha$	Ra	Be	.1611	.3697	.4318	7
2	$\alpha$	Po	Be	.1618	.3624	.3458	7
3	$\alpha$	Pu	Be	.1620	.3593	.3025	7
4	n (slow)	fission	$^{235}\text{U}$	.1658	.3857	.7290	7
5	n th	fission	$^{235}\text{U}$	.1629	.3824	.6543	8
6	n fast	fission	$^{235}\text{U}$	.1632	.3506	.1990	8

Table 1b No.	Proton Energy	Proton Target	n angle to beam	A	B	C	Ref. No.
1	8	C	0	.1615	.4096	1.031	8
2	9	C	0	.1609	.3686	.3913	8
3	11	C	0	.1610	.3956	.8178	8
4	12	C	0	.1609	.3926	.7700	8
5	13	C	0	.1614	.3910	.7502	8
6	11	C	90	.1606	.4172	1.181	8
7	12	C	90	.1628	.4087	1.057	8
8	11	$\text{H}_2\text{O}$	90	.1668	.4252	1.378	8
9	12	$\text{H}_2\text{O}$	90	.1667	.4052	1.036	8
10	13	$\text{H}_2\text{O}$	90	.1661	.4078	1.074	8
11	10	Mg	0	.1653	.4234	1.333	8
12	11	Mg	0	.1643	.4140	1.161	8
13	12	Mg	0	.1640	.4090	1.074	8
14	14.0	Al	10	.1631	.3441	.1323	9
15	15.7	Al	10	.1620	.3396	.06672	9
16	16.3	Al	10	.1621	.3401	.07353	9
17	16.5	Al	10	.1625	.3400	.07496	9
18	18.0	Al	10	.1620	.3392	.06556	9
19	18.5	Al	10	.1632	.3415	.09840	9
20	20	Al	10	.1624	.3397	.07029	9
21	10	Si	0	.1623	.4265	1.371	8
22	11	Si	0	.1632	.4140	1.140	8
23	12	Si	0	.1635	.4120	1.111	8
24	10	S	0	.1664	.4256	1.391	8
25	11	S	0	.1696	.4256	1.439	8
26	12	S	0	.1680	.4243	1.389	8
27	8	Ta	0	.1708	.4285	1.501	8
28	9	Ta	0	.1706	.4270	1.468	8
29	10	Ta	0	.1704	.4253	1.433	8
30	11	Ta	0	.1711	.4211	1.367	8
31	12	Ta	0	.1705	.4207	1.348	8
32	13	Ta	0	.1701	.4206	1.343	8
33	11	Ta	90	.1716	.4250	1.443	8
34	12	Ta	90	.1702	.4253	1.434	8
35	13	Ta	90	.1708	.4224	1.383	8

Table 1b No.	$^3\text{He}$ Energy	$^3\text{He}$ Target	n angle to beam	A	B	C	Ref. No.
36	18	Cu		.1630	.3780	.5881	14

Table 1c No.	Brems. Energy	Brems. Target	angle to beam	A	B	C	Ref. No.
1	16	Pt	90	.1722	.4222	1.397	10
2	16	Pb	90	.1660	.4184	1.256	10
3	13	Bi	90	.1680	.4212	1.325	10
4	16	U	90	.1704	.4090	1.141	10
5	34	$^{16}\text{O}$	55	.1619	.3428	.09096	11
6	34	$^{16}\text{O}$	93	.1615	.3462	.1279	11
7	34	$^{16}\text{O}$	141	.1622	.3488	.1669	11
8	45	$^{238}\text{U}$	90	.1646	.3893	.7646	8
9	55	Be	67.5	.1575	.2374	.2266	12
10	55	Pb	67.5	.1585	.2697	1.207	12
11	65	$^{12}\text{C}$	90	.1601	.2979	.2126	13
12	65	$^{16}\text{O}$	90	.1600	.3145	.2923	13
13	65	$^{24}\text{Mg}$	90	.1606	.2866	.1543	13
14	65	$^{27}\text{Al}$	90	.1608	.2845	.1446	13
15	65	$^{28}\text{Si}$	90	.1596	.2942	.1717	13
16	65	$^{40}\text{Ca}$	90	.1609	.2904	.1864	13
17	65	$^{59}\text{Co}$	90	.1595	.3217	.2675	13
18	65	$^{181}\text{Ta}$	90	.1607	.3185	.3681	13
19	85	Be	67.5	.1601	.1941	.4703	12
20	85	Pb	67.5	.1582	.2506	.4734	12

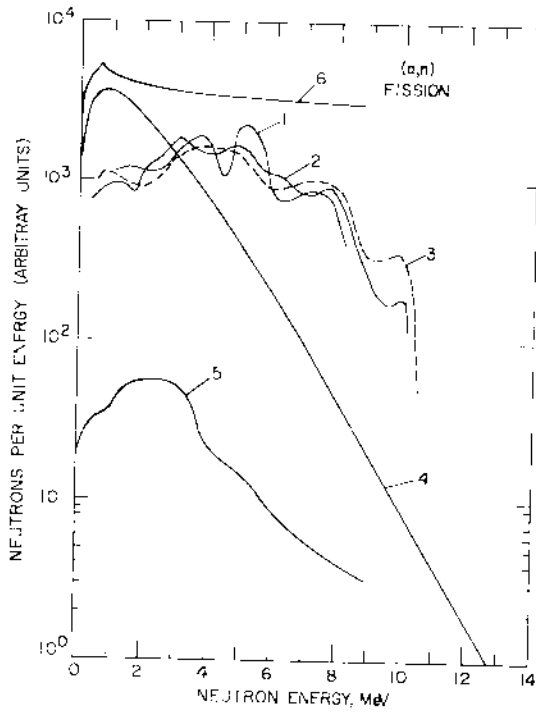


Figure 1

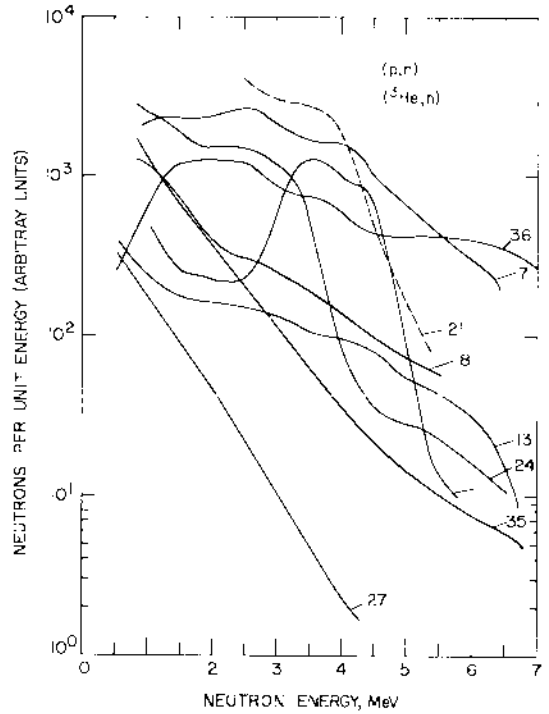


Figure 2

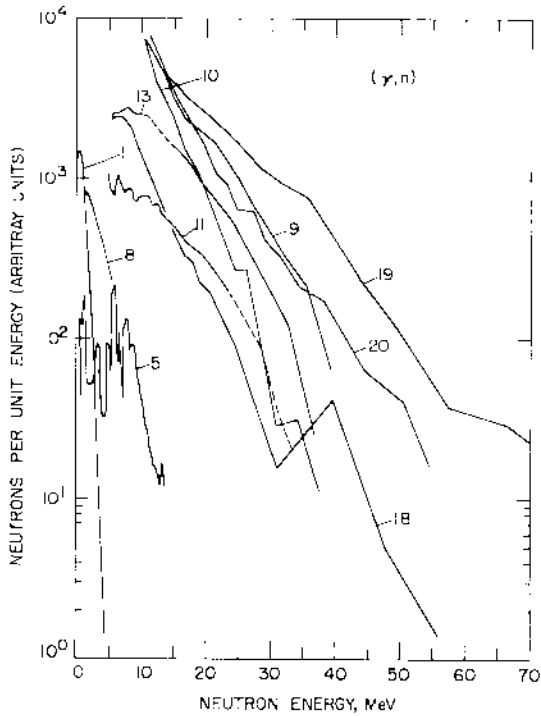


Figure 3

CAPTIONS

1. Neutron spectra from beryllium bombarded by alpha particles in (1) Ra-Be (2) Po-Be and (3) Pu-Be sources, and fission spectra from a (4)  $^{235}\text{U}$ , (5) a thermal reactor core and (6) a faster reactor core. Note that the faster reactor data is incomplete on the high energy side.
2. Neutron spectra produced by proton bombardment of a number of targets obtained at angles to the beam indicated in parenthesis: 1. 8 MeV C(0°), 7. 12 MeV C(90°), 8. 11 MeV C<sub>2</sub>O(90°), 13. 12 MeV Mg(0°), 19. 18.5 MeV Al((10°), 21. 10 MeV Si(0°), 24. 10 MeV S(0°), 27. 8 MeV Ta(0°), and 35. 13 MeV Ta(90°). The neutron spectrum produced by 18 MeV  $^3\text{He}$  incident on Cu 11 is plotted in curve 36.
3. Neutron spectra produced by bremsstrahlung bombardment of the targets indicated with neutrons emerging at the angles indicated in parenthesis: 1. 16 MeV Pt(90°), 5. 34 MeV  $^{16}\text{O}$ (55°), 8. 45 MeV  $^{238}\text{U}$ (90°), 9. 55 MeV Be(67.5°), 10. 55 MeV Pb(67.5°), 11. 65 MeV  $^{12}\text{C}$ (90°), 13. 65 MeV  $^{24}\text{Mg}$ (90°), 19. 85 MeV Be(67.5°), and 20. 85 MeV Pb(67.5°).

drawings varies widely, selected samples of these input data have been replotted in Figures 1, 2, and 3 on semi-log graphs so that shape differences and limitations on some of the data can be appreciated. The  $\alpha$  source and fission spectra are included for completeness though of limited use in accelerator shielding situations.

## II. The Monoenergetic Neutron Dose Equivalent Data

The data in Table 2 is the tissue dose equivalent per neutron due to both neutrons and the  $\gamma$ -rays produced in the shielding concrete for each of energy bins indicated. The original data used to prepare this table came from the three sources already mentioned<sup>(1-3)</sup>. Each had to be adapted to this application.

RAS<sup>(1)</sup> covered concrete-slab penetration by neutrons in 22 energy groups covering the energy range from thermal to 15 MeV and slab thicknesses to 642 g/cm<sup>2</sup>. Similar data reported by one of the authors (ABC)<sup>(2)</sup> allowed extrapolation of this data to thicknesses of 800 g/cm<sup>2</sup>. Interpolation in slab thickness was used to tabulate these data in steps of 50 g/cm<sup>2</sup>.

RAB<sup>(3)</sup> extended the energy range by making calculations at 25, 50, and 75 MeV for slabs as thick as 1500 g/cm<sup>2</sup>. AMPE<sup>(4)</sup> reported calculation for SiO<sub>2</sub> and 5% H<sub>2</sub>O at 100 MeV. Since neutrons interact, at such high energies, largely with individual nucleons, calculations for such a simplified material may fairly be used for a concrete shield calculation. RAS confirmed this by means of a calculation at 50 MeV which showed little difference between a more realistic concrete composition and the SiO<sub>2</sub>-H<sub>2</sub>O model. The results of these two calculations of dose equivalent inside a shield were used to obtain dose-equivalent on the far side of a shielding wall. A correction to account for the drop in dose equivalent near the exit surface due to a lack of backscattering had to be made. This correction process is not precise so some uncertainty is introduced in the results by this step. Since the energies used in the dose equivalent matrix of this report are at 20, 32.5, 50, 70, and 100 MeV it was necessary to interpolate, not only with respect to thickness as was done in the case of the RAS data, but also with respect to incident energy.

Table 2 may be employed with any plane parallel incident neutron energy spectra to obtain the tissue dose equivalent on the far side of a concrete shielding wall.

## III. Computation of the Dose Equivalent

The matrix multiplication used to fold the neutron spectra  $f(E)$  into the dose equivalent matrix  $R(Z, E)$  is

$$D(Z) = \int_{E_1}^{E_2} f(E_1) R(Z, E_1) \Delta E_1$$

The results obtained for about one-third of the spectra are plotted in Figure 4 for values of  $Z$  ranging from 0 to 800 g/cm<sup>2</sup>. Dose in this case is in units rem·cm<sup>2</sup>·n<sup>-1</sup>.

The dose equivalent when plotted on a semi-log plot tends toward a straight line at greater depth which suggested the possibility of an empirical fitting of the data by an equation of the form

$$\ln D(Z) = -A \cdot 10^2 - B \cdot 10^{-1} \frac{Z}{Z_0} + C \cdot 10^{-5} \frac{Z}{Z_0}^2$$

where  $Z_0 = 1 \text{ g/cm}^2$ .

Using this expression as a basis function, a polynomial least squares fit was made for the  $D(Z)$  data for each calculated case. It was found that a reasonable  $\chi^2$  could be obtained if the first and last data points were dropped so, for this case, the values are listed in table 1a, b, and c. Where values of  $D(Z)$  calculated by this approximation are compared to the original data differences of up to 18% are observed but the mean difference is considerably less. Since  $D(Z)$  varies by more than 10 decades for the walls considered, this fit, which corresponds to an uncertainty of about 10 g/cm<sup>2</sup>, is impressively good in the region above 50 g/cm<sup>2</sup>. Below 50 g/cm<sup>2</sup> errors up to a factor of 3 are possible.

Examination of the basis function makes it clear that the first term establishes an incident plane intercept, the second term determines the slope and the third term applies a small correction to the slope.

## IV. Comments on the Results

The first generalization about the dose equivalent as a function of shielding wall thickness is that for three cases, neutrons produced by fission, by  $\text{Be}(n, n)$  processes and by  $(p, n)$  processes shielding thickness required is very similar. Variations of 25 g/cm<sup>2</sup> separate the extremes for  $(\alpha, n)$  and fission processes. Variations of about 60 g/cm<sup>2</sup> separate the extremes for  $(p, n)$  processes. In sharp contrast to this,  $(\gamma, n)$  processes require shielding that is very much a function of energy. For example, a shielding wall adequate for a 16 MeV bremsstrahlung platinum target must be doubled in thickness for an 85 MeV bremsstrahlung lead target producing an equal flux of neutrons at the entrance plane. The dose equivalent calculated for the neutrons produced by bremsstrahlung striking <sup>238</sup>U are an understandable exception due to the admixture of low energy fission neutrons produced.

Small changes in the energy of protons incident on the target material, and in the angle of exit of neutrons leaving the target material, seem to have only minimal effect on the shielding requirements.

There is a lack of data (known to these authors) on experimental determination of dose equivalent for cases involving the inevitable mixture of neutrons and x-rays leaving a thick concrete shield when higher energy neutrons are incident.

## Acknowledgement

The authors are indebted to Gerard Cavanaugh and Theodore Kraft who did much of the data processing for this study. We also appreciate the encouragement from E. Alfred Burrill who showed a keen interest in these calculations as they developed.

TABLE 2

Increment No.	Energy		Monenergetic Neutron Dose Equivalent								
	Interval (eV)		Z=0	Z=50	Z=100	Z=150	Z=200	Z=250	Z=300	Z=350	Z=400
1	0.	4.14E-07	0.10E-08	0.18E-09	0.30E-10	0.51E-11	0.99E-12	0.21E-12	0.46E-13	0.11E-13	0.30E-14
2	4.14E-07	1.12E-06	0.17E-08	0.31E-09	0.48E-10	0.81E-11	0.16E-11	0.33E-12	0.74E-13	0.17E-13	0.42E-14
3	1.12E-06	3.06E-06	0.12E-08	0.35E-09	0.55E-10	0.93E-11	0.18E-11	0.36E-12	0.64E-13	0.20E-13	0.48E-14
4	3.06E-06	1.07E-05	0.12E-08	0.35E-09	0.62E-10	0.11E-10	0.20E-11	0.40E-12	0.92E-13	0.21E-13	0.52E-14
5	1.07E-05	2.90E-05	0.13E-08	0.40E-09	0.66E-10	0.12E-10	0.21E-11	0.43E-12	0.98E-13	0.23E-13	0.55E-14
6	2.90E-05	1.01E-04	0.13E-08	0.42E-09	0.73E-10	0.12E-10	0.23E-11	0.44E-12	0.99E-13	0.24E-13	0.58E-14
7	1.01E-04	5.83E-04	0.13E-08	0.43E-09	0.77E-10	0.13E-10	0.24E-11	0.46E-12	0.10E-12	0.25E-13	0.60E-14
8	5.83E-04	3.35E-03	0.12E-08	0.44E-09	0.81E-10	0.14E-10	0.25E-11	0.50E-12	0.11E-12	0.26E-13	0.63E-14
9	3.35E-03	1.11E-01	0.35E-08	0.44E-09	0.80E-10	0.14E-10	0.25E-11	0.50E-12	0.11E-12	0.25E-13	0.61E-14
10	1.11E-01	5.50E-01	0.16E-07	0.70E-09	0.97E-10	0.17E-10	0.30E-11	0.57E-12	0.13E-12	0.28E-13	0.68E-14
11	5.50E-01	1.11	0.31E-07	0.22E-08	0.19E-09	0.29E-10	0.49E-11	0.95E-12	0.20E-12	0.43E-13	0.92E-14
12	1.11	1.83	0.37E-07	0.64E-08	0.49E-09	0.59E-10	0.91E-11	0.17E-11	0.35E-12	0.74E-13	0.18E-13
13	1.83	2.35	0.36E-07	0.12E-07	0.12E-08	0.15E-09	0.19E-10	0.32E-11	0.49E-12	0.73E-13	0.29E-13
14	2.35	2.46	0.35E-07	0.16E-07	0.31E-08	0.46E-09	0.69E-10	0.10E-10	0.17E-11	0.31E-12	0.65E-13
15	2.46	3.01	0.35E-07	0.14E-07	0.23E-08	0.32E-09	0.43E-10	0.63E-11	0.11E-11	0.21E-12	0.44E-13
16	3.01	4.07	0.36E-07	0.13E-07	0.18E-08	0.24E-09	0.33E-10	0.50E-11	0.83E-12	0.16E-12	0.34E-13
17	4.07	4.97	0.37E-07	0.17E-07	0.32E-08	0.50E-09	0.73E-10	0.12E-10	0.18E-11	0.31E-12	0.61E-13
18	4.97	6.36	0.39E-07	0.17E-07	0.40E-08	0.78E-09	0.14E-09	0.23E-10	0.36E-11	0.63E-12	0.12E-12
19	6.36	8.19	0.41E-07	0.17E-07	0.42E-08	0.84E-09	0.16E-09	0.29E-10	0.53E-11	0.89E-12	0.17E-12
20	8.19	10.0	0.42E-07	0.18E-07	0.43E-08	0.89E-09	0.17E-09	0.32E-10	0.59E-11	0.11E-11	0.20E-12
21	10.0	12.2	0.46E-07	0.18E-07	0.43E-08	0.85E-09	0.16E-09	0.28E-10	0.50E-11	0.87E-12	0.17E-12
22	12.2	15.0	0.56E-07	0.21E-07	0.50E-08	0.11E-08	0.20E-09	0.39E-10	0.70E-11	0.13E-11	0.24E-12
23	15.0	25.0	0.67E-07	0.57E-07	0.19E-07	0.56E-08	0.14E-08	0.30E-09	0.60E-10	0.13E-10	0.31E-11
24	25.0	40.0	0.70E-07	0.62E-07	0.31E-07	0.13E-07	0.46E-08	0.14E-08	0.48E-09	0.15E-09	0.42E-10
25	40.0	60.0	0.67E-07	0.62E-07	0.37E-07	0.20E-07	0.10E-07	0.45E-08	0.19E-08	0.82E-09	0.34E-09
26	60.0	80.0	0.59E-07	0.64E-07	0.44E-07	0.25E-07	0.16E-07	0.82E-08	0.43E-08	0.23E-08	0.12E-08
27	80.0	120.0	0.50E-07	0.60E-07	0.46E-07	0.33E-07	0.22E-07	0.14E-07	0.85E-08	0.52E-08	0.31E-08

			Z=450	Z=500	Z=550	Z=600	Z=650	Z=700	Z=750	Z=800
1	0.	4.14E-07	0.80E-15	0.23E-15	0.65E-16	0.18E-16	0.	0.	0.	0.
2	4.14E-07	1.12E-06	0.11E-14	0.26E-15	0.67E-16	0.17E-16	0.	0.	0.	0.
3	1.12E-06	3.06E-06	0.12E-14	0.30E-15	0.76E-16	0.19E-16	0.	0.	0.	0.
4	3.06E-06	1.07E-05	0.23E-14	0.32E-15	0.80E-16	0.20E-16	0.	0.	0.	0.
5	1.07E-05	2.90E-05	0.14E-14	0.37E-15	0.84E-16	0.21E-16	0.	0.	0.	0.
6	2.90E-05	1.01E-04	0.15E-14	0.36E-15	0.88E-16	0.21E-16	0.	0.	0.	0.
7	1.01E-04	5.83E-04	0.15E-14	0.37E-15	0.91E-16	0.22E-16	0.	0.	0.	0.
8	5.83E-04	3.35E-03	0.16E-14	0.38E-15	0.94E-16	0.23E-16	0.	0.	0.	0.
9	3.35E-03	1.11E-01	0.15E-14	0.36E-15	0.85E-16	0.20E-16	0.	0.	0.	0.
10	1.11E-01	5.50E-01	0.17E-14	0.40E-15	0.97E-16	0.23E-16	0.	0.	0.	0.
11	5.50E-01	1.11	0.20E-14	0.43E-15	0.94E-16	0.21E-16	0.	0.	0.	0.
12	1.11	1.83	0.42E-14	0.98E-15	0.24E-15	0.53E-16	0.14E-16	0.	0.	0.
13	1.83	2.35	0.67E-14	0.16E-14	0.36E-15	0.83E-16	0.19E-16	0.	0.	0.
14	2.35	2.46	0.14E-13	0.29E-14	0.60E-15	0.13E-15	0.28E-16	0.	0.	0.
15	2.46	3.01	0.97E-14	0.21E-14	0.46E-15	0.10E-15	0.22E-16	0.	0.	0.
16	3.01	4.07	0.25E-14	0.17E-14	0.36E-15	0.80E-16	0.18E-16	0.	0.	0.
17	4.07	4.97	0.12E-13	0.25E-14	0.50E-15	0.10E-15	0.19E-16	0.	0.	0.
18	4.97	6.36	0.22E-13	0.40E-14	0.74E-15	0.14E-15	0.26E-16	0.	0.	0.
19	6.36	8.19	0.31E-13	0.57E-14	0.11E-14	0.20E-15	0.38E-16	0.	0.	0.
20	8.19	10.0	0.37E-13	0.70E-14	0.13E-14	0.25E-15	0.48E-16	0.	0.	0.
21	10.0	12.2	0.32E-13	0.59E-14	0.12E-14	0.22E-15	0.43E-16	0.	0.	0.
22	12.2	15.0	0.46E-13	0.87E-14	0.17E-14	0.32E-15	0.61E-16	0.12E-16	0.	0.
23	15.0	25.0	0.58E-12	0.14E-12	0.28E-13	0.57E-14	0.13E-14	0.30E-15	0.67E-16	0.16E-16
24	25.0	40.0	0.12E-10	0.39E-11	0.11E-11	0.28E-12	0.75E-13	0.22E-13	0.48E-14	0.10E-14
25	40.0	60.0	0.14E-09	0.59E-10	0.24E-10	0.95E-11	0.37E-11	0.15E-11	0.53E-12	0.20E-12
26	60.0	80.0	0.60E-09	0.29E-09	0.15E-09	0.62E-10	0.36E-10	0.20E-10	0.92E-11	0.46E-11
27	80.0	120.0	0.19E-08	0.11E-08	0.64E-09	0.36E-09	0.21E-09	0.12E-09	0.65E-10	0.35E-10

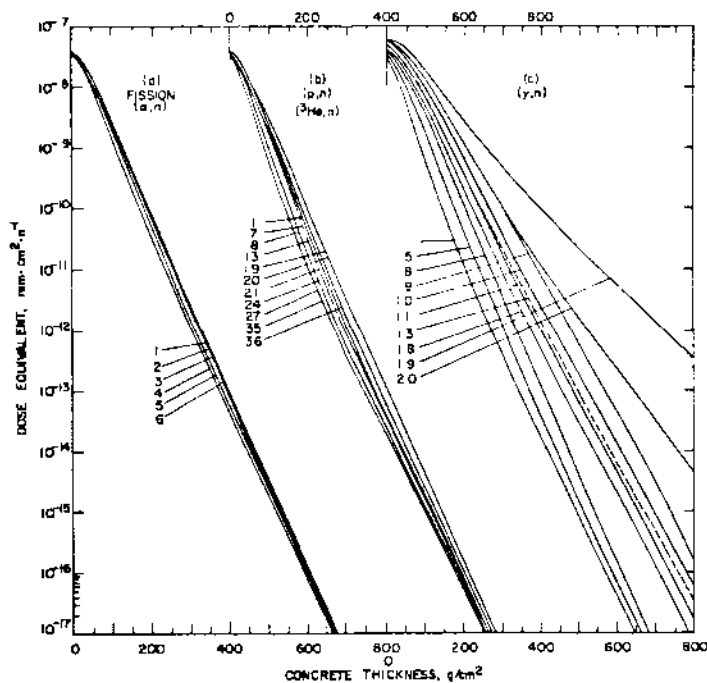


Figure 4. Tissue dose-equivalent at the far side of a concrete shielding wall for plane parallel incident neutron spectra. Fission process and  $\alpha$ -beryllium sources indicated by number are listed in Table 1a and shown in (a).  $(p,n)$  and  $(^3\text{He},n)$  data are shown in (b) and  $(\gamma,n)$  data is shown in (c). The concrete thickness axis has been shifted by 400 g/cm<sup>2</sup> for (b) and (c) in order to avoid confusion of closely spaced data.

#### REFERENCES

1. R.W. Roussin and F.A.R. Schmidt, "Adjoint  $S_n$  Calculations of Coupled Neutron and Gamma-Ray Transport Through Concrete Slabs," Nucl. Eng. and Des. **15**, 3, 319 (1971).
2. R.G. Alsmiller, Jr., F.R. Myatt, J. Barish and W.W. Engle, Jr., "Shielding Against Neutrons in the Energy Range 50 to 400 MeV," Nucl. Instr. and Meth. **72** 213 (1969) (Also published as OROU-TM-2554).
3. R.W. Roussin, R.G. Alsmiller, Jr. and J. Barish, "Calculations of the Transport of Neutrons and Secondary Gamma-Rays Through Concrete for Incident neutrons in the Energy Range 15 to 75 MeV," Nucl. Eng. and Des. **24**, 2 (1973).
4. Private Communication, A.S. Chilton, "Effect of Material Composition on Neutron Penetration of Concrete slabs," Nat. Bur. Stds. Rep. 10425, June 1971 (unpublished).
5. R.G. Jaeger, Editor, Engineering Compendium on Radiation Shielding Vol. 1, Shielding Fundamentals and Methods (Chapter 8) Springer-Verlag, New York (1970).
6. National Committee on Radiation Protection and Measurement, "Protection Against Neutron Radiation," NCRP Report No. 10 NCRP Wash (1970).
7. H.T. Price, G.C. Horton, and K.T. Spinney, "Radiation Shielding" Pergamon Press, New York (1957) p. 146, 151.
8. International Commission on Radiation Units "Neutron Fluence, Neutron Spectra and Kerma" ICRU Report 13 (1969).
9. G.R. Holeman, D.M. Shaw and K.W. Price, "Stray Neutron Spectra and Comparison of Measurements with Discrete Ordinate Calculations" Proc. of the Second International Conference on Accelerator Dosimetry and Experience, CONF 691101 USAEC Div. of Tech. Info., Stanford (1969) p. 552.
10. Y.Y. Glazunov, M.V. Savin, I.N. Safina, E.F. Formakin and V.A. Khokhlov, "Photoneutron Spectra of Platinum, Bismuth, Lead and Uranium," Soviet Physics, JETP **29**, 1284 (1964).
11. V.V. Verbinski and J.C. Courtney, "Photoneutron Spectra and Cross Sections for  $^{12}\text{C}$  and  $^{16}\text{O}$ ," Nuclear Physics, **73** 398 (1965).
12. W.W. Kaushal, E.J. Winhold, P.F. Yergin, H.A. Medicus and R.H. Augustson, "Fast Photoneutron Spectra Due to 50-85 MeV Photons," Phys. Rev. **175** 1310 (1968).
13. F.W.K. Pirk, "Energy Spectra of Photoneutrons at Excitation Energies up to 60 MeV," Proc. of the International Nuclear Physics Conf., Gatlinburg, Tennessee 952 (1966): Academic Press, New York and London (1967).
14. J.B. McCaslin and A.R. Smith, "Radiation Measurements and Shielding Study of the Berkeley, 27-inch  $^3\text{He}$  Cyclotron," Proc. of the Second International Conference on Accelerator Dosimetry and Experience, CONF. 691101 USAEC Div. of Tech. Info., Stanford (1969) p. 486.



## EVALUATION OF PERSONNEL EXPOSURES IN A HIGH-ENERGY, HIGH-INTENSITY BETA-GAMMA FIELD\*

Bill L. O'Neal  
Sandia Laboratories  
Albuquerque, New Mexico 87115

### Abstract

A high-energy, high-intensity beta-gamma radiation field occurred in experimental areas of a tunnel complex at the NTS. The beta-to-gamma dose rate ratio in free air was as high as 28 to 1 (83 rad/hr vs 3 R/hr) and the beta-particle maximum energy was 3.5 MeV. Evaluation and recovery of experiments resulted in personnel radiation doses approaching the AEC guidelines. The methods described here were used in evaluating the radiation field and determining an estimate of the relative dose to the skin, gonads, and lens of the eye as measured by both thermoluminescent dosimeters (TLD) and film badges. In addition, problems not normally associated with mixed-field, beta-gamma dosimetry are discussed.

### 1. Introduction

Most health physicists involved with mixed-field, beta-gamma dosimetry usually find that the whole-body penetrating dose is the limiting factor in controlling personnel exposures. The dose contributions due to the beta particles may be small because of rapid absorption of the particles in air, protective clothing, and shielding, or are limited to the extremities, which have a MPD 15 times greater than that of the whole body. However, when one experiences a radiation field in which working conditions limit the protective methods such as shielding or distance, and in which the beta-to-gamma dose rate ratio and the beta energy is unusually high, the dose to the skin, the gonads or the lens of the eye may be the controlling factor. The dose to these organs in such a field is difficult to estimate because of the beta-absorption processes in both active and passive radiation detectors and the absorption between the surface and the sensitive volume of the critical organ.

Such a radiation field resulted from a nuclear-weapon effects test conducted in a tunnel complex at the Nevada Test Site. Failure of the stemming material caused a release of a complex spectrum of fission products into the experimental areas. Recovery of the experiments about ten months later resulted in controlled personnel doses approaching the AEC guidelines. The purpose of this paper is to describe the methods used in estimating the

---

\* This work was supported by the U. S. Atomic Energy Commission.

relative dose to potential critical organs, as measured by TLD's and film badges, and to discuss the health physics problems created by this unique radiation field. While many similar radiation fields were experienced, the discussion is limited mainly to the area of greatest exposure potential.

2. Source Term Evaluation

A major consideration in providing as accurate as possible a dose estimate for protection purposes was the evaluation of the field-source geometry and the radiation spectrums present. This was especially important in the areas where the dose rates were relatively high and over-exposures could occur in a short time. The geometry of a typical experimental area after the experiments were removed (Figure 1), was that of a right circular steel cylinder with a diameter of 65 inches and a length of about 30 feet.

Fission products and induced activity existed on and in the pipe walls. A powder-like dust was found generously spread over the floor of the pipe and clinging to the walls. Figure 2 shows a Ge(Li) gamma spectrum of the dust sample. The major fission products identified in the sample were  $^{106}\text{Ru-Rh}$ ,  $^{103}\text{Ru}$ ,  $^{137}\text{Cs}$ ,  $^{141}\text{Ce}$  and  $^{144}\text{Ce}$ . A ratio of 6:1 was observed between  $^{106}\text{Ru-Rh}$  and  $^{103}\text{Ru}$ , and a ratio of 20:1 between  $^{106}\text{Ru-Rh}$  and  $^{137}\text{Cs}$ . These ratios were not what might have been expected at this time based on fission yields and decay data. The amount of cerium present could not be estimated because of the high Compton region. Minor contributions to the sample were identified as  $^{124}\text{Sb}$ ,  $^{95}\text{Zr-Nb}$ ,  $^{54}\text{Mn}$ ,  $^{60}\text{Co}$ ,  $^{110\text{m}}\text{Ag}$  and  $^{140}\text{Ba-La}$ .

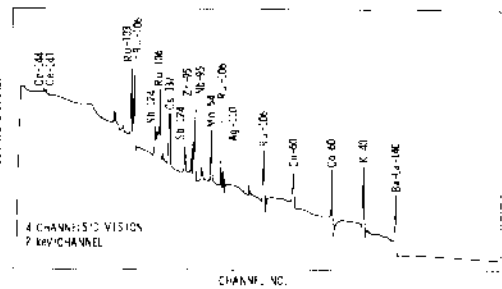
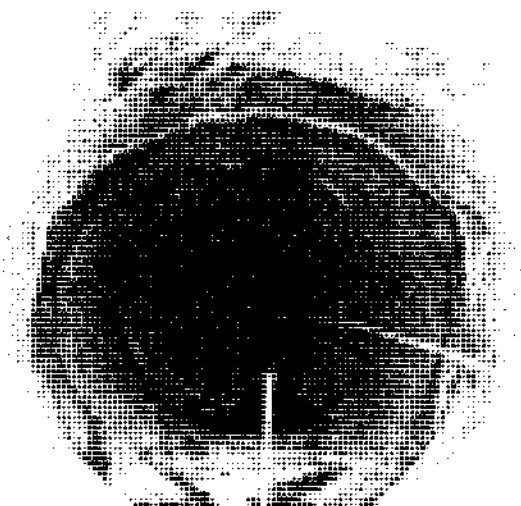
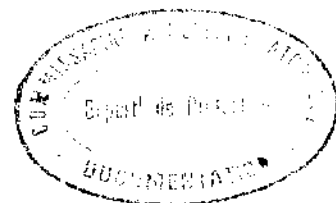


Figure 1. Geometry of experimental area

Figure 2. Ge(Li) gamma spectrum of sample collected in experimental area

Figure 3 shows a beta spectrum of the sample as measured by a 1.3-cm thick plastic scintillator (Pilot B) and a pulse-height analyzer. The spectrum indicated a maximum beta-particle energy of about 3.4 MeV, which corresponds to the beta-particle energy from  $^{106}\text{Rh}$  of 3.54 MeV.<sup>1</sup> The lower



energy value of 3.4 MeV was due to incomplete energy absorption, since the range of the beta particles was greater than the thickness of the scintillator. The range of a 3.5-MeV beta particle in aluminum is 0.6 cm<sup>2</sup>. The equivalent range in tissue and plastic ( $\rho = 1 \text{ gm/cm}^3$ ) is inversely proportional to their densities and is equal to 1.7 cm. A more detailed analysis of the spectrum indicated the relative magnitude of the beta hazard. The mean energy was about 1.0 MeV, with 65% of the beta particles having energies greater than 700 keV. The 700-keV point represents the energy absorbed before reaching the lens of the eye tissue at a depth of about 3.0 mm beneath the cornea.<sup>3</sup>

It was realized that the spectrum of beta energies in the experimental area may be considerably different from that obtained from the sample because of multiple scattering. A beta absorption curve in aluminum of the sample was compared to the theoretical absorption calculated by equation 1 and 2;<sup>4</sup> the results are shown in Table I.

$$\mu/\rho (\text{cm}^2/\text{gm}) = 17 E_{\text{max}}^{-1.43} \quad (1)$$

$$I = I_0 e^{-(\mu/\rho)(x)(\rho)} \quad (2)$$

It can be seen that the two methods compare very favorably even though the sample consists of a composite mixture of beta-emitting isotopes.

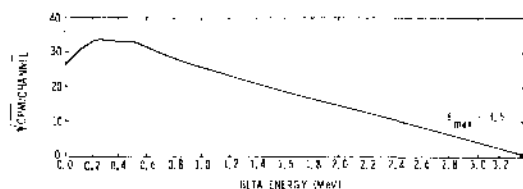


Figure 3. Beta spectrum of sample collected in experimental area

TABLE I  
Beta Absorption Data  
for the Composite Sample

Al Thickness (mg/cm <sup>2</sup> )	Actual % Transmission	Theoretical % Transmission
13.50	96	96
34.56	90	91
109.00	78	73
171.45	66	62
276.44	50	46
426.60	32	30
619.65	17	17
856.98	7	9
1089.14	3	5
1375.65	2	2
1640.53	1	1

The closeness of the comparison suggested a technique of estimating the beta spectrum inside the pipe. TLD's located inside and outside full-face masks of individuals collecting samples showed an approximate HVL of 0.16 gm/cm<sup>2</sup> or an absorption coefficient of 4.4 cm<sup>2</sup>/gm. Equation 1 and 2 then gave an approximate effective  $E_{\text{max}} = 2.6 \text{ MeV}$  and an absorption curve (Figure 4) which could be used for depth dose determinations and instrument correction factors. This relationship is valid provided the distance of penetration is well within the range of the beta particles.

### 3. Evaluation of Field Uniformity and Magnitude

Radiation-monitoring personnel dressed in appropriate anticontamination clothing surveyed the experimental area with several different portable radiation instruments calibrated to <sup>60</sup>Co radiations (PIC-6, E-520, Radector,

and Cutie Pie). Each instrument showed a wide variation of beta-plus-gamma readings but indicated a relatively uniform response throughout the length of the pipe, with a few localized hot spots. The wide variations in instrument response were expected because of differences in wall materials, wall thicknesses, and area of beta windows. The Cutie Pie was the only instrument that provided an estimate of the beta-plus-gamma exposure rate; the other instruments were limited either by an inadequate beta window or by saturation. The Cutie Pie, when roughly corrected for wall absorption, gave a beta-plus-gamma reading of ~100 rad/hr. Average gamma readings obtained from the PIC-6 and E-520 were ~2 R/hr.

However, a more accurate definition of the field strength was needed. TLD-100's were calibrated to <sup>60</sup>Co gamma radiation and uranium slugs with an assumed beta dose rate at the surface of 233 mrad/hr.<sup>5</sup> The TLD's, positioned on an exposure wheel, were exposed in the experimental area. The results, shown in Figure 5, indicated a uniform dose rate from one edge of the pipe to the other with only a slight increase close to the walls. The estimated free air exposures as measured by beta-shielded and unshielded TLD's were 83 rad/hr and 3 R/hr, or a dose-rate ratio of 28:1. These estimates were believed to be accurate and were used to limit exposure times.

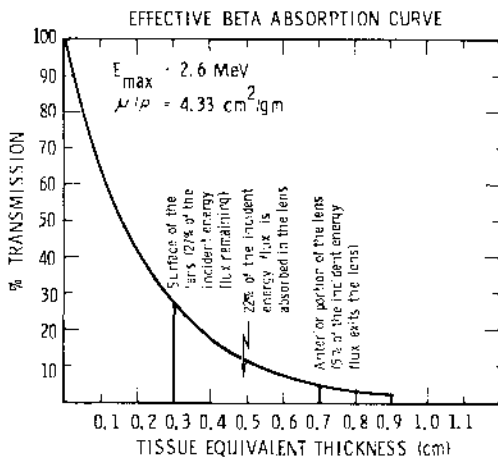


Figure 4. Effective beta absorption curve

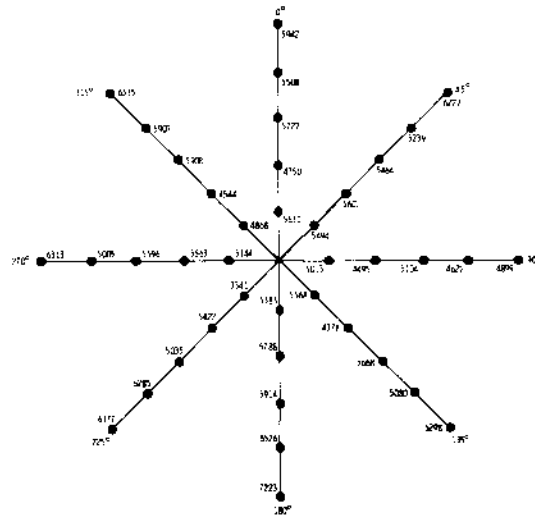


Figure 5. Exposure wheel

#### 4. Simulated Exposure Results

In order to measure and control personnel exposures more accurately during experiment evaluations or recovery operations, it was necessary to determine what perturbation effect an individual's body had when moving in the radiation field, and what organ(s) could receive the greatest radiation dose.

A TLD holder made of lucite was designed to be used in a beta-gamma field where the maximum beta-particle energy was equal to 3.5 MeV. The holder consisted of an open window region for beta-plus-gamma measurements

and a beta-shielded region 1.7 cm in thickness for gamma measurements. After rad-R response correction factors were applied, simple subtraction provided a relative estimate of both beta and gamma components. The TLD badge and the official NTS film badge, placed next to one another under suitable protective clothing at various locations on the body (Figure 6), were simultaneously exposed in the radiation field. Two different exposure conditions were simulated. One individual randomly moved about the field while a second individual remained in a relatively fixed position.

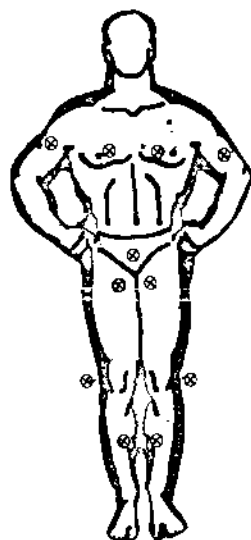


Figure 6. Dosimeter locations

Tables II and III show the dosimeter results. The gamma results for both the film and TLD's show a relatively uniform exposure independent of body location. Comparison of the gamma results between the two dosimeters agreed within  $\pm 30\%$ . Beta results depended on body location and the degree of body attenuation afforded the dosimeter. Comparison of the beta results between the two dosimeters, while not dramatic, were better than expected. Variations were generally less than a factor of 2. Generally the TLD readings were higher than the film readings. This was attributed in part to the absorption in the film wrapper ( $\sim 25 \text{ mg/cm}^2$ ), open-window edge effects and film response for a beta energy spectrum other than that used in film calibration. The rad/R ratio was the greatest on the back of the individual and suggested that the skin was the critical organ. Tables IV and V show the effect of estimating the dose to other areas of the body when the analysis is based on a single dosimeter worn on the chest. The readings less than unity indicate an underestimate of surface dose. In most cases, however, no error greater than a factor of 2 was observed.

The readings less than unity indicate an underestimate of surface dose. In most cases, however, no error greater than a factor of 2 was observed.

TABLE II

Simulated Exposure:  
Subject Randomly  
Moved about the Radiation Field

DOSIMETER LOCATION	FILM mR	TLD mR	TLD FILM	FILM rads	TLD rads	TLD FILM	RD g/y
1:1 INSIDE RIGHT LEG	140	224	0.89	2385	1981	0.83	17
1:2 INSIDE LEFT LEG	120	193	1.61	1830	2437	1.33	15
1:3 OUTSIDE RIGHT KNEE	145	142	1.02	2995	4021	1.34	27
1:4 OUTSIDE LEFT KNEE	150	154	1.02	2942	2238	0.76	24
1:5 INSIDE RIGHT LEG GROIN LEVEL	100	223	2.03	925	1503	1.62	9
1:6 INSIDE LEFT LEG GROIN LEVEL	105	113	1.07	1510	773	0.51	14
1:7 INFRONT OF GONADS	100	131	1.31	1745	2016	1.16	17
1:8 RIGHT SIDE WAIST LEVEL	105	125	1.20	2125	2956	1.39	23
1:9 LEFT SIDE WAIST LEVEL	95	-	-	1910	-	-	20
1:10 RIGHT SIDE CHEST LEVEL	95	97	0.98	1575	2409	1.53	12
1:11 LEFT SIDE CHEST LEVEL	95	-	-	1575	-	-	12
1:12 MIDDLE RIGHT UPPER ARM	150	216	1.44	3595	3554	1.02	24
1:13 MIDDLE LEFT UPPER ARM	140	-	-	3370	-	-	24
1:14 MIDDLE OF BACK, CHEST LEVEL	135	159	1.18	2453	4359	1.77	28
1:15 MIDDLE OF BACK, WAIST LEVEL	110	129	1.17	2290	2925	1.21	30
1:16 FRONT, WAIST LEVEL, OUTSIDE COVERALLS	130	105	0.80	2980	4241	1.42	23
1:17 BACK, WAIST LEVEL, OUTSIDE COVERALLS	150	222	1.48	3165	5152	1.64	21
1:18 CONTROL	-	-	-	-	-	-	-

TABLE III

Simulated Exposure:  
Subject Remained in a Fixed  
Position in the Radiation Field

DOSIMETER LOCATION	FILM mR	TLD mR	TLD FILM	FILM rads	TLD rads	TLD FILM	RD g/y
1:1 INSIDE RIGHT LEG	200	206	1.03	1230	2291	1.86	5
1:2 INSIDE LEFT LEG	170	222	1.29	2210	2416	1.06	21
1:3 OUTSIDE RIGHT KNEE	205	301	1.47	2345	4681	2.00	16
1:4 OUTSIDE LEFT KNEE	250	283	1.13	2715	4480	1.65	16
1:5 INSIDE RIGHT LEG GROIN LEVEL	175	224	1.28	1935	3229	1.67	14
1:6 INSIDE LEFT LEG GROIN LEVEL	180	185	1.03	1990	2257	1.13	11
1:7 INFRONT OF GONADS	175	161	0.92	1953	1413	0.72	16
1:8 RIGHT SIDE WAIST LEVEL	140	151	1.08	2285	4321	1.89	16
1:9 LEFT SIDE WAIST LEVEL	125	148	1.18	2865	2972	1.04	19
1:10 RIGHT SIDE CHEST LEVEL	140	158	1.13	2880	2559	1.19	14
1:11 LEFT SIDE CHEST LEVEL	130	144	1.11	2485	2266	0.91	14
1:12 MIDDLE RIGHT UPPER ARM	205	217	1.06	3330	3196	1.05	27
1:13 MIDDLE LEFT UPPER ARM	175	257	1.46	2290	3457	1.51	14
1:14 MIDDLE OF BACK, CHEST LEVEL	215	250	1.16	3370	4591	1.36	20
1:15 MIDDLE OF BACK, WAIST LEVEL	185	134	0.72	2925	5393	1.84	42
1:16 FRONT, WAIST LEVEL, OUTSIDE COVERALLS	200	157	0.78	3485	4032	1.15	18
1:17 BACK, WAIST LEVEL, OUTSIDE COVERALLS	305	204	0.67	3305	7128	2.17	35
1:18 CONTROL	-	-	-	-	-	-	-

TABLE IV

Surface Dose Ratio of Chest to Other Body Areas: Subject Randomly Moved about the Radiation Field

DOSIMETER LOCATION	RATIO OF CHEST TO OTHER BODY AREAS			
	FILM (mR)	TLD (mR)	FILM (mrad)	TLD (mrad)
121 INSIDE RIGHT LEG	0.58	0.75	0.52	0.76
122 INSIDE LEFT LEG	0.79	0.48	0.81	0.78
133 OUTSIDE RIGHT KNEE	0.65	0.63	0.56	0.47
144 OUTSIDE LEFT KNEE	0.63	0.57	0.51	0.25
151 INSIDE RIGHT LEG, GROIN LEVEL	0.95	0.46	1.56	1.8
161 INSIDE LEFT LEG, GROIN LEVEL	0.90	0.32	0.98	7.3
171 INFRONT OF GONADS	0.75	0.11	0.25	0.75
181 RIGHT SIDE, WAIST LEVEL	0.90	0.74	0.70	0.55
191 LEFT SIDE, WAIST LEVEL	1.0	-	0.78	-
110 RIGHT SIDE, CHEST LEVEL	1.0	1.0	1.0	1.0
111 LEFT SIDE, CHEST LEVEL	1.0	-	1.0	-
112 MIDDLE RIGHT UPPER ARM	0.68	0.43	0.41	0.52
113 MIDDLE LEFT UPPER ARM	0.68	-	0.44	-
114 MIDDLE OF BACK, CHEST LEVEL	0.72	0.58	0.60	0.44
115 MIDDLE OF BACK, WAIST LEVEL	0.65	0.72	0.65	0.48
116 FRONT, WAIST LEVEL, OUTSIDE COVERALLS	0.73	0.50	0.50	0.45
117 BACK, WAIST LEVEL, OUTSIDE COVERALLS	0.68	0.42	0.47	0.37
118 CONTROL	-	-	-	-

TABLE V

Surface Dose Ratio of Chest to Other Body Areas: Subject Remained in a Fixed Position in the Radiation Field

DOSIMETER LOCATION	RATIO OF CHEST TO OTHER BODY AREAS			
	FILM (mR)	TLD (mR)	FILM (mrad)	TLD (mrad)
121 INSIDE RIGHT LEG	0.58	0.75	0.37	0.96
122 INSIDE LEFT LEG	0.79	0.69	0.76	0.92
133 OUTSIDE RIGHT KNEE	0.66	0.50	0.74	0.47
144 OUTSIDE LEFT KNEE	0.54	0.53	0.54	0.45
151 INSIDE RIGHT LEG, GROIN LEVEL	0.71	0.67	0.89	0.65
161 INSIDE LEFT LEG, GROIN LEVEL	0.75	0.82	0.87	1.05
171 INFRONT OF GONADS	1.00	0.96	0.95	1.5
181 RIGHT SIDE, WAIST LEVEL	0.96	1.0	0.76	0.55
191 LEFT SIDE, WAIST LEVEL	1.2	1.02	0.93	0.77
110 RIGHT SIDE, CHEST LEVEL	1.0	1.0	1.0	1.0
111 LEFT SIDE, CHEST LEVEL	1.0	1.0	1.0	1.0
112 MIDDLE RIGHT UPPER ARM	0.56	0.70	0.53	0.38
113 MIDDLE LEFT UPPER ARM	0.77	0.60	0.75	0.64
114 MIDDLE OF BACK, CHEST LEVEL	0.63	0.66	0.51	0.48
115 MIDDLE OF BACK, WAIST LEVEL	0.73	1.13	0.61	0.40
116 FRONT, WAIST LEVEL, OUTSIDE COVERALLS	0.58	0.96	0.49	0.37
117 BACK, WAIST LEVEL, OUTSIDE COVERALLS	0.44	0.74	0.52	0.31
118 CONTROL	-	-	-	-

### 5. Evaluation and Control of Personnel Dose

The primary concerns in controlling personnel exposures were the dose to the skin at a corresponding depth of  $\sim 7 \text{ mg/cm}^2$  and the dose to the lens of the eye at a depth of  $\sim 3 \text{ mm}$ . The gonads were not considered a limiting organ due to the beta absorption that occurs between the surface and the sensitive depth at approximately  $1 \text{ cm}$ .<sup>6</sup> The guidelines followed are those recommended in the AEC manual chapter 0524: (1) Skin:  $30 \text{ rem/yr}$ ,  $10 \text{ rem/qt yr}$ , (2) Lens:  $5 \text{ rem/yr}$ ,  $3 \text{ rem/qt yr}$ , (3) Cornea:  $15 \text{ rem/yr}$ ,  $5 \text{ rem/qt yr}$ . The control and evaluation of personnel doses were based on the readings from the official NTS film badges and from TLD's located inside face masks. The film badges were worn beneath protective clothing and as close as possible to the body. In most cases only one film badge was worn, and it was worn on the chest.

The gamma readings from the film were taken to be representative of the true surface skin dose. The beta readings from the film were adjusted according to the location of the badge on the body and by a correction factor to compensate for the lower beta response when compared to the TLD data. If the badge was worn on the back, the reading was corrected for the lower beta response by multiplying by 1.5. If the badge was worn on the chest, the reading was corrected for both the lower beta response and for body attenuation by multiplying by 2.5. The gamma readings and the beta readings were then summed for a skin-dose estimate. No beta absorption was accounted for between the surface and the sensitive skin depth at  $7 \text{ mg/cm}^2$ .

The TLD's located inside the face mask provided surface dose estimates for the eye. The gamma dose was obtained from the film badge. The beta surface dose was obtained by simply subtracting the gamma component from the TLD. Since the shape of the energy spectrum remains nearly constant throughout the absorption and the angular distribution pattern for the beta particles remains substantially constant during absorption, estimates of the beta depth dose for the lens or other organs were obtained from the absorption curve in Figure 4. The dose to the lens tissue (thickness  $\sim 4 \text{ mm}$ ) was determined to be  $\sim 22\%$  of the incident dose.<sup>7</sup> This was felt to be somewhat conser-

vative due to the solid angle represented by the lens compared to the solid angle represented by the TLD located in the face mask. The dose to the gonads was about 1% of the surface dose.

No individual's exposure exceeded a reportable dose value according to the AEC guidelines. However, if the skin-dose limits recommended in the NCRP Report No. 39 of 15 rem/yr or 5 rem/qt yr were followed, a closer control of personnel exposures would have been necessary.

## 6. Conclusions

The radiation field described, though unusual, is not limited to the Nevada Test Site. A similar field can result from accidents at reactors or fuel-reprocessing plants.

The radiation field consisted of high-energy beta particles with an  $E_{\max}$  of 3.5 MeV. The field strength was relatively uniform throughout the cylindrical geometry with a free air dose rate of 83 rad/hr and 3 R/hr.

Beta-absorption formulas used for single-nuclide continuous beta spectra appear to give good approximations for samples containing a composite mixture of beta-emitting radionuclide. Therefore the determination of an effective end point energy can provide relative depth dose data for health protection purposes.

Correlation between the TLD and film badge were within  $\pm 30\%$  for the gamma exposures and varied by no more than a factor of 2 for the beta doses. The TLD readings were assumed to provide more accurate surface beta dose estimates, and the film badge readings were adjusted accordingly. Surface doses were also adjusted according to the location of the film badge on the body.

Estimates of beta depth doses were obtained from the surface dose, beta absorption relative to the tissue involved, and the depth and thickness of the sensitive region.

## References

1. C. Michael Lederer, Jack M. Hollander and Isadore Perlman, Table of Isotopes (New York: John Wiley and Sons, 1967), p. 50.
2. Radiological Health Handbook, Revised Edition (Rockville, Maryland: U. S. Department of Health, Education and Welfare, 1970), p. 123.
3. Handbook 59, Recommendations of the NCRP On: Permissible Dose From External Sources of Ionizing Radiation (Washington: National Bureau of Standards, 1954), p. 40.
4. Robley D. Evans, The Atomic Nucleus (New York: McGraw-Hill Book Co., 1955), pp. 627-8.
5. Radiological Health Handbook (op. cit.), p. 204.
6. Handbook 59 (op. cit.), pp. 38-40.
7. Robley D. Evans, Private communications (1972).

## DOSE, DOSE EQUIVALENT, EFFECTIVE DOSE AND CELL SURVIVAL FROM NEGATIVE PIONS\*

H. A. Wright, R. N. Hamm, and J. E. Turner  
Health Physics Division, Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830

### Abstract

Much interest has been shown recently in the use of negative pions for cancer radiotherapy. Several facilities are being constructed which will provide pion beams of sufficient intensity for radiotherapy and clinical trials are planned. Calculations are now being made to provide necessary information for the radiotherapist to plan the treatment of a tumor patient and also for the health physicist in insuring the protection of the patient as well as those working in the vicinity. Monte Carlo computer codes have been developed which take into account all of the relevant physical processes involved as pions penetrate tissue, e.g., inelastic nuclear interactions and the transport of all secondary particles produced, multiple coulomb scattering, range straggling, etc. Good agreement has been obtained between the results of these calculations and experimental measurements of absorbed dose. The codes can provide detailed information about the energy and spatial distributions of each type of primary and secondary particle throughout the phantom and also the LET distribution of the energy deposited. In this investigation these codes have been used to calculate the spatial distributions of absorbed dose and to study the effects of applying various types of weighting factors to the components of dose. The ICRP recommended quality factors have been applied as functions of LET to determine the spatial distribution of dose equivalent. Experimental RBE values for cell survival have been applied as functions of LET to determine the spatial distribution of "effective" dose. The cell survival model of Katz has also been applied for estimating cell survival of human T-1 cells. The results of these various weighting schemes will be discussed.

### Introduction

Negative pion beams have been used for a number of years in radiobiological experiments. More recently, attention has been focused on their use for radiotherapy. At the end of its trajectory, a stopped negative pion is captured by an atomic nucleus, causing the nucleus to disintegrate into energetic, high-LET fragments, producing a "star". A large amount of energy is thus deposited within a small volume around the capture site. The interactions of pions are complex and

---

\* Research sponsored jointly by the National Science Foundation and the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.



difficult to measure. Few experimental data exist. Fortunately, a great deal of information can be obtained from calculations which, of course, should be checked by measurements whenever possible. This paper reports some results of various calculations for negative pion beams. Some of these results are parts of manuscripts to be published in the open literature.

### Description of Computational Programs

The Monte Carlo computer code, PION-1<sup>1,2</sup> was developed for the study of dose deposition by pion beams incident on water and tissue targets. The program includes all physical interactions known to be relevant: electronic interactions (expressed by means of stopping power), pion nuclear interactions, multiple coulomb scattering, and range straggling. Muon and electron beam contamination can also be included. The complete histories of all primary and secondary particles, arising from the irradiation of a target by a pion beam, are obtained. The computer program furnishes complete microscopic data for all particles. These data can be tabulated in any desired form to study dose as a function of position, LET distributions, etc. In what follows we shall calculate, for a particular pion beam, incident on a tissue target, depth dose curves, dose equivalent, average quality factors, RBE values, and cell survival.

### Specification of Beam

The quantities below are calculated as averages in a thin cylindrical volume element of radius 3 cm, called the detector, placed at different depths along the beam axis. A beam of approximately elliptical cross section 6 x 7 cm was used for these calculations. The beam was the same as that employed in an earlier study,<sup>2</sup> except that no contamination (muons and electrons) is included here. The mean momentum of pions in the beam is 175 MeV/c. A Gaussian distribution is assumed with one-half of the pions having momenta within 2% of the mean value. The particle histories of 10,000 incident pions and all of the secondary products they generate were used in making each set of calculations.

### Absorbed Dose

The absorbed dose, averaged over the dimensions of the 3 cm detector, is shown in Figure 1 as a function of depth in the irradiated tissue target. The various components that contribute to the total absorbed dose are also shown. The curve labeled " $\pi$ " represents the ionization dose delivered by pions, that labeled "H" shows the dose from heavy particles (i. e., mass number > 1), "N" represents the neutron dose, and "P" the proton dose. Except in the peak the proton dose is almost identical to that from heavy particles.

The depth-dose curve is characterized by a plateau region to a depth of about 17 cm and by a peak which covers the region from 17 to about 23 cm. The ratio of the maximum dose to that at the surface is 3.3 for this pure beam of pions. The relative contributions to dose in the plateau and peak regions are given in Table 1. The high LET components, which arise from pion-nuclear interactions, contribute 30% of the dose in the peak.

Individual contributions to the absorbed dose can be weighted in various ways to make additional predictions about pion beams. Some examples are given in the next three sections.

### Dose Equivalent

For occupational exposure to ionizing radiation, the International Commission on Radiological Protection has recommended<sup>3</sup> the use of certain quality factors,  $Q$ , for weighting the absorbed dose deposited in different LET intervals. These values of  $Q$  were used to weight the absorbed dose components as a function of LET for each primary pion and secondary particle to obtain the dose equivalent at each depth in the target. Above 1750 MeV/cm, the value  $Q = 20$  was used. The resulting curve for the dose equivalent,  $H$ , as a function of depth is shown in Figure 2 together with the absorbed dose curve obtained from Figure 1.

These results show that the dose equivalent has its peak value approximately one cm beyond the depth at which the absorbed dose is a maximum. The dose equivalent peaks at a greater depth because a larger number of pions stop there. This fact can be seen from Figure 1, where the "star" dose has its maximum. Even though the dose is smaller behind the peak, the weighted contribution from high LET components is greater.

The individual contributions to the dose equivalent in the plateau and peak regions are also summarized in Table 1. Dividing  $H$  by  $D$  gives the "average quality factor,"  $\overline{QF}$ , for the pion beam at various depths. A somewhat more extensive study of quality factors for both negative and positive pion beams is being reported elsewhere.<sup>4</sup> The value of  $\overline{QF}$  is approximately 1.6 in the plateau region (about 5 cm depth) and rises to a value near 5 behind the region of peak dose.

### Use of Todd's RBE Values for Human T-1 Kidney Cell Survival

Todd has summarized RBE values as functions of LET for different survival levels of T-1 human kidney cells exposed to ion beams.<sup>5</sup> These values have been used as weighting factors for the absorbed dose in different LET intervals to obtain a weighted total dose, or "effective dose" as a function of depth for the pion beam. The ratio of the effective dose and the absorbed dose provides an estimate of the RBE for the beam at different depths. The weighting factors for 50% and 1% survival levels have been used to compute the curves labeled Todd in Figure 3. These curves give the estimated RBE for a given survival level at a given depth. The results indicate that the RBE's are somewhat greater than unity in the plateau region and rise to maximum values of 1.7 and 1.4 at depths slightly beyond the depth of the peak dose. The present calculations, utilizing the code PION-1,<sup>1</sup> represent a refinement of previous work.<sup>6</sup>

### Use of Katz's Model

Katz has developed a model for calculating cell survival.<sup>7</sup> This model has been incorporated to estimate human T-1 kidney cell survival levels as functions of depth in the target exposed to the pion beam. The dose was normalized so that the cell survival probability at the surface of the target was 50% in one case (190 rad at the surface) and 30% in another (280 rad at the surface). Figure 4 shows the results obtained. The survival probability in both cases increases slightly in the first 10 cm of depth, reflecting the small decrease in the absorbed dose (Figure 1). At 15 cm the survival probability begins to drop rapidly, reaching a minimum around 20.5 cm. The upper curve, for 50% survival at the surface, drops about 2 orders of magnitude to its minimum; the 30% curve drops 3 orders of magnitude.

RBE values were also calculated with Katz's model for given survival levels at each depth. Results for 50% and 1% survival are shown in Figure 3 by the curves labeled Katz. These curves can be compared directly with those labeled Todd which illustrate the same quantities computed by using Todd's RBE values<sup>5</sup> as weighting factors. The RBE values found with Katz's model at large depths are larger than those based on Todd's values for the survival levels considered here. At smaller depths Katz's 1% curve and Todd's 50% curve are about equal. The discrepancy between the curves for the same survival levels could be due to a number of factors. Perhaps the most fundamental question is the validity of applying Todd's RBE values from experiments with individual ions to the star components produced by pion capture.

#### References

1. H. A. Wright et al., in preparation.
2. J. E. Turner, J. Dutrannois, H. A. Wright, R. N. Hamm, J. Baarli, A. H. Sullivan, M. J. Berger, and S. M. Seltzer, *Radiation Research* 52, 229 (1972).
3. *Health Physics* 9, 357 (1963).
4. H. A. Wright, R. N. Hamm, and J. E. Turner, *Health Physics*, to be published.
5. Paul Todd, *Radiation Research Suppl.* 7, 196 (1967).
6. J. R. Dutrannois, H. A. Wright, J. E. Turner, and R. N. Hamm, *Int. J. Radiation Biology* 23, 421 (1973).
7. R. Katz, B. Ackerson, M. Homayoonfar, and S. C. Sharma, *Radiation Research* 47, 402 (1971).

#### Figure Captions

- Figure 1. Absorbed dose calculated as a function of depth for detector of radius 3 cm. As described in text, various contributions to the total dose are also shown: " $\pi$ " shows the dose from pion ionization, "H" that from heavy particles (mass number > 1), "P" from protons, and "N" from neutrons.
- Figure 2. Dose equivalent, H, and absorbed dose, D, as functions of depth.
- Figure 3. Comparison of RBE values for 50% and 1% survival levels based on Todd's summary of RBE and on Katz's model.
- Figure 4. Cell survival probabilities for two examples calculated as functions of depth with Katz's model. In one case the survival level at the surface is 50%; in the other case, 30%.

Table 1. Relative Contributions (Percent) to Absorbed Dose  
in Plateau\* and Peak\*\* Regions

	Absorbed Dose		Dose Equivalent	
	Plateau	Peak	Plateau	Peak
Pion Ionization	89	38	55	10
Heavy Particles	6	30	34	64
Protons	4	27	4	14
Neutrons	1	5	6	12

\* Averaged between 5 cm and 6 cm.

\*\* Averaged between 20 cm and 21 cm.

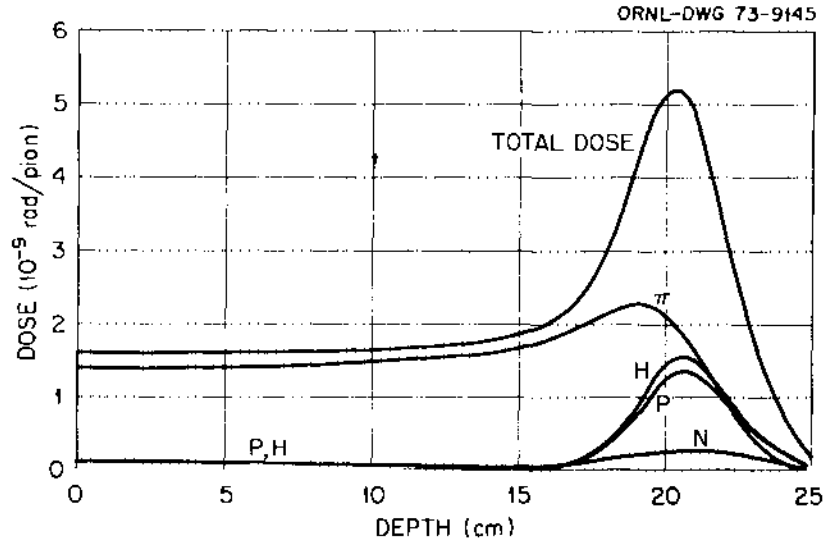


Figure 1.

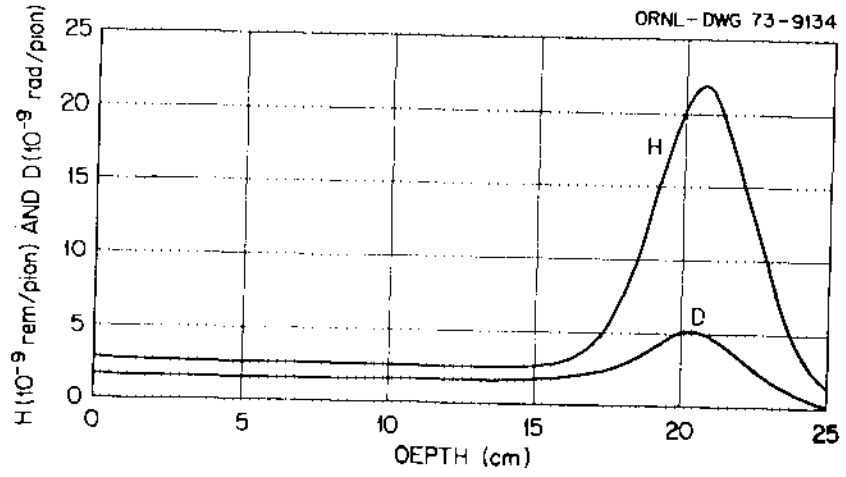


Figure 2.

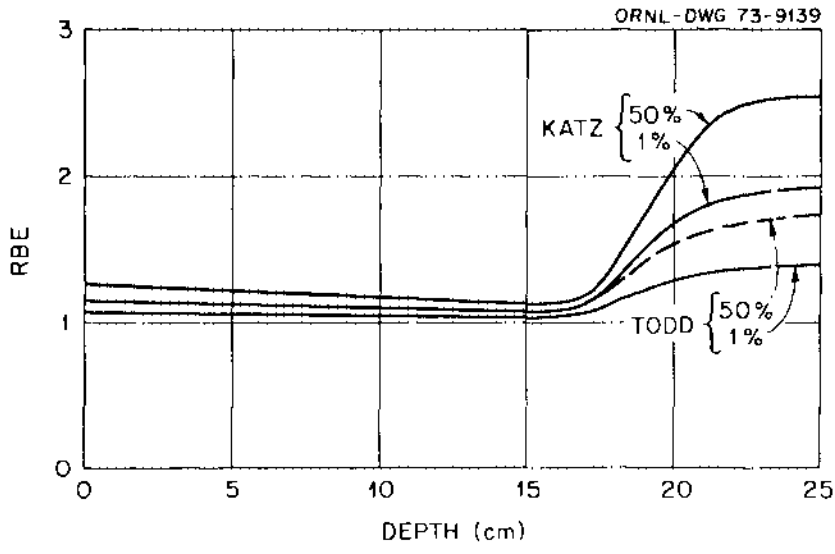


Figure 3.

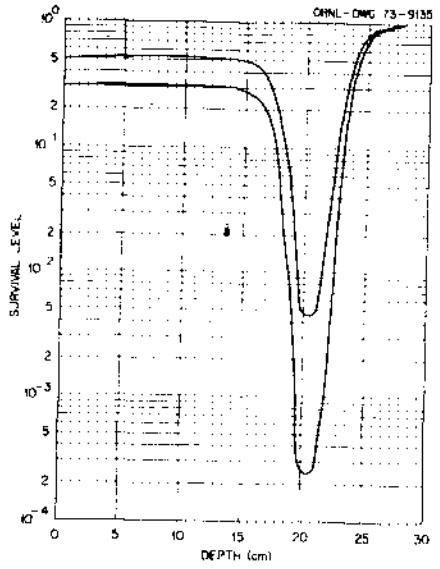


Figure 4.

712

# METABOLISM OF URANIUM AND TRANSURANIUM ELEMENTS

BIOMEDICAL FOLLOW-UP OF THE MANHATTAN PROJECT PLUTONIUM WORKERS

L. H. Hempelmann

Strong Memorial Hospital, University of Rochester, Rochester, New York

W. H. Langham (deceased), G. L. Voelz, and C. R. Richmond

Health Division, Los Alamos Scientific Laboratory  
University of California, Los Alamos, New Mexico

## Abstract

Long-term studies have been performed on 25 men who were exposed to plutonium during World War II at what is now the Los Alamos Scientific Laboratory. Almost all of the subjects had body burdens of plutonium ranging from 0.1-1.3  $\mu\text{g}$  or 6-80 nCi of relatively pure plutonium-239 as estimated from the urine assay method for plutonium used at Los Alamos prior to 1950. This paper reconstructs the war-time exposure conditions, discusses the estimates of body and lung burdens based primarily on urine assay for plutonium, and recapitulates the medical studies that have continued during the intervening years.

## Introduction

This is the story, first, of how 25 young men were heavily exposed to plutonium at what is now the Los Alamos Scientific Laboratory (at Los Alamos, New Mexico) in the days of the Manhattan Project during World War II and, second, of what has happened to them in the subsequent 27 years. All were sent to Los Alamos (Project Y) in 1944 or 1945 and given various technical jobs processing plutonium in the Chemistry and Metallurgy Research Division. All subjects had body burdens of plutonium estimated from the urine assay for plutonium used at Los Alamos before 1950 that ranged from 0.1-1.2  $\mu\text{g}^1$  (0.006-0.08  $\mu\text{Ci}$ ).

With the production and processing of plutonium on a milligram scale early in 1944 and in kilogram lots in 1945, Manhattan Project workers were exposed to a new radioactive hazard on an unprecedented scale. Since there was no practical basis for the safe handling of plutonium, the experience of the radium industry was adapted to the processing of plutonium at LASL. However, the problem of protection against plutonium and radium in practice differed by many orders of magnitude. To complicate the safe handling of plutonium further, there were no sensitive portable alpha particle counters or air samplers. Laboratory and air-borne contamination could be detected only qualitatively by paper swipes of surfaces or external nares measured in a stationary proportional alpha counter, and finally there was no method of determining body burdens of plutonium until the spring of 1945.<sup>2</sup>

The first studies of the chemistry and metallurgy of plutonium were carried out in the now destroyed D Building, an old wooden temporary building shown in Fig. 1. The exhausts of individual hoods, most without filters, can be seen.



Fig. 1. The original wooden D Building which housed the chemists and metallurgists in CMR Division.

Although every known method of protecting the workers against inhalation or ingestion of plutonium was used, the exposure conditions were deplorable by present-day standards. The makeshift methods at hand were simply inadequate to prevent exposure of the workers despite the most stringent safety regulations. As an example, unsuspected contaminated areas at laboratory benches of up to 35  $\mu\text{g}$  of plutonium were detected by the swipe method. Cumulative quantities of up to 0.5  $\mu\text{g}$  were swiped out of the external nares of some workers despite conscientious use of respirators. The most dramatic accident occurred when the first 8 g of plutonium (all that existed at that time) were being processed. The sample was spilled once on the floor, recovered, and then spilled into a centrifuge cup.

By February 1945, when kilogram quantities of plutonium began to arrive at LASL for processing and fabrication into nuclear devices, protective measures had improved considerably, and a method of assaying the body burden of plutonium had been developed. When the war ended in August 1945, 29 workers had been removed from their jobs because they contained measurable body burdens up to 1.2 g as determined by the urine assay methods available at that time. Of the 29 men, one died of a coronary, and three were dropped because future lists showed no detectable body burden.

#### Clinical, Laboratory, and Radioactivity Observations

##### Medical Observations

In 1953, a program for periodic examination of these men (financed by the U. S. Atomic Energy Commission) was established. In 1953 and again in 1955, 22 and 25 subjects of the series, respectively, were examined by physicians associated with the U. S. Atomic Energy Commission. All 25 of the men were examined by their family physicians in 1960, 17 in 1966, and 24 in 1970. In late 1971 and early 1972, 22 of the 25 subjects traveled to Los Alamos for a complete study including urine assays for plutonium, *in vivo* measurements for plutonium-239 in the chest, pulmonary cytology, and chromosome analyses. The results of these studies will be reported in detail.<sup>3</sup> Roentgenograms were also

taken of the chest, pelvis, and teeth. Except for the ailments that one would expect in a group of men mostly in their early fifties, all subjects examined were in remarkably good health. One man had died in 1959 of a coronary at age 38. All men were actively working, most as successful executives. No roentgenographic changes in the lungs or bones were apparent. The lamina dura of the jaws were intact in all cases.

Estimates of Plutonium Body Burdens from Urine Assay

The estimates of body burden of plutonium as determined by assay of the urine of the subjects made during the 20-yr observation period (1953-1972) are shown in Table I. The 1972 version of the PUQFUA (Plutonium Body Burden From

TABLE I

Plutonium Body Burden Estimates Based upon Urine Assay Data for the Subjects<sup>a</sup>

<u>Case Code No.</u>	<u>1953</u>	<u>1962</u>	<u>1972</u>
1	0.03 - 0.06	0.01	0.206
2	0.006 - 0.032	--	0.03
3	0.08	0.13	0.42
4	0.08	0.14	0.26
5	0.08	0.14	0.18
6	0.06	0.07	0.14
7	0.06	0.08	0.15
8	0.04	0.05	0.11
9	0.06	0.11	0.11
10	0.05	0.03	0.10
11	0.03	0.03	0.05
12	0.03	0.02	0.12
13	0.02	0.04	0.005
16	0.006	0.002	0.03
17	0.04	0.09	0.13
18	0.04	0.04	0.10
19	0.03	0.06	0.02
20	0.02	0.02	0.05
21	0.02	0.03	0.04
22	0.02	0.02	0.05
23	0.02	0.04	0.04
24	0.006	0.01	0.03
25	0.006	0.01	0.01
26	0.02	0.03	0.006
27	0.02	0.03	0.05

<sup>a</sup>Microcurie ± approximately 50%.



Urine Analysis) code was used to estimate what is considered to be the best value for the body burden of the subjects.<sup>4</sup> In all but 2 cases, the current estimates of body burden are higher than those in 1953, usually by a factor of 2-3 and sometimes by a factor of 5-6. Values in the last column of Table I range from 0.005-0.42  $\mu\text{Ci}$  plutonium-239,240, or from approximately 1/8-10 times the current maximum permissible body burden (0.04  $\mu\text{Ci}$ ) for occupational workers. Eighteen of the 25 values listed for 1972 are equal to or exceed the 0.04  $\mu\text{Ci}$  value. Comparing the relatively small quantities of plutonium deposited in the body (excluding the lungs) with the large amounts to which the subjects had been exposed, we can only conclude that the gastrointestinal tract has a remarkable ability to exclude plutonium from entering the body. Had plutonium been as readily absorbed as radium, all subjects would unquestionably have lethal body burdens of radioactivity.

To evaluate the possible consequences of bone doses of this magnitude, we must refer to animal data. The most extensive of these studies has been carried out at the University of Utah Medical School since December 1, 1952, when the first group of 6 beagles were injected intravenously with plutonium-239 citrate.<sup>5</sup> Injected doses ranged from 0.016-2.9  $\mu\text{Ci}/\text{kg}$  body weight for 6 groups of about 12 dogs each. To date, the 0.016  $\mu\text{Ci}/\text{kg}$  dose level is of interest, as 4 dogs developed osteosarcomas, giving a tumor incidence of 33%.<sup>5</sup> Average time from injection to death for these animals was 9.9 yr, and the average skeletal dose from injection to death was estimated to be about 80-90 rads delivered at about 8-9 rads/yr.<sup>6</sup>

It is difficult to estimate even crudely bone and liver doses from the data in Table I. However, some estimates can be obtained if numerous simplifying assumptions are made. For example, the average body burden for 1972 in Table I is about 0.10  $\mu\text{Ci}$  which, if equally divided between bone and liver, would deliver about 0.7 rad/yr and 2.9 rads/yr, respectively, to these tissues. If no loss occurred during the 27 yr since exposure, the corresponding total doses to bone and liver would be about 19 and 78 rads. For the highest body burden listed in Table I (case No. 3), the estimated dose rates for bone and liver would be  $\sim$ 2.9 and 12.2 rads/yr, respectively. If the entire average burden to the group was limited to bone, the dose rate would be 1.4 rads/yr and the 27-yr dose, using the same assumptions, would be about 38 rads to the skeleton. For case No. 3, the comparable values would be about 5.9 rads/yr and 195 rads. These estimates, as indicated above, are very crude and should not be considered as being otherwise.

#### Determination of Plutonium in the Body by *In Vivo* Measurements

During the most recent medical examinations at Los Alamos, estimates were made of the amount of plutonium in the chest (lung and respiratory lymph nodes) of each subject using an *in vivo* lung counter. Measurements also were made of the liver region in several subjects and of the hands of persons known to have had skin wound contaminated by plutonium. Positive counts suggesting chest burdens ranging from 3 to about 10 nCi were obtained for 14 of 21 persons measured. However, in no case did the estimated chest burden exceed the minimum detectable level at the 95% confidence level. Seven of the 14 subjects with positive chest counts had estimated chest burdens of 7 nCi or greater and may be considered (at the 68% level of confidence) to have statistically significant chest burdens of from 7-10 nCi.

The only direct measurement of plutonium in the lungs in this study was made on the operative specimens of subject No. 2 who had surgery for a benign lung tumor. Table II shows results of analyses for plutonium-239. The concentration of plutonium-239 was approximately the same in the tumor and lung tissue, while the concentration of plutonium-239 in bone was approximately half.

TABLE II

Plutonium-239 Content of Tissues  
Removed from Subject No. 2 in May 1971<sup>a</sup>

<u>Tissue</u>	<u>pCi/g Wet Weight</u>
Lung	3.85
Lymph node	205.00
Hamartoma	3.40
Rib	1.61

<sup>a</sup>After ashing and dissolution of tissue.

The highest concentration, observed in the lymph node tissue, is consistent with experimental findings in dogs exposed to PuO<sub>2</sub> by inhalation, as mentioned above. It is well established that the concentration in lymph nodes relative to that in lung tissue increases as a function of time following exposure. If one assumes a homogeneous distribution of plutonium-239 throughout the 1 g and lymph nodes and a total lung weight of 1000 g and respiratory lymph node weight of 20 g, the total plutonium burden of the lungs and respiratory lymph nodes is approximately 8 nCi roughly equally divided between lung and lymph node. For reference, the total amount of plutonium in the lung of case No. 2 (estimated by tissue assay to be 3.85 nCi) is approximately 550 times contemporary total lung burdens in humans in the United States exposed to fallout resulting from plutonium dispersed by atmospheric weapons tests. Estimates of the chest burden of plutonium-239 of subject No. 2, based on extrapolation from analysis of lung and lymph node tissue, are in reasonable agreement (a factor of about 2) with estimates based on chest-counting.

Chromosome analysis of lymphocytes in the peripheral blood revealed no abnormalities, and studies of exfoliated pulmonary cells showed mild to marked dysplastic changes particularly in heavy smokers.

#### Vital Statistics Information

Although our study group is relatively small, about 25 men, the magnitude of their plutonium burdens, the long time since exposure, and their cooperativeness collectively make them a unique and extremely valuable group. However, because something like 16-20% of all deaths annually in the United States are from cancer, one might be concerned about the size of the group as 4 or 5 might be expected to die from "naturally occurring" cancer had they never been exposed to plutonium. However, evidence obtained from experimental animal studies very strongly indicates that the kinds of cancer induced by plutonium are not all increased in a nonspecific way but, rather, consist primarily of lung carcinomas, liver cancer, and bone sarcomas from the modalities of exposure involved.

We have used vital statistics data<sup>7</sup> to estimate the probabilities of death from certain cancer types over a 65-yr period from age 20 to age 85. The total probabilities per 100,000 deaths for tumor type 23 (bronchus, trachea, and lung, specified as primary), type 16 (liver), and type 48 (bone) are 3023, 578, and 152, respectively, for United States white males. Thus, for a group of 25, we would expect 0.76, 0.15, and 0.04 deaths from tumor types 23, 16, and 48, respectively. Therefore, in lieu of other information, the occurrence of one lung

cancer could be ascribed to nonradiation factors, whereas the occurrence of one liver cancer might be considered suggestive of plutonium induction, and the occurrence of one bone sarcoma would be statistically significant.

One also needs to consider the individual smoking histories because of the difference in lung cancer incidence history between smokers and nonsmokers. In addition, these subjects have been exposed to a large number of chemicals, some of which are known to cause biological effects, both during the Manhattan Project and subsequent employment. The following is a list of materials to which one person has been exposed: organic plasticizers, xylene, toluene, urea, formaldehyde, sulfonic acid, sodium thiosulfate, tetraethyl lead, freon, ethyl chloride, fluorinated hydrocarbons, and industrial sludges containing lead and mercury. It is also known that several of our subjects could have been significantly exposed to beryllium at a time when virtually no safety precautions were observed because of its alleged lack of toxicity. Hopefully, these follow-up studies will be continued throughout the lifetime of these subjects. This information, even though very limited, is human experience of the most relevant kind for establishing value judgments where inadequate data exist for formulating risk evaluations.

#### Acknowledgments

This work was supported by the Division of Biomedical and Environmental Research, U. S. Atomic Energy Commission, under Contract AT-(30-1)4284 to the School of Medicine and Dentistry, University of Rochester, and under Contract W-7405-ENG-36 to the University of California, Los Alamos, New Mexico.

This report covers information and work that has been carried out over a period of many years and has involved persons too numerous to mention who have contributed to the success of the project.

#### References

1. W. H. Langham and J. B. Storer, Eds., "Annual Report of the Biomedical Research Group of the Health Division, 1952," Los Alamos Scientific Laboratory Report LA-1537 (June 1953), p. 8.
2. W. H. Langham, "Determination of Plutonium in Human Urine," Los Alamos Scientific Laboratory Report LAMS-603 (December 1947); also U. S. Atomic Energy Commission Report MDDC-1555, Technical Information Division, Oak Ridge, Tenn. (March 1945).
3. L. H. Hempelmann, W. H. Langham, C. R. Richmond, and G. L. Voelz, Manhattan Project Plutonium Workers: A Twenty-Seven Year Follow-Up Study of Selected Cases, Health Phys. (1973), in press.
4. J. N. P. Lawrence, PUQFUA, An IBM 704 Code for Computing Plutonium Body Burdens, Health Phys. 8, 61 (1962).
5. C. W. Mays and T. F. Dougherty, Progress in the Beagle, Studies at the University of Utah, Health Phys. 22, 793 (1972).
6. C. W. Mays, G. N. Taylor, W. S. S. Jee, and T. F. Dougherty, Speculated Risk to Bone and Liver from  $^{239}\text{Pu}$ , Health Phys. 19, 601 (1970).
7. F. Burbank, "Patterns in Cancer Mortality in the United States: 1950-1967," National Cancer Institute Monograph 33 (May 1971).

## SOLUBILITY CHANGES OF $^{238}\text{Pu}$ OXIDE IN WATER SUSPENSION AND EFFECT ON BIOLOGICAL BEHAVIOR AFTER INHALATION BY BEAGLE DOGS\*

J. F. Park, D. L. Catt, D. K. Craig, R. J. Olson and V. H. Smith

Biology Department, Battelle, Pacific Northwest Laboratories  
Richland, Washington, 99352, U.S.A.

Abstract -- Beagles were exposed to aerosols of  $^{239}\text{PuO}_2$  or  $^{238}\text{PuO}_2$  prepared by identical methods of calcining the oxalate at  $750^\circ\text{C}$  for 2 hours. The  $\text{PuO}_2$  was stored in water suspensions of 2 to 3 mg  $\text{PuO}_2/\text{ml}$  for various periods until required for exposure at which time suspensions of suitable concentrations were prepared by dilution of the stock. Aerosols were generated by nebulizing these suspensions. Ultrafilterability of the  $^{239}\text{PuO}_2$  suspension remained stable ranging from 0.1 to 0.2% over a 16 month period. Dogs exposed to  $^{239}\text{PuO}_2$  during this time and sacrificed 30 to 140 days postexposure had more than 98% of the body burden at death in the lungs and thoracic lymph nodes. Dogs exposed to  $^{238}\text{PuO}_2$  6 months after preparation of the stock suspension had 64% and 50% in the lungs and thoracic lymph nodes, with 23% and 34% in the skeleton and 8% and 11% in the liver at 30 and 90 days postexposure, respectively. Ultrafilterability of the stock  $^{238}\text{PuO}_2$  suspension was 25%. X-ray diffraction analyses of the  $^{239}\text{PuO}_2$  and of freshly prepared  $^{239}\text{PuO}_2$  yielded the expected peaks, but the  $^{238}\text{PuO}_2$  that had been in water suspension for 9 months showed no X-ray peaks. Dogs exposed to freshly prepared  $^{239}\text{PuO}_2$  with 0.2% ultrafilterability showed more than 98% of the Pu in the lungs and thoracic lymph nodes 30 and 60 days postexposure, while the ultrafilterability of the water suspension changed from 0.2% to 16% during the 60-day postexposure period. Radiation damage to  $^{238}\text{PuO}_2$  may be responsible for the differences in the behavior of  $^{238}\text{PuO}_2$  and  $^{239}\text{PuO}_2$  in water suspension and in vivo.

### INTRODUCTION

In preparation for a life-span dose-effect-relationship study with beagle dogs comparing inhaled  $^{239}\text{PuO}_2$  and  $^{238}\text{PuO}_2$ , small groups of dogs were exposed to compare the short-term retention and translocation of the two isotopes before exposing the larger group of dogs. The dogs that inhaled  $^{238}\text{PuO}_2$  showed much more translocation of Pu to the liver and skeleton during a 3-month postexposure period than dogs exposed to  $^{239}\text{PuO}_2$ . Results of preliminary studies to investigate the difference in behavior of the two isotopes are the subject of this paper.

### METHODS

Eighteen month-old beagle dogs were exposed for 5 to 30 minutes to aerosols of  $^{238}\text{PuO}_2$  or  $^{239}\text{PuO}_2$ . These oxides were prepared by identical methods, involving calcining plutonium oxalate at  $750^\circ\text{C}$  for 2 hours. After calcining, the  $\text{PuO}_2$  was stored in water at a concentration of 2 to 3 mg  $\text{PuO}_2/\text{ml}$ . This suspension was stored (i.e., aged) until required for exposure of dogs, when suspensions of suitable concentrations were prepared by dilution of the stock suspension and aerosols were generated by nebulizing these suspensions<sup>1</sup>. Dogs

were exposed via a mask from an aerosol exposure chamber as described previously<sup>2</sup>.

Table 1 shows the groups of dogs available for comparison. The aged <sup>238</sup>PuO<sub>2</sub> stock suspension had been in water suspension for 6 months when the dogs were exposed. Ultrafilterability, measured by the methods of Lindenbaum and Westfall<sup>3</sup>, using 24 Å pore-size visking-type membrane, of this aged stock suspension was 25% and the aerosolized particle size distribution, determined using a cascade type impactor<sup>4</sup>, was smaller than that of the freshly prepared <sup>238</sup>PuO<sub>2</sub> or the aged <sup>239</sup>PuO<sub>2</sub>.

Table 1. Experimental Groups for Comparison of the Behavior of Inhaled <sup>238</sup>PuO<sub>2</sub> and <sup>239</sup>PuO<sub>2</sub> in Beagles

Pu Suspension	Number of Dogs	Sacrifice (Days After Exposure)	Age of Stock Suspension	Ultra-filterability of stock Suspension	Particle Size	
					AMAD*	GSD**
Aged <sup>238</sup> PuO <sub>2</sub>	3	27-37	6 mo	25%	0.8	2.5
Aged <sup>238</sup> PuO <sub>2</sub>	3	91-92	6 mo	25%	0.9	2.5
Fresh <sup>238</sup> PuO <sub>2</sub>	1	29	48 h	0.2%	2.3	2.3
Fresh <sup>238</sup> PuO <sub>2</sub>	2	54	48 h	0.2%	2.0	2.3
Fresh <sup>238</sup> PuO <sub>2</sub>	2	78	48 h	0.2%	2.2	2.3
Aged <sup>239</sup> PuO <sub>2</sub>	3	28-30	16 mo	0.2%	2.6	1.7
Aged <sup>239</sup> PuO <sub>2</sub>	4	140-142	3 mo	0.2%	2.5	1.8

\* AMAD Mean activity median aerodynamic diameter

\*\* GSD Mean geometric standard deviation

The freshly prepared <sup>238</sup>PuO<sub>2</sub> was stored in water suspension only 48 hours before exposing the dogs. Ultrafilterability of this suspension was about 0.2% and the aerosolized particle size distribution was more like the distribution for the <sup>239</sup>PuO<sub>2</sub> aerosols.

The <sup>239</sup>PuO<sub>2</sub>, prepared in the same way as the <sup>238</sup>PuO<sub>2</sub>, was in suspension for 16 months prior to exposure of one group of dogs and for 3 months prior to exposure of the other group. The ultrafilterability and particle size distribution of the aerosols for both <sup>239</sup>PuO<sub>2</sub> groups was similar.

Dogs were sacrificed from 1 to 5 months after exposure. Tissues and excreta were analyzed for Pu content by liquid scintillation counting following ashing and dissolution in HNO<sub>3</sub>-HF<sup>5</sup>.

## RESULTS

Table 2 shows the distribution of plutonium in the dogs at sacrifice expressed as mean % final body burden. The final body burden ranged from 0.01 to 4.8 µCi in the dogs. No influence of dose on Pu tissue distribution was observed. The dogs exposed to <sup>239</sup>PuO<sub>2</sub> had nearly all of the Pu in the lungs and thoracic lymph nodes (98-99%) at 30 and 140 days after exposure with very little translocation to other tissue, except the thoracic lymph nodes, regardless of the age of the suspension.

Table 2. Tissue Distribution of Inhaled Pu in Beagles

Pu Suspension	Sacrifice (Days After Exposure)	Mean % Final Pu Burden			
		Lungs	Thoracic Lymph Nodes*	Liver	Skeleton
Aged $^{238}\text{PuO}_2$	27-37	63	1.0	7.5	23
Aged $^{238}\text{PuO}_2$	91-92	43	6.7	11	34
Fresh $^{238}\text{PuO}_2$	29	98	0.7	0.3	0.5
Fresh $^{238}\text{PuO}_2$	54	93	5.0	0.6	0.9
Fresh $^{238}\text{PuO}_2$	78	91	5.0	0.8	1.8
Aged $^{239}\text{PuO}_2$	28-30	97	0.8	0.07	0.2
Aged $^{239}\text{PuO}_2$	141-142	96	4.0	0.01	0.05

\* Tracheobronchial, mediastinal and sternal lymph nodes

The Pu tissue distribution in the dogs exposed to fresh  $^{238}\text{PuO}_2$  was similar to the dogs exposed to  $^{239}\text{PuO}_2$  with 96 to 98% of the final body burden in the lungs and thoracic lymph nodes at 29 to 78 days after exposure. Translocation was primarily to the thoracic lymph nodes. These dogs had more Pu in the skeleton and liver than the dogs exposed to  $^{239}\text{PuO}_2$ . There was a trend toward  $^{238}\text{Pu}$  translocation from the lung and accumulation in the thoracic lymph nodes, liver and the skeleton with time after exposure.

The Pu tissue distribution in the dogs exposed to aged  $^{238}\text{PuO}_2$  suspension was very different from the dogs exposed to aged  $^{239}\text{PuO}_2$  suspensions or dogs exposed to fresh  $^{238}\text{PuO}_2$  suspensions. There was much more translocation to liver, 8 to 11% of the final body burden; and skeleton, 23 to 34% of the final body burden in the dogs exposed to aged  $^{238}\text{PuO}_2$ . This appeared reasonable, since the particle size of the aged  $^{238}\text{PuO}_2$  aerosols was smaller than that of the fresh  $^{238}\text{PuO}_2$  and  $^{239}\text{PuO}_2$  and since the ultrafilterability of the aged  $^{238}\text{PuO}_2$  suspension was 25% compared to 0.2% for the fresh  $^{238}\text{PuO}_2$  suspension and aged  $^{239}\text{PuO}_2$  suspension. A higher solubility of the aged  $^{238}\text{PuO}_2$  was expected because of the relatively greater surface area with smaller particles.

Table 3 shows the fraction of total plutonium deposited in the dog that was retained in the body or excreted in urine or feces. The fraction retained was largest for inhaled  $^{239}\text{PuO}_2$  followed in descending order by aged  $^{238}\text{PuO}_2$  and fresh  $^{238}\text{PuO}_2$ . Both  $^{238}\text{Pu}$ -exposed groups excreted a larger fraction in the feces than did the  $^{239}\text{PuO}_2$ -exposed dogs. The fraction excreted in the urine of the dogs exposed to aged  $^{238}\text{Pu}$  was about 10 times larger than the fraction excreted in the urine of the other two groups but still represented less than 0.5% of the plutonium initially deposited.

It seemed surprising that storage in water for 6 months could so markedly change the physical and biological behavior of the  $^{238}\text{PuO}_2$ . X-ray diffraction analyses of 19 month old suspensions of  $^{239}\text{PuO}_2$  and freshly prepared 72 hour old  $^{238}\text{PuO}_2$  suspensions showed the expected peaks but  $^{238}\text{PuO}_2$  that had been in water suspension for 9 months showed no X-ray peaks, indicating an alteration in crystal structure. We measured the ultrafilterability of  $^{238}\text{PuO}_2$  suspensions at periodic intervals following their preparation with the results shown in Table 4. Electron micrographs of the aerosolized aged  $^{238}\text{PuO}_2$  particles collected on thermal precipitator<sup>6</sup> grids were much smaller and more regularly shaped than the fresh  $^{238}\text{PuO}_2$  and  $^{239}\text{PuO}_2$  particles (Figure 1).

Table 3. Retention and Excretion of Inhaled Pu in Beagles

Pu Suspension	Sacrifice (Days After Exposure)	MEAN % INITIAL PU BURDEN		
		Final Body Burden	Fecal Excretion	Urinary Excretion
Aged $^{238}\text{PuO}_2$	31	67	32	0.36
Aged $^{238}\text{PuO}_2$	92	64	36	0.48
Fresh $^{238}\text{PuO}_2$	29	27	73	0.04
Fresh $^{238}\text{PuO}_2$	54	25	75	0.02
Fresh $^{238}\text{PuO}_2$	78	18	81	0.06
Aged $^{239}\text{PuO}_2$	29	85	16	0.05
Aged $^{239}\text{PuO}_2$	141	92	8	0.07

\* Tracheobronchial, mediastinal and sternal lymph nodes

Table 4. Ultrafilterability of  $^{238}\text{PuO}_2$  Suspension

Age of Stock Suspension (Days)	Ultrafilterability Mean $\pm$ SD (%)
1	0.17 $\pm$ 0.04
3	0.38 $\pm$ 0.04
9	1.70 $\pm$ 0.17
26	8.1 $\pm$ 2.3
35	12.1 $\pm$ 0.49
65	15.8 $\pm$ 0.70

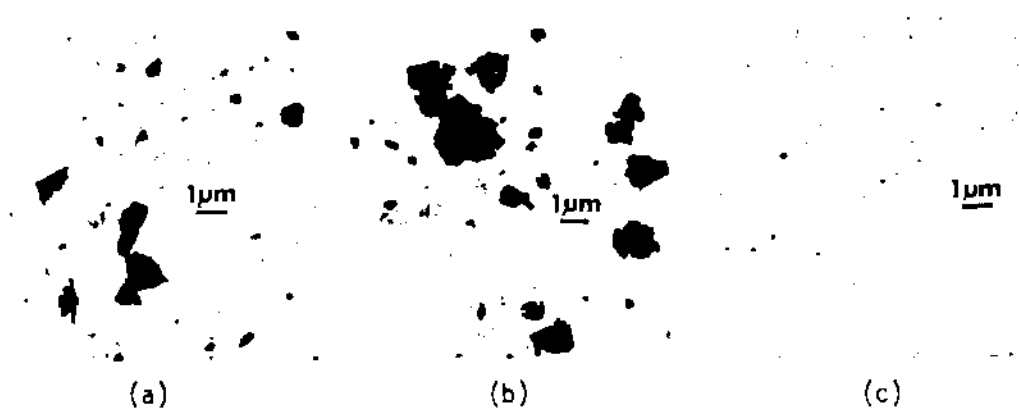


Figure 1. Electron Micrographs of (a) Aged  $^{239}\text{PuO}_2$  Aerosol, (b) Fresh  $^{239}\text{PuO}_2$  Aerosol and (c) Aged  $^{238}\text{PuO}_2$  Shadowed at  $26^\circ$ .

### DISCUSSION

The smaller AMAD of the aged  $^{238}\text{PuO}_2$  may have influenced the fraction translocated to the liver and skeleton; however, the lack of X-ray defraction peaks, the size and shape of the particles and the higher ultrafilterability indicate a change in the chemical form of the  $^{238}\text{PuO}_2$  during storage in water. The trend with time after exposure toward increased translocation from the lung and accumulation of Pu in the liver and skeleton of the dogs exposed to freshly prepared  $^{238}\text{PuO}_2$ , suggest that similar changes in chemical form of  $^{238}\text{PuO}_2$  may occur in the animals. Rats exposed to  $^{238}\text{PuO}_2$  showed a high rate of translocation to the skeleton - 11% of the body burden at 20 days and over 20% at a year or more after exposure - compared to less than 2% in the skeleton of rats exposed to  $^{239}\text{PuO}_2$ . In other studies, dogs exposed to  $^{238}\text{PuO}_2$  aerosols have shown higher rates of translocation to skeleton, up to 13% of the body burden after 6 months, compared to less than 1% for  $^{239}\text{PuO}_2$ -exposed dogs<sup>8</sup>. Table 5 shows distribution of  $^{238}\text{Pu}$  in the tissue of dogs 5 to 6 years after inhalation of  $^{238}\text{PuO}_2$ , compared to dogs sacrificed at similar times after inhalation of  $^{239}\text{PuO}_2$ . The much greater translocation of  $^{238}\text{Pu}$  to the skeleton and liver, despite the low ultrafilterability and fresh state of the water suspension employed in generating the aerosol, suggest that solubilization of the  $^{238}\text{PuO}_2$  occurs to a significant degree within the dog, as well as in water suspension. The  $^{239}\text{PuO}_2$ -exposed dogs died due to lung tumors and the  $^{238}\text{PuO}_2$ -exposed dogs died due to bone tumors<sup>9,10</sup>.

*Table 5. Tissue Distribution of Pu in Beagles After Inhalation of  $^{238}\text{PuO}_2$  and  $^{239}\text{PuO}_2$*

<u>Tissue</u>	Percent of Final Body Burden <sup>+</sup>					
	$^{238}\text{PuO}_2$ *			$^{239}\text{PuO}_2$ **		
	58***	60	62	54	57	68
Lung	6	7	17	55	49	35
Lymph Nodes	10	11	9	24	27	37
Liver	23	33	22	16	15	19
Skeleton	55	43	47	3	5	4
All Other Tissues	6	6	5	2	4	5
Final Body Burden ( $\mu\text{Ci}$ )	2.5	2.3	2.2	1.8	1.2	1.4

+ Values are for individual dogs

\* Calcined 350°C, ultrafilterability 1-2%, CMD 0.1  $\mu\text{m}$ , animal exposed 2 days after preparation of the water suspension

\*\* Calcined 350°C, ultrafilterability <1%, CMD 0.1-0.5  $\mu\text{m}$

\*\*\*Months after exposure

The chemical and physical differences between  $^{238}\text{PuO}_2$  and  $^{239}\text{PuO}_2$ , which caused the differences in their behavior in water suspension and in vivo, are not completely understood. Higher in vitro solubility of respirable  $^{238}\text{PuO}_2$  particles, as compared to  $^{239}\text{PuO}_2$  particles, has also been reported by Raabe, et al.<sup>11</sup>.

Due to the small amount of information on the biological behavior of  $^{238}\text{PuO}_2$ , it has generally been assumed that  $^{238}\text{PuO}_2$  would behave like  $^{239}\text{PuO}_2$ . This is quite evidently not the case in the animal and it may not be the case in environmental contamination.

Since both isotopes are present in varying proportions in reactors, and in the wastes from fuel reprocessing, we should learn how the ratio of  $^{238}\text{Pu}$  to  $^{239}\text{Pu}$  influences the physical and biological behavior of mixtures. We should



also learn if this is related to the high specific activity of  $^{238}\text{Pu}$  and whether other alpha-emitting, high specific activity radionuclides present in the fuel cycle may influence the chemical, physical and biological behavior of the mixture of radionuclides.

Acknowledgment -- The authors would like to acknowledge the contribution of J. Barnett, E. F. Blanton, V. T. Faubert, J. Fitzner, M. Snyder, G. L. Webb and E. L. Wierman for technical assistance in various phases of the work, of A. C. Case, J. P. Herring, G. J. Powers and J. L. Ryan for Pu preparation and analyses and of V. A. Harmon, Westinghouse Hanford Company for X-ray diffraction analyses.

\* Work performed under U. S. Atomic Energy Commission Contract AT(45-1)-1830.

#### REFERENCES

1. Craig, D. K., R. L. Buschbom and J. P. Herring. Health Phys. 24: 637-644 (1973).
2. Craig, D. K., J. M. Thomas, J. R. Decker and J. F. Park. Health Phys. 22: 845 (1972).
3. Lindenbaum, A. and W. Westfall. Int. J. Appl. Radiat. Isotopes, 16: 545 (1965).
4. Mercer, T. T., M. I. Tillery and G. J. Newton. J. Aerosol Sci. 1: 9 (1970).
5. Keough, R. F. and G. J. Powers. Analyt. Chem. 42: 419 (1970).
6. Walkenhorst, W. Beitr. Silikoforsch. 18: 27 (1952).
7. Stuart, B. O., W. J. Bair, W. J. Clarke and E. B. Howard. Technical Report No. AFWL-TR-68-49, Air Force Weapons Laboratory, Kirtland Air Force Base, New Mexico (1968).
8. Park, J. F., E. B. Howard and W. J. Bair. Technical Report No. AFWL-TR-69-75, Air Force Weapons Laboratory, Kirtland Air Force Base, New Mexico (1969).
9. Park, J. F., W. J. Bair and R. H. Busch. Health Phys. 22: 803-810 (1972).
10. Park, J. F. et al. In: Pacific Northwest Laboratory Annual Report for 1973, BNWL-1850, Pt. 1. Battelle-Northwest, Richland, WA (In Press).
11. Raabe, O. G., G. M. Kanappily and H. A. Boyd. In: Inhalation Toxicology Research Institute/Annual Report for 1972, LF 46: 24-30. Lovelace Foundation for Medical Education and Research, Albuquerque, NM.

THE BIOLOGICAL DISPOSITION OF  $\text{Es}(\text{NO}_3)_3$  IN RATS AFTER INTRAVENOUS,  
INTRAMUSCULAR AND SUBCUTANEOUS ADMINISTRATION\*

V. H. Smith

Battelle, Pacific Northwest Laboratories, Richland, WA, 99352, U.S.A.

Abstract -- It would be a rare event that individuals would acquire hazardous depositions of einsteinium, but as a member of the actinide elements, and to be prepared for that rare event, its behavior in vivo is of interest.

Adult, female, Wistar-strain rats were injected with 4.76  $\mu\text{Ci}$  Es as the nitrate in 0.20 cc pH 2.0 solution (90% ultrafilterable) intravenously (i.v.), intramuscularly (i.m.) and subcutaneously (s.c.).

Regardless of the injection route,  $\text{Es}(\text{NO}_3)_3$  was retained preponderantly in the skeleton up to 24 days after administration. The liver burden decreased from 26% of the initial Es administered i.v. at 4 hours to 14% at 1 day and <2% at 24 days. The skeletal content increased to about 70% on day 7 and decreased to about 56% by day 24. The liver and skeletal retention at 24 days from the s.c. and i.m. injections was slightly less than from the i.v. route. Injection site retention was greater for the s.c. route, 16%, than for the i.m. route, 8%. The popliteal lymph nodes adjacent to the i.m. injection site retained about 0.7% of the injected dose/gram compared to about 0.1 that value for the other lymph nodes and routes. Kidney concentrations at 24 days were higher from the i.m. and s.c. routes, 0.56%/g, and 0.43%/g, than from the i.v. route, 0.33%/g and all were higher than the liver concentrations of 0.11-0.16%/g wet weight. In common with other actinides, Es was concentrated more in the vertebrae and sternum than in other bones. Excretion was higher initially in the urine, but by the 5th day more was being excreted in the feces. Skin absorption was very high, about 4% of the available Es (2.5  $\mu\text{Ci}/\text{cm}^2$ , pH 2 nitrate solution) being absorbed in 30 min.

INTRODUCTION

The production of californium-252 is accompanied by small, but potentially hazardous, amounts of einsteinium-253; an alpha-emitting actinide with a physical half-life of 20.47 days. During isolation and as the properties of Es are being studied in various laboratories, the potential for human-exposure

exists. The biological disposition of Es is of interest to predict its behavior in man, as a basis for estimating the efficacy of therapy procedures and to compare

---

\*This paper is based on research performed under the United States Atomic Energy Commission Contract AT(45-1)-1830.

with the behavior of actinides lower in atomic number. Based on its ionic radius, the single ionization state of +3 and the shielding arrangement of the 5f electrons, Es should behave similarly to the lanthanide, terbium. For such comparisons,  $\text{Es}(\text{NO}_3)_3$  was administered to rats since this is a form in which other actinides are commonly encountered.

#### EXPERIMENTAL

$^{253}\text{Es}$   $\text{EsCl}_3$ , obtained from the Oak Ridge National Laboratory, was converted to  $\text{Es}(\text{NO}_3)_3$  by repeated evaporation with  $\text{HNO}_3$ . The resulting material was kept in 2N  $\text{HNO}_3$  and just prior to use portions were converted to a pH 2.0 solution as required. Using Visking membranes the  $\text{Es}(\text{NO}_3)_3$  was shown to be at least 90% ultrafilterable. Groups of 8 female, Hill-Top Wistar rats of about 275-300 grams were injected via the tail vein under light ether anesthesia, intramuscularly in the gluteus maximus, or subcutaneously in the nuchal region, without anesthesia, using 0.20 cc of a pH 2 solution containing 4.76  $\mu\text{Ci}$   $^{253}\text{Es}$ . A group of anesthetized rats had 0.786  $\text{cm}^2$  of shaved skin, between the shoulder blades, exposed for 30 minutes to 2.0  $\mu\text{Ci}$  Es in 0.15 ml of a pH 2 nitrate solution.

Animals were housed 4 rats per cage. Pooled excreta collections were made from each cage. Tissues and excreta were ashed and assayed for Es content by methods described previously<sup>1</sup>. Data have been corrected for radioactive decay to the time of administration.

#### RESULTS

Intravenous Es was translocated to skeleton as shown in Figure 1, apparently peaking around day 7; the early liver burden quite rapidly decreased. All soft tissues appeared to lose about a fourth to a half their 7 day Es burden by day 24 (Table I), indicating a rapid turnover; much greater than for Pu.

Only about 4% of the Es remained at the intramuscular injection site

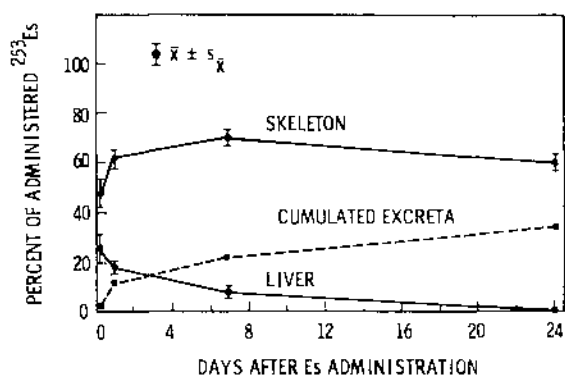


Figure 1. Distribution of  $^{253}\text{Es}$  After Intravenous Injection of  $\text{Es}(\text{NO}_3)_3$  to Rats

by 24 days, whereas 16% remained at the subcutaneous site (Figure 2).

#### $^{253}\text{Es}$ DISTRIBUTION IN RATS 24 DAYS AFTER INJECTION OF $\text{Es}(\text{NO}_3)_3$

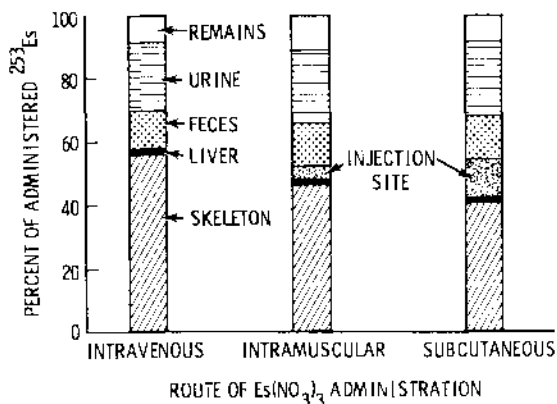


Figure 2.  $^{253}\text{Es}$  Distribution in Rats 24 Days Following the Injection of  $\text{Es}(\text{NO}_3)_3$  by Various Routes

The popliteal lymph nodes were about 6 times higher near the intramuscular injection site than the nodes from the intravenous rats (Table I). Similarly, the axillary nodes near the site of the subcutaneous injection were slightly higher than the corresponding nodes from intravenously injected rats, while the mesenteric lymph nodes for all three routes retained Es in about the same concentrations as other soft tissues. Generally, tissue concentrations were similar for the three routes of administration with the intravenous route having slightly higher retention in the skeleton at 24 days. The amount of Es remaining in the rat was rela-

Table 1.  $^{253}\text{Es}$  Concentration in Tissues After Administration of 4.76  $\mu\text{Ci}$   $\text{Es}(\text{NO}_3)_3$  to Rats

TIME OF MEASUREMENT	PERCENT DOSE/GRAM ( $\bar{x} \pm S_{\bar{x}}$ )			
	7 DAYS	24 DAYS	24 DAYS	24 DAYS
ROUTE	INTRAVENOUS	INTRAVENOUS	INTRAMUSCULAR	SUBCUTANEOUS
TISSUE				
FEMUR*	8.8 $\pm$ 0.4	6.8 $\pm$ 0.2	5.3 $\pm$ 0.5	4.7 $\pm$ 0.3
LIVER	0.88 $\pm$ 0.09	0.16 $\pm$ 0.03	0.11 $\pm$ 0.02	0.11 $\pm$ 0.01
KIDNEY	0.86 $\pm$ 0.05	0.33 $\pm$ 0.05	0.56 $\pm$ 0.08	0.43 $\pm$ 0.05
SPLEEN	0.63 $\pm$ 0.01	0.21 $\pm$ 0.01	0.13 $\pm$ 0.02	0.13 $\pm$ 0.04
LUNGS	0.12 $\pm$ 0.001	0.10 $\pm$ 0.01		
OVARIES	0.15 $\pm$ 0.02	0.074 $\pm$ 0.009	0.062 $\pm$ 0.009	0.032 $\pm$ 0.006
HEART	0.093 $\pm$ 0.002	0.059 $\pm$ 0.009		
MUSCLE	0.012 $\pm$ 0.002	0.0098 $\pm$ 0.0018	0.012 $\pm$ 0.004**	0.006 $\pm$ 0.005**
ADRENALS	0.12 $\pm$ 0.03	0.047 $\pm$ 0.008	0.040 $\pm$ 0.007	0.040 $\pm$ 0.006
THYMUS	0.042 $\pm$ 0.007	0.029 $\pm$ 0.008	0.015 $\pm$ 0.003	0.015 $\pm$ 0.001
LUMPH NODES:				
MESENTERIC	0.12 $\pm$ 0.05	0.066 $\pm$ 0.005	0.058 $\pm$ 0.005	0.029 $\pm$ 0.006
HEPATIC	0.047 $\pm$ 0.008	0.024 $\pm$ 0.005		
SPLENIC	0.12 $\pm$ 0.005	0.055 $\pm$ 0.012		
TRACHEO-BRONCHIAL	0.13 $\pm$ 0.03	0.027 $\pm$ 0.009		
POPLITEAL		0.061 $\pm$ 0.008	0.70 $\pm$ 0.18	0.088 $\pm$ 0.018
AXILLARY		0.048 $\pm$ 0.011		

\*BASED ON ASH WEIGHT. ALL OTHER TISSUES BASED ON WET WEIGHT.

\*\*MUSCLE SAMPLED REMOTE FROM INJECTION SITE.

tively independent of the route of administration taking into account the amount remaining at the injection sites (Figure 2).

Among the bones of the skeleton, Es concentration was greatest in the vertebrae and sternum (Table 2). The lower concentration in the bones of the feet may be influenced by difficulty in completely removing the surrounding tissue, giving high weights and thus, lowering the observed concentration values; however, the distal bones may have a lower concentration than other bones. Dividing the amount of Es present in the entire skeleton by that found in one femur gave a value of 23.7 (95% confidence interval = + 1.6) based on 16 such comparisons. This value did not appear to vary significantly in

Table 2. Concentration of  $^{253}\text{Es}$  in the Skeleton 24 Days After Intravenous Injection of 4.76  $\mu\text{Ci}$   $\text{Es}(\text{NO}_3)_3$  at pH 2

BONE	% DOSE/GRAM ASH WEIGHT $\bar{x} \pm S_{\bar{x}}$
FEMUR	6.8 $\pm$ 0.2
STERNUM	7.8 $\pm$ 0.3
VERTEBRAE	7.9 $\pm$ 0.7
RIBS	6.1 $\pm$ 0.3
PELVIS	6.6 $\pm$ 0.8
FEET	4.3 $\pm$ 0.4
REMAINDER	6.5 $\pm$ 0.5

rats tested at 4 hours to 24 days, or to be affected by the route of administration.

The patterns of  $^{253}\text{Es}$  excretion over the 21 day collection period are

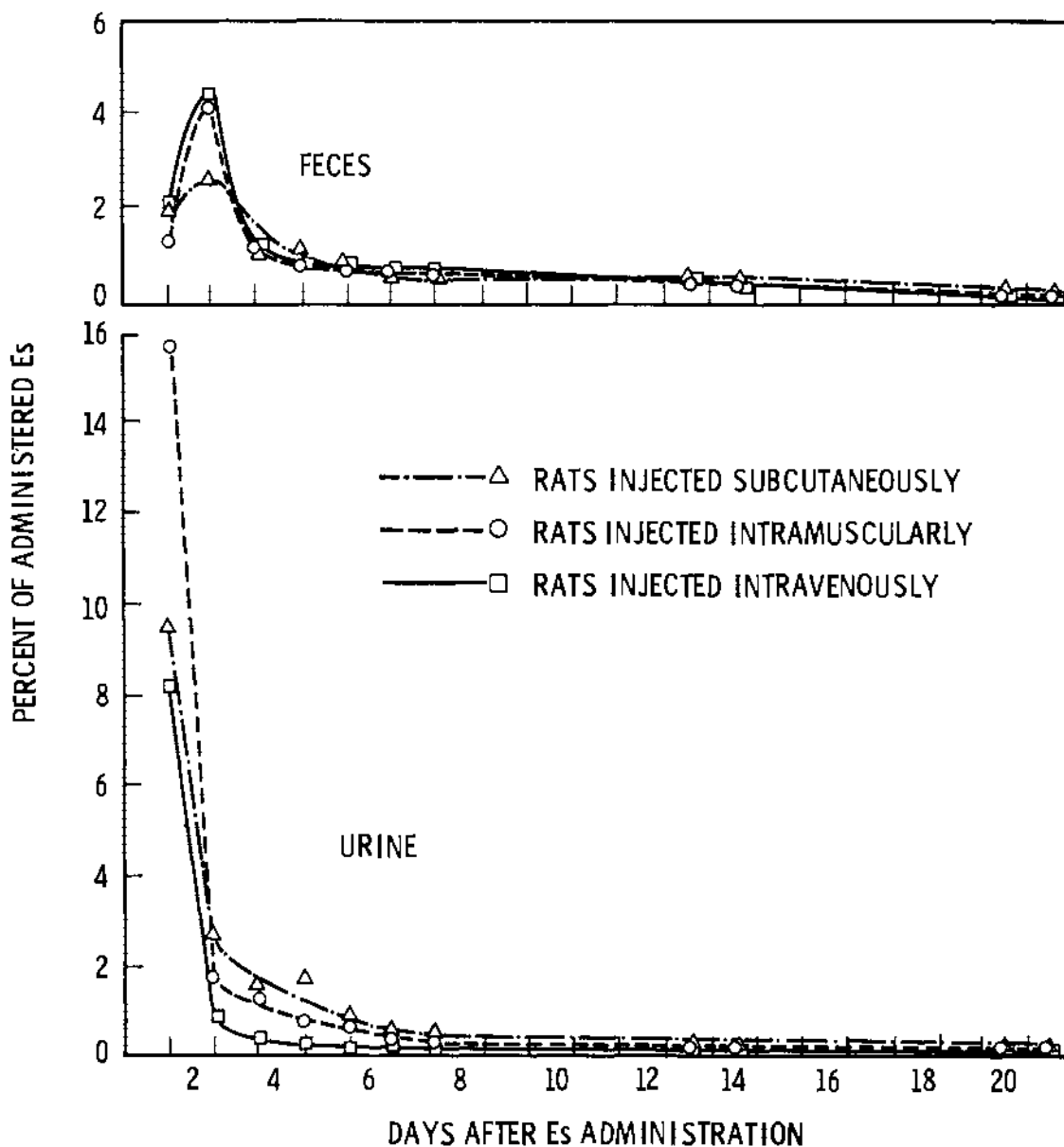


Figure 3. Excretion of  $^{253}\text{Es}$  by Rats After Intravenous, Intramuscular and Subcutaneous Administration of  $\text{Es}(\text{NO}_3)_3$  (Error limits are discussed in the text.)

shown in Figure 3. With few exceptions the range of the means of the two sub-groups about their common mean, represented by the symbol on the figure, was less than 10%. Exceptions were at day 4 of the subcutaneously injected animals where their range was almost  $\pm 20\%$  of the combined value and the first day urine results from the intravenous rats which were  $\pm 78\%$  of the combined mean. The latter was due to poor collection and it is suspected their value should be higher since corresponding literature values

using einsteinium chloride in rats<sup>1</sup> or the citrate in mice<sup>2</sup> show greater than 20% of the dose excreted in urine the first day. The high urinary output of Es following intramuscular injection indicates a rapid translocation from the injection site. After about 1 week, more Es is excreted in the feces than urine but both are low and similar in magnitude by the 21st day. There was more Es found in the 2nd days feces than over the 1st 24 hours' collection but this may be due to generally smaller fecal

output on day 1 than observed in subsequent collections.

The rats receiving Es solution on the skin absorbed nearly 4% of the total available activity, 2.5  $\mu\text{Ci}/\text{cm}^2$ , in 30 minutes. About  $10^{-2}$   $\mu\text{Ci}$  were found in the urine,  $10^{-1}$   $\mu\text{Ci}$  in feces, 7, 4, 2 and  $0.6 \times 10^{-4}$   $\mu\text{Ci}$  in or on the skin, and in the skeleton, liver, and kidneys, respectively. While the skin site had been gently washed to a detectable level of <1-2 cpm using an alpha survey probe, the feces might be high from additional cleaning of the area by the rat and subsequent swallowing, although the nuchal region would not readily lend itself to such cleaning.

#### DISCUSSION

Generally, Es injected as the nitrate was more mobile in soft tissues than actinides with lower Z. For example, Es leaves rat liver initially with a biological half-time of about 5 1/2 days compared to 18 days<sup>3</sup> for Am, and is retained at the intramuscular injection site to the extent of 8% of the dose at 24 days compared to 16%, 37% and 90% retention at 30 days after intramuscular injection with Cf, Cm and <sup>239</sup>Pu nitrate, respectively<sup>4</sup>. These data and the high retention in skeleton and low liver retention support the predictions of Durbin<sup>5</sup> and demonstrate the highly chemical nature of the actinide behavior in the biological milieu. Based on the biological solubility one could predict favorable response to chelation therapy and this is the case<sup>2,7</sup>. There appears to be little difference between the tissue distribution after injecting Es as the nitrate, chloride<sup>1</sup> or citrate<sup>2</sup> which is in contrast to similar salts of Am, Cm or Pu<sup>8</sup>.

Those lymph nodes expected to drain the subcutaneous and intramuscular injection sites showed higher concentrations of Es than similar nodes away from the site. This suggests some Es transport to the local nodes as particulates, although the ultrafiltration tests made on the injection solutions, even a few hours after the rats were injected, showed almost complete solubility. Einsteinium, with a

similar ionic radius and biological distribution in rats, resembles the rare earth, terbium, in its behavior<sup>5</sup>. Apparently it is less subject to forming particulate hydroxides *in vivo*, or to altering its properties through valence state changes, than are Pu and to a lesser extent Am and Cm.

#### CONCLUSIONS

Es behaves biologically like a lanthanide and is more mobile and soluble *in vivo* than Pu, Am, and Cm. It moves rapidly from intramuscular deposits of its nitrate, mainly into bone, where it is slowly released with a biological half-time of >40 days. This is still twice the physical decay rate so that the bone receives most of the available radiation. In contrast, about half the Es initially deposited in the soft tissues is lost from the tissues before having a chance to decay.

#### ACKNOWLEDGEMENTS

The author wishes to thank K. E. McDonald, D. W. Baxter, and J. W. Barnett for excellent technical assistance.

#### REFERENCES

1. F. P. Hungate, J. E. Ballou, D. D. Mahlum, M. Kashima, V. H. Smith, C. L. Sanders, D. W. Baxter, M. R. Sikov, and R. C. Thompson. Preliminary data on <sup>253</sup>Es and <sup>249</sup>Bk metabolism in rats. *Health Phys.* 22: 653-656 (1972).
2. H. G. Parker, S. R. Wright, A. de G. Low-Beer, and D. J. Yaeger. The metabolism of <sup>253</sup>Es in mice. *Health Phys.* 22: 647-651 (1972).
3. V. S. Stepanov and G. A. Zalikin. Distribution and kinetics of the elimination of americium-241 from the rat organism. *Radiobiology USSR* 10: 240 (1970) (AEC-tr-7154).
4. M. Morin, J.-C. Nenot, and J. Lafuma. Comparison du devenir biologique du californium et de celui d'autres transuraniens. *Compt. Rend. Acad. Sci., Series D.*, 276: 1209-1211 (1973).
5. P. W. Durbin. Distribution of the transuranic elements in mammals. *Health Phys.* 8: 665-671 (1962).

6. J. A. Mewhinney, P. L. Ziemer, and R. R. Landoit. Retention and distribution of injected californium-252 in the rat. Health Phys. 21: 857-859 (1971).
7. V. H. Smith. Therapeutic removal of internally-deposited transuranium elements. Health Phys. 22: 765-778 (1972).
8. International Commission on Radiological Protection. The Metabolism of Compounds of Plutonium and Other Actinides. ICRP Publ. 19, Table 6.1. Pergamon Press, New York, 1972.

RETENTION AND ELIMINATION OF BERKELIUM-249-CALIFORNIUM-249  
FOLLOWING ACUTE ACCIDENTAL INHALATION

J. Rundo and J. Sedlet  
Argonne National Laboratory  
Argonne, Illinois 60439

Abstract

A case of accidental inhalation of a small quantity of an ignited mixture of  $^{249}\text{Bk}$  and its decay product,  $^{249}\text{Cf}$ , was studied by body radioactivity measurements and excretion analyses during the first year after intake. The initial chest content of about 3 nCi of  $^{249}\text{Cf}$  declined according to a 2-component exponential function of time (half-lives of 25 days and 1210 days). Except for an initial rapid clearance via the feces, the urinary and fecal excretion rates of both nuclides increased with time until 60-70 days after intake, and then declined. If the early fecal excretion was neglected, the results could be described as the difference between two exponential components with half-times of 15-30 days and 90-130 days.

Introduction

This paper presents the results of the first year's study of an inhalation intake of  $^{249}\text{Bk}$  and  $^{249}\text{Cf}$  by a chemist. Some of the mixture, ignited on a tantalum disc, became airborne, as evidenced by the early detection of extensive surface contamination in the area. Fecal and urinary collections began immediately, and body radioactivity measurements were begun one week later. The circumstances of the incident strongly suggested an inhalation exposure. A sample of the contamination was used to prepare standards for the body radioactivity and excretion measurements. The Bk/Cf ratio in this sample was assumed to be the same as in the inhaled material. Berkelium-249 decays with a 314 day half-life, principally by emitting a beta particle with an end-point energy of 0.125 MeV, to  $^{249}\text{Cf}$ , an alpha emitter whose principal particles have energies of about 5.8 MeV. The californium isotope also emits 333 and 388 keV gamma-rays in 16 and 72% of its decays, respectively.<sup>1</sup>

This case contains some unusual and interesting features. There is little reported information on the behavior of californium in the body. Since the active material in this incident was ignited before intake, it was presumably highly insoluble, and the behavior of insoluble forms of the actinides in the body is of considerable interest because of the potential widespread use of these elements. Finally, the parent-daughter relationship of the two nuclides, coupled with the long half-life of the daughter, presents some interesting problems in data interpretation.

Experimental Techniques

Investigations of the radioactivity in the subject were made in a steel room in an underground laboratory.<sup>2</sup> Measurements were made with two large NaI(Tl) detectors, positioned above and below the supine subject and placed alternately at the levels of the mid-sternum and of the lower abdomen. All the spectra observed at the former position showed a peak due to  $^{249}\text{Cf}$  at 0.39 MeV while those at the latter did not. The spectra measured with the counter



in the abdominal region were used, after appropriate normalization, as "backgrounds" for the spectra from the chest region. For calibration measurements a standardized source of  $^{249}\text{Cf}$  was placed in turn in each of the six spaces of a lung phantom similar to that devised by Miller,<sup>3</sup> and the gamma-ray spectra were summed to give an average spectrum from a simulated distributed source. For the energy band 295-485 keV a calibration factor of 5.61 pCi/cpm was obtained when the responses from both detectors were combined.

An acid solution of the contaminant was standardized by counting in calibrated alpha and beta particle counters. The Bk/Cf activity ratio in the contaminant, measured on day 2, was 306 and the ratio at other times was calculated. The ratio of the half-lives, 418 for Cf/Bk, is similar to the initial activity ratio (Bk/Cf), so that the complete decay of the original  $^{249}\text{Bk}$  will nearly double the  $^{249}\text{Cf}$  activity. Samples of urine and feces were analyzed by specially developed radiochemical techniques and simultaneous liquid scintillation counting of both nuclides. The separation of californium from the natural thorium in the feces was based on differences in their distribution between aqueous solution and di(2-ethylhexyl)phosphoric acid. Details of the techniques will be described elsewhere.

### Results and Discussion

The excretion results are plotted in Figures 1 and 2. The  $^{249}\text{Bk}$  data have

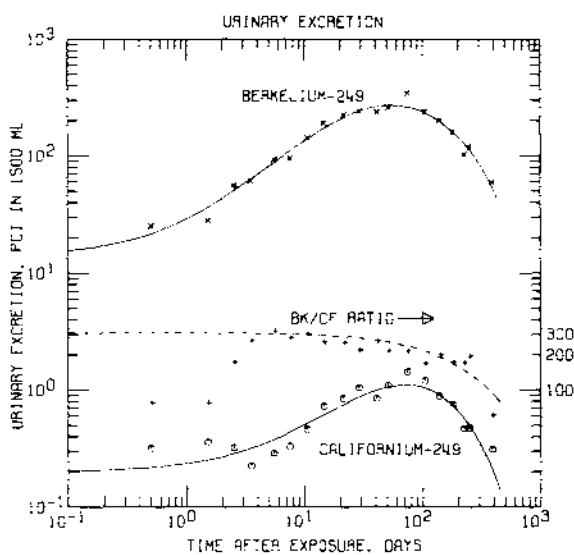


Figure 1.

Urinary excretion of the two nuclides (x and o) and the curves fitted by least-squares analysis. The dashed curve shows the predicted behavior of the  $^{249}\text{Bk}/^{249}\text{Cf}$  activity ratio in the contaminant; the measured ratios of the activities in urine are shown as points (+) for comparison.

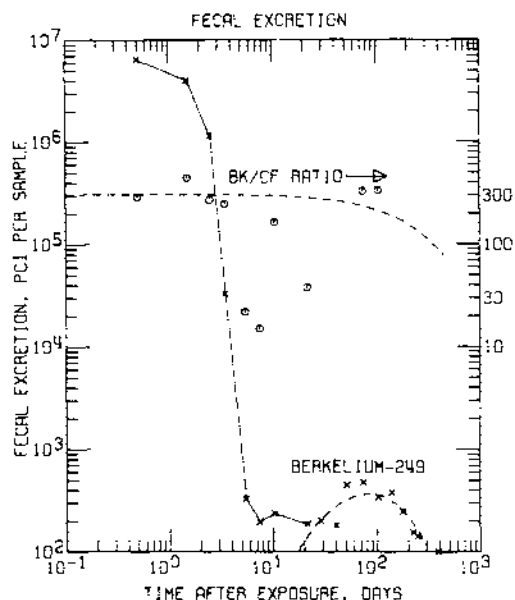


Figure 2.

Fecal excretion of  $^{249}\text{Bk}$  (x); the lower dashed curve is the result of a least-squares fit to the points from 21.5 days onwards. The calculated  $^{249}\text{Bk}/^{249}\text{Cf}$  activity ratio in the contaminant is plotted as in Fig. 1, and the ratios of the activities in feces are included (o).

been corrected for radioactive decay from the time of exposure. The  $^{249}\text{Cf}$  data have been corrected for the growth of  $^{249}\text{Cf}$  from  $^{249}\text{Bk}$  in vivo with the assumption that the newly-formed  $^{249}\text{Cf}$  was excreted at the same rate as that inhaled. In the interpretation of the results, a urine volume of 1500 ml and a single fecal sample were assumed to comprise the daily elimination. The analyses of the fecal samples for  $^{249}\text{Cf}$  have not been completed, but the elimination pattern shown by the available data is similar to that of  $^{249}\text{Bk}$ . The excretion rate in the feces decreased during the first week with a half-time of about 0.5 day. After about 10 days, an increase in the fecal excretion rate similar to that found for urine became apparent. The curves through the excretion data are weighted least-squares fits of the difference between two exponential terms, representing the initial increase and the subsequent decrease in excretion rates. The data for the fecal excretion of  $^{249}\text{Bk}$  for the first 21 days were ignored in making the least-squares analysis. The parameters of the excretion equation,

$$E(\text{pCi}/1500 \text{ ml or pCi/sample}) = -C_1 \exp\left(-\frac{0.693}{T_1}t\right) + C_2 \exp\left(-\frac{0.693}{T_2}t\right) \quad (1)$$

are given in Table I. The equations represent the data reasonably well, and within the statistical errors the  $C_1/C_2$  ratios are all unity as required for zero excretion at zero time. The same processes appear to control the excretion of both nuclides. This is reassuring although not particularly surprising, since the nuclides were inhaled in the same form and have similar chemical properties.

TABLE I. Excretion Equation Parameters From Least-Squares Fitting

	$C_1$	$T_1$	$C_2$	$T_2$
$^{249}\text{Bk}$ - urine	$396 \pm 40$	$16 \pm 2$	$410 \pm 42$	$132 \pm 12$
$^{249}\text{Bk}$ - feces	$1255 \pm 300$	$33 \pm 7$	$1100 \pm 340$	$91 \pm 13$
$^{249}\text{Cf}$ - urine	$2.2 \pm 1.0$	$30 \pm 10$	$2.4 \pm 1.0$	$109 \pm 36$

A plot of  $^{249}\text{Cf}$  chest content against time suggested a two-component exponential function and a non-linear, weighted, least-squares fit was made of such a function. The results are shown in the upper part of Figure 3, where the smooth curve drawn through the points has the equation:

$$\text{Content (pCi)} = 480 \exp\left(-\frac{0.693}{25}t\right) + 2350 \exp\left(-\frac{0.693}{1210}t\right).$$

If this were a true representation of the retention in the chest, its first derivative (with a negative sign) would describe the excretion rate, and in the lower part of Figure 3 this is plotted as the broken curve. The points along the curve are the measured total  $^{249}\text{Cf}$  excretion (uncorrected for growth) on those days for which measurements are complete. After the early massive fecal clearance is complete, there is respectable agreement between the predicted and observed excretion rates. However, additional data may indicate that the agreement is due partly to a fortuitous combination of different half-times that result in agreement over a limited time interval.

This analysis may be an over-simplification because of the growth of new  $^{249}\text{Cf}$ . By the time of the latest measurement (day 388) an additional 1.25 nCi of  $^{249}\text{Cf}$  would have been formed and the question, as yet unanswered, arises as to what extent this remained in the chest or was excreted preferentially. The initial chest content of 2.8 nCi  $^{249}\text{Cf}$  must have been accompanied by 870 nCi of  $^{249}\text{Bk}$ . The stability of the ratio of the counting rates from the front and back was studied. The mean value was 1.90, and the standard deviation (0.16)

was similar to the statistical standard errors of the individual ratios (0.12 to 0.17). This suggested that the ratio was constant (within statistics), and it indicated that there was no major change in the distribution of  $^{249}\text{Cf}$  in the chest during the first year.

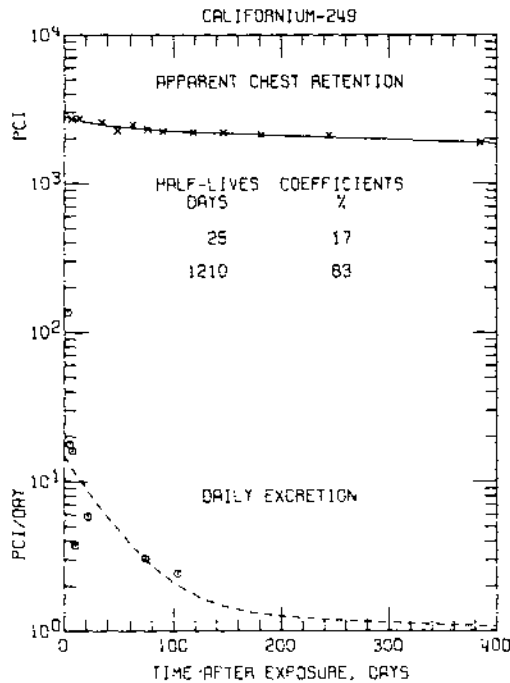


Figure 3.

Apparent chest retention (x) of the  $^{249}\text{Cf}$ , based on external gamma-ray counting; the regression equation is drawn as a smooth curve, and its first derivative as a dashed curve. For comparison with the latter, observed total excretion data are shown (o).

From the Bk/Cf ratios in Figures 1 and 2, some differences can be seen in the excretion behavior of the two nuclides. An excretion mechanism favoring  $^{249}\text{Cf}$  is apparent only in the first three urine samples, whereas the fecal excretion appears to favor  $^{249}\text{Cf}$  for a period from 4 days on, but not at about 100 days. The data are insufficient as yet to establish a pattern.

Integration of the excretion equations gives those portions of the body content to be excreted with the biological half-times thus far evident. These values, calculated with the assumption that the  $^{249}\text{Cf}$  fecal elimination was proportional to its urinary excretion, were equivalent to 25% of the measured  $^{249}\text{Cf}$  burden (2.8 nCi) and 20% of the calculated  $^{249}\text{Bk}$  burden (870 nCi). These appear to be reasonable fractions of an inhaled, insoluble material to be eliminated with half-times as short as 90-130 days.

The excretion pattern is consistent with the interpretation that much of the activity was taken into the body in an "unexcretable" form and, with the early fecal clearance neglected, this form required conversion to a second form before it could be excreted. If both the conversion and the excretion are exponential with rate constants,  $\lambda_1$  and  $\lambda_2$ , respectively, then the observed excretion rate, E, will be:

$$E = \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} A_0 \exp(-\lambda_1 t) - \left( \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} A_0 - B_0 \right) \exp(-\lambda_2 t), \quad (2)$$

where  $A_0$  and  $B_0$  are the amounts of "unexcretable" and "excretable" material inhaled, respectively. The equation is analogous to that which describes two successive radioactive decays. The model is similar to that proposed by Healy<sup>4</sup> for the excretion of insoluble plutonium from the lung, and it differs from his only in that he used a power function to describe the excretion portion

rather than an exponential function. Since equation (2) is in the same form as equation (1), the parameters  $A_0$  and  $B_0$  can be evaluated from the results in Table I. The results obtained with the assumptions that  $B_0$  was alternatively zero or non-zero are given in Table II. The amount presumably inhaled in "excretable" form ( $B_0$ ) was small compared to  $A_0$ , and within statistics, may be taken as zero. Although the validity of the model is not established, it does account for the increase in excretion rate with time. Similar increases after inhalation of insoluble particulate plutonium have been observed by several workers,<sup>5,6</sup> and were attributed by them to the low solubility of the plutonium compound.

TABLE II. Evaluation of Excretion Parameters

	$B_0 = 0$	$B_0 \neq 0$	
	$A_0$	$A_0$	$B_0$
<sup>249</sup> Bk - urine	67 nCi	66 nCi	0.014 nCi
<sup>249</sup> Bk - feces	106 nCi	113 nCi	-0.16 nCi
<sup>249</sup> Cf - urine	0.26 nCi	0.25 nCi	$2 \times 10^{-4}$ nCi

Evidence for the insolubility of the inhaled contaminant was obtained from the chemical behavior of the material excreted in the feces. In the first few fecal samples, 25% to 70% of the activity could not be readily dissolved. This percentage decreased to 2% or less in the samples collected after day 10. Further, the activity in solutions of the early fecal samples was initially only poorly extracted, although it could be coprecipitated with non-isotopic carriers. At the same time, tracer solution added to aliquots of such samples was efficiently extracted. The activity could be rendered extractable by vigorous chemical treatment. Subsequent fecal samples contained activity that, based on its chemical behavior, had undergone a change to a more soluble form.

#### References

- 1 Lederer, C. M., Hollander, J. M., and Perlman, I., Table of Isotopes, John Wiley and Sons, Inc., New York (1968).
- 2 Rundo, J., Keane, A. T., and May, H. A., "Low-Background Counting Rooms," in ANL-7860, Part II, pp. 257-263 (1971).
- 3 Miller, C. E., "The Human Spectrometer," in ANL-5829, pp. 144-166 (1958).
- 4 Healy, J. W., Am. Ind. Hyg. Assoc. Quarterly, **18**, 261 (1957).
- 5 Campbell, E., Moss, W. D., Schulte, H. F., and Kressin, I. K., "Urine and Fecal Excretion Following an Inhalation Exposure," (Abstract), in ANL-8014, p. 21 (1972).
- 6 Wood, Jr., W. R. and Sheehan, W. E., Am. Ind. Hyg. Assoc. J., **32**, 58 (1971).

This work was performed under the auspices of the U. S. Atomic Energy Commission.

TOXICOLOGY OF TRANSPORTABLE URANIUM:  
A CRITICAL CONTRIBUTION TO HEALTH PROTECTION OF WORKERS

F. Breuer

CNEN, Division of Health and Controls, Rome, Italy

E. Righi

CNEN, Medical Service

of CNEN National Laboratories, Frascati (Rome), Italy

SUMMARY

The chemical toxicity of uranium has a remarkable importance in the evaluation of the risk of internal contamination due to transportable compounds of such element. The risk becomes most relevant when one has to deal with natural uranium or long-lived uranium isotopes. The maximum permissible concentrations recommended by ICRP and the other limit values proposed are based on the results of experiments that, though complete and valid in their general outline, are affected by the limited technical possibilities available when effected. Since now the possibilities of detecting and evidencing renal lesions are remarkably increased; it could be useful to review the problem of the chemical toxicity of uranium both on the basis of the advanced techniques available and on the experience gained in the field of health protection in nuclear environments. In fact modern techniques allow a relatively easy detection of even slight modifications and of their characteristics; at the same time they permit to carry out the analysis of very small biological samples. In the present work it is also stressed that studies carried out on other renal toxic substances, like cadmium, are giving very interesting results.

The occupational exposure of workers to the hazards of radioactive contamination involves health protection problems which still deserve further studies and research. It is well known that since the first definition of protection criteria suggested by international and national qualified organisms, several variations and advances have been introduced in the successively published recommendations<sup>1,2,3,4,5,6</sup>.

Among the different nuclides taken into consideration to

control the risk of internal contamination, particular attention must be given to natural uranium and to long-life isotopes of this element: in fact their incorporation in man and higher animals is remarkably important as far as their chemical toxicity is concerned. Such a risk becomes of primary importance when we have to deal with those compounds of U-235, U-238 and natural uranium which are easily absorbed<sup>3,4,6</sup>. These toxicological assumptions represent, therefore, the basic reference for the definition of limit levels in radiation protection: their recognition is the result of a great bulk of studies and experiments considering both the biochemical and the morphological aspects<sup>7</sup>.

However, it should be observed that, despite the precision and the extension of such investigations, not negligible perplexities still exist on the actual physiopathological significance of the proposed limit levels. Just to quote an example, it is rather difficult to evaluate, on a histological level, findings described as "slight or extremely mild renal-tubular changes"<sup>7</sup>. Similarly, it is sometimes far from easy to use the available protection criteria to solve real problems, such as the correct evaluation of a possible renal damage in the case of a continuous contamination when the incorporation rate widely varies in time<sup>8</sup> and consequently to give in this case a right and reliable interpretation of the excretion data.

The problem is worth while a more modern evaluation made on the basis of recent knowledge in the field of renal physiology and physiopathology. First of all it is necessary to take into consideration one of the most sensitive indicator of uranium intoxication, "albuminuria", which, together with catalasuria and aminoaciduria, permits the detection of an early renal failure (in the form of tubular lesions) due to the introduction of such heavy metal in a soluble form<sup>7,10</sup>.

Albuminuria has practically been considered one of the most important reference symptoms, that is to say a kind of "threshold" manifestation used in the definition of protection parameters. Now, the modern knowledge on the structure and origin of urinary proteins, as well as on the relationships existing between the different types of proteinuria and the morphological and functional nephropathic modifications, brings about a series of complex problems of interpretation emerging from analyses such as the electrophoresis, the immunoelectrophoresis, gel-filtration, ultracentrifugation, chromatography, immunodiffusion, etc.<sup>11</sup>.

Obviously the simple quantitative evaluation of "albuminuria" in many cases results nowadays semeiologically inadequate. For instance in chronic pielonephritis, a quantitative description of proteinuria, often not too high, does not always describe a severe renal damage<sup>12</sup>. Consequently the importance of "albuminuria" on a physiopathogenetic level appears to be decreased: an evaluation must be made in quantitative and qualitative terms of "pro-

teinuria", or, better, of "proteinurias". More recently, in fact, on the basis of their electrophoretic behaviour, the fractions forming the "proteinuria" have been accurately detected and a morphological distinction has been made possible as far as "pathologic proteinurias" are concerned, in "glomerular" and "tubular" proteinurias<sup>11,13,14,15,16,17</sup>.

The electrophoretic behaviour of proteinurias in the different possible combinations has often shown to be remarkably consistent with the different pathogenetic and clinical phases of several nephropathies. The observation of an electrophoretic picture indicating a renal damage, confirmed at the biopsy, in the presence of very poor, or even not detectable, urinary proteins, shows the great possibilities given by modern methods.

We did not find in the current specialized literature studies of this kind, related to the characterization of proteinuria caused by uranium. However, it might be interesting to try to outline some diagnostic possibilities even if on a theoretical basis.

Taking into account the capacity of soluble natural uranium of causing lesions to proximal tubules, the hypothesis that the "expected" proteinuria might essentially be of a tubular kind (simple or mixed) appears to be justifiable.

Though considering the incertitudes connected to the origin and formation processes of the tubular proteinuria, the latter presents morphological and structural features which allow a rather exact differentiation: it is made by micromolecular aggregates (molecular weight: 20,000 - 50,000) detectable by means of an ultracentrifuge; it often shows an anomalous behaviour in the presence of precipitating factors, boiling included; its content in urines is generally poor and sometimes not even detectable<sup>11</sup>. Consequently diagnostic difficulties may arise owing both to the commonly low values and to the irregular reactivity to precipitation also when the content is high.

However, these difficulties can be overcome by the electrophoretic examination (on cellulose acetate or on gels of acrylamide or of starch). In fact the morphological picture permits a satisfactory characterization also when the quantitative determination, carried out with methods commonly used in clinical research, gives scarce results or no results at all. The electrophoretic morphology of tubular proteinuria is characterized (on cellulose acetate) by the prevalence of globulins as compared to albumin; by a low intensity of the transferrinic fraction; and by the presence of homogeneous bands with differing mobility in the gamma zone.

The "glomerular" proteinuria presents a rather different picture; the albumin fraction is remarkably wide and intense, and the transferrinic band is very sharp. Obviously mixed forms also exist and are often encountered; they appear as the result of

various overlappings of the pure forms.

The first studies on tubular proteinurias have been carried out by Butler and Flynn in 1958<sup>13</sup>. But we would like to stress that at the same time Luessenhop and coll. established the minimum dose of hexavalent uranium able to cause, when injected intravenously, albuminuria in man<sup>10</sup>.

The evaluation criteria used in this study to detect the "albuminuric threshold" were based on methods which are no more adequate to give an evaluation of the kidney functionality, especially at the level of the "intravenous tolerance dose of hexavalent uranium for man"<sup>10</sup>. It is also necessary to observe that a correct interpretation of this symptom is sometimes made more difficult by the frequent presence of proteinuria in patients affected by neoplasia<sup>18</sup>, as the examined subjects actually were<sup>10</sup>.

In 1959 Publication 2 of ICRP<sup>3</sup> reported, among others, the MPC in air and water for natural uranium, U-235, and U-238 in soluble form. These values, already contained in the 1954 "Recommendations"<sup>2</sup> are still substantially unchanged. In fact the modifications appearing in Publication 6 for the value of MPC in water derive from the different hypothesis accepted for intestinal absorption of uranium<sup>4</sup>; while the consideration of its ability in producing renal lesions remained the same. In any case, it should be observed that the MPC values for soluble uranium have been subjected to some criticism<sup>19</sup> since their very first appearance.

In urines of experimental animals intoxicated with hexavalent uranium, the presence of plasma-albumin and of various plasma-globulins was shown by electrophoresis (on paper?)<sup>7</sup>. It is interesting to compare these results to those obtained in the case of tubular proteinuria.

In fact, the tubular proteinuria (in its pure form) shows on paper electrophoresis an albumin/globulins ratio lower than 1, and the presence of a fraction alpha-2 of a remarkable entity, a fraction beta relatively high and, in some cases, a fraction with anomalous mobility in the cathodic sense (post-gamma). By means of electrophoresis on starch gel the globulinic fractions show a pre- and post-albuminic, alpha-2, beta and post-gamma mobility<sup>11,14,16,17</sup> (we already described the electrophoretic results on cellulose acetate).

This topic shows many difficulties in any case. Several questions have been posed, since the first experiments, concerning the passage of plasmatic albumin into urines, especially in the frequent absence of appreciable glomerular lesions in the case of small doses of uranium<sup>7,10</sup>. On the other hand the genesis of tubular proteinuria involves many aspects which are still to be explained<sup>20,21,22</sup>.



It is also very interesting to compare the preceding observations with analogous aspects supplied by toxicological studies of another heavy metal, cadmium, also responsible of renal lesions mainly at the tubular level. The proteinuria evidenced by Friberg in 1950 in workers exposed to cadmium dusts and vapours<sup>23</sup> was deeply studied; the characters distinguishing it from proteinurias found in nephritis and nephroses<sup>24</sup> are now well known.

The cadmic protein (molecular weight ranging from 20,000 to 30,000; sedimentation constant of about 2.2 S) does not precipitate when boiling, or when treated with picric acid; it is evidenced by a precipitation with nitric acid, with 25% trichloroacetic acid, with 3% sulfosalicylic acid. The electrophoretic spectrum includes four or five fractions with a low albumin/globulins ratio; 50% approximately of the components behave as gamma-globulins in electrophoresis, immunoelectrophoresis, and DEAE-cellulose chromatography.

By means of specific antisera it was possible to demonstrate that about 30% of cadmium proteins is constituted by L chains of gamma-globulins<sup>25</sup>.

It could be wise to recall here that also recently the difficulty of evidencing the peculiar micromolecular proteinuria of subjects exposed to cadmium by means of conventional methods has been stressed; while electrophoretic methods permit to recognize such proteinuria with reliable results<sup>26</sup>.

The reported aspects of cadmium toxicology are intended as an example showing the difficulties and complexity of protection studies and demonstrate that these kind of problems do not exclusively concern uranium.

The observations made on the electrophoretic behaviour of proteinurias, represent a partial view of the problem, but they are in any case sufficient, according to our opinion<sup>27</sup>, to select the importance of some controls not only at the damage level, but rather at the level of "contact" of uranium with the kidney. On the other hand, the concept of threshold is being critically reviewed, as evidenced during the Symposium of the American Academies of Industrial Hygiene and Occupational Medicine held in Cincinnati in February 1970<sup>28</sup>.

The definition of dose without effect and of acceptable reaction advanced on that occasion undoubtedly involves aspects of biological philosophy extremely delicate and, in any case, the necessity of having investigation means of high sensitivity and specificity.

Summing up, on the basis of recent knowledge in the field of physiopathology and diagnostics a check appears to be auspicious of the validity of the values established for uranium years ago and still used: in order to confirm them or, if necessary, to

adequate them to more modern criteria. On the other hand, the methods available today could permit to carry out an adequate observation in man too, in the range of the present "admissibility", and at very low levels without involving undue risks.

As far as, in particular, the electrophoretic multifractionation of urinary proteins is concerned, the present availability of techniques which can be carried out also on not concentrated urines<sup>29</sup> can permit wider experiments on small animals, such as rats for which it has already been established that the lethal dose for uranium relatively to the body weight is well comparable to that for man<sup>8,10</sup>.

The aim of these observations will be reached if the above discussions show the necessary requirements for a translation into a useful research program.

#### REFERENCES

1. Maximum permissible amounts of radioisotopes in the human body and maximum permissible concentrations in air and water. USA National Bureau of Standard, Handbook 52, 1953.
2. Recommendations of the International Commission on Radiological Protection (Revised December 1, 1954). Brit.J. Radiol., Supplementum n. 6, London, 1955.
3. Report of Committee II on permissible dose for internal radiation. ICRP Publication 2, Pergamon Press, New York, 1959.
4. Recommendations of International Commission on Radiological Protection. ICRP Publication 6, Pergamon Press, Oxford, 1964.
5. Recommendations of International Commission on Radiological Protection. ICRP Publication 9, Pergamon Press, Oxford, 1966.
6. Report of Committee 4 on evaluation of radiation doses to body tissues from internal contamination due to occupational exposure. ICRP Publication 10, Pergamon Press, Oxford, 1968.
7. Voegtlin C., Hodge H.C., eds.: Pharmacology and toxicology of uranium compounds. Books 1 to 4. McGraw-Hill Book Company, Inc., New York, 1949-1953.
8. Eve I.S.: Some suggested maximum permissible single intakes of uranium. Health Physics, 10, 773, 1964.
9. Sunderman F.W., Sunderman F.W.Jr., eds.: Laboratory diagnosis of kidney diseases. Warren H. Green, Inc., St. Louis, Missouri, 1970.
10. Luessenhop A.J. et al.: The toxicity in man of hexavalent uranium following intravenous administration. Amer. J. Roentgenol., 79, 83, 1958.
11. Maiorca R., Scarpioni L.: Le proteinurie. Fisiopatologia e diagnostica. Il Pensiero Scientifico editore, Roma, 1968.
12. Lenti G., Vercellone A.: La pielonefrite cronica. Aggiornamenti clinicoterapeutici. Ed. Minerva Medica, 1960.

13. Butler E.A., Flynn F.V.: The proteinuria of renal tubular disorders. *Lancet*, 2, 978, 1958.
14. Butler E.A., Flynn F.V.: The occurrence of post-gamma protein in urine; a new protein abnormality. *J.Clin.Path.*, 14, 172, 1961.
15. Traeger J. et al.: Analyse électrophorétique des protéinuries des glomérulonéphrites et des syndromes néphrotiques. *J.Urol. Néphrol. (Paris)*, 71, 1056, 1965.
16. Traeger J. et al.: Analyse électrophorétique des protéinuries rencontrées au cours des atteintes tubulaires rénales. *J.Urol. Néphrol. (Paris)*, 71, 728, 1965.
17. Traeger J. et al.: Les protéinuries des atteintes tubulaires rénales. *Path.Biol.*, 14, 5, 1966.
18. Leyton G.R. et al.: Chemical studies and physical characteristics of urinary proteoses in cancer. *Clin.Chim.Acta*, 9, 157, 1964.
19. Hursh J.B.: Suggested urinary tolerance levels for enriched uranium. UR-515, 1957.
20. Butler E.A. et al.: A study of urine proteins by two-dimensional electrophoresis with special reference to the proteinuria of renal tubular disorders. *Clin. Chim.Acta*, 7, 34, 1962.
21. Creeth J.M. et al.: An ultracentrifuge study of urine proteins with particular reference to the proteinuria of renal tubular disorders. *Clin. Chim. Acta*, 8, 406, 1963.
22. Harrison J.F., Northam B.E.: Low molecular weight urine protein investigated by gel filtration. *Clin. Chim.Acta*, 14, 679, 1966.
23. Friberg L.: Health hazards in the manufacturing of alkaline accumulators with special reference to chronic cadmium poisoning. *Acta Med. Scand.* 138, suppl. 240, 1, 1950.
24. Patty F.A. ed.: *Industrial hygiene and toxicology*. Cap.XXVII, Interscience Publishers, Inc., New York, 1967.
25. Vigliani E.C., Pernis B., Amante L.: Etudes biochimiques et immunologiques sur la nature de la protéinurie cadmique. *La Med.Lavoro*, 57, 321, 1966.
26. Pujol M. et al.: Tubulopathie des intoxications chroniques par le cadmium. *Arch. Mal. Prof.*, 31, 637, 1970.
27. Righi E., Arzano S.: Il protidogramma elettroforetico su urine non concentrate nella valutazione della idoneità al lavoro con rischio di contaminazione radioattiva. *CNEN Report LNF-72/82*, Frascati (Rome), 1972.
28. Hatch T.F.: Thresholds: do they exist? *Arch. Environ. Health*, 22, 687, 1971.
29. Brancaccio D.: Multifrazionamento elettroforetico di proteine urinarie non concentrate. *Arch. Ital. Urol. Nefrol.*, 42, 315, 1969.

# THE EFFECT OF NATURAL PHYSIOLOGICAL STRESSES AND ARTIFICIAL HORMONAL STRESSES ON THE RETENTION OF AMERICIUM AND PLUTONIUM BY RAT BONE

J.G. Hollins, M.C. Storr, A. Durakovic  
National Research Council of Canada  
Ottawa, Canada, K1A 0R6

## Abstract

The retention of americium and plutonium by rat bone varies with age and sex, and during pregnancy and lactation. While the calcium content of bone was substantially reduced by lactation, the retention of americium and plutonium was scarcely affected. When the food consumption of lactating rats was restricted to that of control virgin rats fed ad libitum, the retention of americium by the bone of the lactating mothers was greater than that of the controls. The effect of parenteral administration of parathyroid hormone and a restricted dietary intake of calcium was also to increase the retention of americium at a time when bone calcium was being reduced. In contrast, the retention of americium by rat bone during the intense anabolic period of a mother immediately after lactation was less than that of the controls. The retention of americium and plutonium by bone was also reduced after either a series of intraperitoneal injections of calcium gluconate or parathyroidectomy.

## Introduction

The relation between the incorporation of radionuclides by the skeleton and the induction of neoplastic processes in trabecular bone is well known. It has led to many efforts to reduce the burden of that important class of osteotropic elements, the actinides, principally by means of chelating agents. The work reported here provides an alternative approach to this problem by studying the retention of americium and plutonium by bone during periods of anabolic and catabolic activity in the skeleton. We have examined the effects of age, sex, pregnancy and lactation. Having established some variations of the retention of americium and plutonium with these natural stresses, we continued with investigations of the effects of the artificial stresses of parenteral administration of parathyroid hormone, calcitonin, estrogen, and calcium gluconate, a diet free of calcium, and parathyroidectomy.

## Experimental

The animals used in these experiments were specific-pathogen-free rats of the Sprague-Dawley strain bred in our laboratories. The rats were given standard laboratory food, unless otherwise indicated, and tap water ad libitum. Americium-241 and plutonium-239 were administered by intravenous injection. A sublingual vein of a rat anesthetized lightly with Halothane was used. Americium and plutonium were administered as 1% citrate solutions at pH7. The injection volume of 0.1 ml contained 50 nCi of Am-241 or Pu-239. The radionuclides were supplied by the Radiochemical Centre, Amersham.

The radioactivity of rats and bones was measured by counting the 60 keV gamma ray of Am-241 with sodium iodide detectors. Plutonium in bones was measured by liquid scintillation counting after extraction with di (2 ethyl hexyl) phosphoric acid, following the method of Keough and Powers<sup>1</sup>.

## Results

### Age and sex

The retention of americium and plutonium by the whole skeleton and femora of growing and mature female rats one week after injection is presented in Table 1. The retention of americium is greater in the younger rats, while the retention of plutonium is greater in the older rats. In Table 2 the variation with sex of the retention of americium and plutonium by femora of 15 week old rats is presented. The skeletal retention of americium was higher in male than in female rats in a series of experiments with rats of four ages<sup>2</sup>. This is illustrated in Table 2 for rats of 15 weeks and results for plutonium are added. Although the males retained significantly more americium in their bones than the females ( $P = 0.01$ ), the retention of plutonium by bone showed very little dependence on sex, with a slightly higher retention by females than by males ( $P = 0.3$ ).

Table 1. Retention of americium and plutonium 7 days after injection.  
% of injected dose.\*

		Age at time of injection	
		5 weeks	15 weeks
<u>Skeleton</u>	Am	30.6 ± 0.5	20.8 ± 0.4
	Pu	62.6 ± 2.1	77.1 ± 3.0
<u>Femora</u>	Am	3.0 ± 0.2	2.0 ± 0.2
	Pu	7.3 ± 0.5	7.7 ± 0.5

\* all results presented in these tables are mean values ± standard errors of the means for at least 5 rats.

Table 2. Retention of americium and plutonium by 15 week old rats 7 days after injection.

		% of injected dose.	
<u>Skeleton</u>	Am	40.7 ± 3.7	28.1 ± 3.1
<u>Femora</u>	Am	4.1 ± 0.4	2.0 ± 0.2
	Pu	7.1 ± 0.2	7.7 ± 0.5

### Pregnancy and Lactation

During pregnancy and lactation the female rat undergoes many physiological changes that compensate for the increased nutritional demands of either the fetus or the newborn. There is a general increase in the consumption of food and a correspondingly increased intestinal absorption of essential minerals.

Of greatest importance for our studies are the changes that occur in bone. The metabolism of the bone minerals is in positive balance during pregnancy, with the formation of many new trabeculae in bone as calcium salts are laid down. The process is reversed in lactation. With the production of milk, the calcium in blood is utilized with a resulting resorption of trabecular bone. Pregnancy and lactation thus provide tools that permit an examination of the movements of americium and plutonium during both positive and negative calcium balance. This is

of some importance, for Herring has shown that both americium and plutonium are associated with resorbing rather than with active bone surfaces<sup>3</sup>.

The retention of americium by the femora of pregnant rats is less than that of the control rats (Table 3). The increase in the weight of the femur during pregnancy lends even more emphasis to this observation when viewed as a concentration. These trends were also observed in measurements of the whole skeleton. The period of greatest anabolic activity occurs at the end of pregnancy and it was this interval of pregnancy that showed the greatest reduction in the retention of americium by femur and the whole skeleton. Skeletal tissue in the stage of active calcification thus possesses a reduced ability to incorporate americium.

Table 3. Retention of americium during pregnancy, % of injected dose.

	Controls	0 - 10 d	12 - 22 d	17 - 22 d
Skeleton	26.1 ± 1.7	25.1 ± 2.9	22.4 ± 1.0	22.0 ± 0.6
Femur	1.03 ± 0.04	1.11 ± 0.07	0.96 ± 0.04	0.82 ± 0.02

Table 4. Retention of americium and plutonium by the ends and shafts of femur after 22 days of lactation, % of injected dose.

		Ends	Shafts
Am	lactating rats	0.45 ± 0.01	0.14 ± 0.01
	control rats	0.42 ± 0.02	0.12 ± 0.01
Pu	lactating rats	1.75 ± 0.14	0.40 ± 0.03
	control rats	2.04 ± 0.27	0.49 ± 0.07

Table 5. Specific activity of ends and shafts of femur after 22 days of lactation, % of injected dose/g dry bone.

Am	lactating rats	1.40 ± 0.05	0.65 ± 0.02
	control rats	0.94 ± 0.05	0.51 ± 0.03
Pu	lactating rats	5.5 ± 0.4	1.9 ± 0.2
	control rats	4.5 ± 0.6	2.0 ± 0.3

Table 6. Retention of americium by one femur at the end of lactation after restricted consumption of food.

	Food	Retention, % dose	Specific Activity, %/g dry bone
Control	ad lib	0.92 ± 0.05	1.26 ± 0.07
Mother	ad lib	0.86 ± 0.06	1.56 ± 0.11
Mother	17 g/d	1.09 ± 0.07	1.87 ± 0.12
Mother	10, 15 g/d	1.18 ± 0.06	2.14 ± 0.11

Table 7. Retention of americium by one femur in mother rats 10 days after weaning.

	Retention, % dose	Specific Activity, %/g dry bone
Mother	0.62 ± 0.04	1.09 ± 0.09
Control	0.92 ± 0.08	1.39 ± 0.13

The retention of americium and plutonium by bone of lactating rats is affected very little during the 22-day period of lactation of the rat (Table 4), bearing in mind the decimation of the trabeculae of bone by the end of lactation. The amounts of americium and plutonium were not significantly different in either the ends or shafts of femur at the end of lactation (P's between 0.2 and 0.3). The weight of the ends of the femur is substantially reduced and Table 5 shows that the ability of cortical bone to concentrate americium and plutonium is increased by the catabolic activity of lactation, the effect being greater for americium than for plutonium.

The stress of restriction of food consumption by lactating rats has also been studied. Normally, the daily consumption of food by a lactating rat increases steadily during lactation and reaches a level four times greater at the end of lactation than at the beginning. In one experiment, lactating mothers were restricted to the weight of food consumed ad libitum by control rats - 17 g/d. In a second experiment, the mothers were restricted to 10 g/d for the first 15 days of lactation and 15 g/d for the last 7 days of lactation. The retention of americium by femur under these stresses is presented in Table 6. The effect of the stress is to increase the retention of americium by the femur, the greater the stress the greater the increase in retention. It is therefore evident again that increased catabolic activity results in an increased retention of americium by rat bone.

Immediately after weaning, a mother undergoes a period of intense anabolic activity, during which the bone mineral used up during lactation is replaced. We examined the metabolism of americium during this anabolic period by injecting americium on the day of weaning and measuring the retention 10 days later. The retention of americium by the femur of the mothers was substantially lower than that of the controls (67%) (Table 7). While the weights of the femurs of the mothers were still significantly lower than those of the controls 10 days after weaning, the ability of the bone to concentrate americium, as demonstrated by the specific activity, was also substantially reduced (78% of controls).

#### Effects of artificial stresses

We summarise here the effect on the retention of americium and plutonium by bone of parenteral administration of parathyroid hormone, calcium gluconate and calcitonin. The effects of a diet free of calcium and of parathyroidectomy are also described.

Parathyroid hormone. The work of Herring showed that resorbing surfaces of bone are important in the retention of americium and plutonium and our investigation of the metabolism of americium and plutonium during reproduction suggested that the physiological stresses that affect bone building have a measurable influence on the retention of these elements by bone. We therefore investigated the effect of parathyroid hormone, because of its role as a regulator of bone resorption. We injected subcutaneously 50 units of PTH twice a day to male rats

of about 450 g body weight. The rats were also fed a diet free of calcium. Americium and plutonium were injected 7 days after the rats had been on this regime and the animals were killed and their tissues analysed after a further 14 days. The results for femur are presented in Table 8. The diet without calcium had a significant effect in increasing the retention of americium and plutonium. The parathyroid hormone further increased the retention of americium but had no further effect on the retention of plutonium. These results therefore confirm the affinity of americium and plutonium for resorbing surfaces, but point out again that the behaviour of americium and plutonium in bone is certainly different quantitatively, if not qualitatively.

Table 8. Effect of parathyroid hormone and a diet without calcium on the retention of americium and plutonium by one femur of male rats 14 days after injection, % of injected dose.

	Am	Pu
PTH + diet	1.77 ± 0.05	3.58 ± 0.08
Diet alone	1.45 ± 0.06	3.58 ± 0.09
Control	1.23 ± 0.05	3.38 ± 0.08

Table 9. Effect of calcium gluconate on the retention of americium and plutonium by one femur of male rats 5 hours after injection, % of injected dose.

	Am	Pu
Calcium gluconate	1.00 ± 0.04	1.29 ± 0.06
Control	1.20 ± 0.02	1.38 ± 0.08

Table 10. Effect of calcitonin on the retention of americium and plutonium by one femur of male rats 5 hours after injection, % of injected dose.

	Am	Pu
Calcitonin	1.15 ± 0.05	1.45 ± 0.37
Control	1.09 ± 0.04	1.38 ± 0.08

Table 11. Effect of parathyroidectomy on the retention of americium by one femur of male rats 5 hours after injection, % of injected dose.

PTX	1.09 ± 0.10
Control	1.26 ± 0.04

Calcium gluconate. Having established that the increase in resorption produced by a diet deficient in calcium or by parathyroid hormone increases the retention of americium and plutonium, the opposite effect was sought. Repeated administration of calcium gluconate was used to increase the level of calcium in the blood and reduce the bone resorption. Intraperitoneal injection of calcium gluconate at half-hourly intervals, starting 30 minutes before injection of americium and plutonium, resulted in a significantly reduced retention of americium by femur 5 hours later ( $P \sim 0.01$ ) and a smaller decrease in the retention of plutonium that was not significant ( $P \sim 0.3$ , Table 9).

Calcitonin. The effect of calcitonin on the plasma calcium level is the opposite of the effect of parathyroid hormone<sup>4</sup>. Administration of salmon calci-



tonin to rats at a dose of 2 units/kg produced a transitory reduction of plasma calcium from 100 mg/l to 70 mg/l. The effect lasted for about 4 hours. We administered calcitonin intraperitoneally at the same time as americium and plutonium and repeated the calcitonin after 2-1/2 hours. The rats were killed 5 hours after administration of the radionuclides. The retention of americium by femur is presented in Table 10. The administration of calcitonin in this experiment had no effect on the retention of americium or plutonium by femur.

Parathyroidectomy. An alternative method of reducing the level of bone resorption was to remove the endogenous source of parathyroid hormone. Parathyroidectomy was performed 4 days before the injection of americium. Blood calcium levels were reduced to about one-half of normal by this operation. The effect of parathyroidectomy on the retention of americium by femur 5 hours after injection (Table 11), was similar to that found with parenteral administration of calcium gluconate. The decrease in retention of americium was 14% with parathyroidectomy and 17% after parenteral administration of calcium gluconate.

### Discussion

The use of stresses that increase or decrease resorption of bone, either by natural or artificial means, increases and decreases the retention of americium by bone, with similar but smaller effects for plutonium. The biggest difference, while statistically significant, is only a 40% change in the retention of americium. The largest effect observed so far is the increase in retention after administration of parathyroid hormone. The decrease obtained with administration of calcium gluconate is more modest. Our experiments with calcitonin have not yet been completed, but no effect has yet been observed. The modest size of the changes obtained so far does not offer much encouragement for the use of hormones or other pharmaceutical agents that modify bone metabolism as therapeutic agents to reduce the retention of transuranic elements by bone. However, there are other agents that influence the metabolism of bone and it might be of interest to study the effect of, say, growth hormone, calcium ascorbate, or combinations of these agents with a chelating agent.

It has sometimes been assumed that americium and plutonium behave similarly as contaminants of biological systems and, while similarities are apparent in this work, there are some notable differences, with the retention of plutonium much less affected by variations in bone metabolism than that of americium.

### References

1. R.F. KEOUGH and G.J. POWERS, *Anal. Chem.* 42, 419 (1970).
2. A. DURAKOVIC, J.G. HOLLINS, M.C. STORR, *Health Phys.* 24, 541 (1973).
3. G.M. HERRING, J. VAUGHAN, M. WILLIAMSON, *Health Phys.* 8, 717 (1962).
4. J.M. VAUGHAN, *The Physiology of Bone*, p. 180. Clarendon Press, Oxford (1970).

## RADIATION ACCIDENTS

### INDUSTRIAL RADIATION INCIDENTS IN THE UNITED KINGDOM

By C D Burgess and E G Weatherley

HM Factory Inspectorate, Department of Employment,

London, United Kingdom

#### Abstract

Certain categories of radiation accidents, incidents and excessive radiation exposures are reportable to HM Factory Inspectorate. This paper describes the method of analysing reports of excessive radiation doses. The analysis includes the processes and locations associated with these reports, the magnitude of the radiation doses received and the age groups of the exposed persons. Causes are divided into management errors, operator errors and equipment failure. The analysis shows that industrial radiography on engineering construction sites is the process which produces most excessive radiation exposures. Unsuitable equipment, inadequate supervision and training of radiographers are found to be the main causes of excessive exposures.

#### Introduction

##### Objective

The purpose of this paper is to review and analyse radiation incidents reported to HM Factory Inspectorate between 1968 and 1972, to identify areas where radiation doses in excess of the maximum permissible are occurring, to indicate where, if any, additional effort needs to be deployed in industry and what particular problems need to be solved. Incidents occurring during 1972 are analysed in detail.

##### Scope

This paper deals with incidents reported to HM Factory Inspectorate under the Ionising Radiations (Sealed Sources) Regulations 1969.<sup>1</sup> The application of these regulations has been described in previous papers.<sup>2,3</sup> In particular they do not apply to hospitals, nor to research and teaching establishments. Incidents reported under the Ionising Radiations (Unsealed Radioactive Substances) Regulations 1968<sup>4</sup> have been excluded because these regulations only apply to a small proportion of persons employed in establishments where unsealed radioactive substances are used and consequently would not be representative.

##### Reportable Incidents

Reportable incidents are defined as those reportable to HM Factory Inspectorate under the regulations;<sup>1</sup> namely

- (a) Incidents where it appears that a person has received a radiation dose in excess of the maximum permissible dose specified in the regulations.
- (b) Breakage or leakage of sealed sources.
- (c) Lost or mislaid sealed sources.

Only category (a) is analysed and discussed. The word incident is used in preference to accident because most excessive exposures result from chronic causes rather than acute accident situations.

Notification

Where a dose assessment on a single film badge is in excess of the maximum permissible it is most unlikely that the incident remains unreported due to the close liaison between HM Factory Inspectorate and approved dosimetry laboratories. On the other hand where it arises from the summation of a number of film badge dose assessments over the calendar quarter the position is probably less satisfactory. It is, of course, impossible to assess whether significant numbers of persons not wearing film badges receive doses in excess of the maximum permissible, but the operation and enforcement of the various legislative requirements in the United Kingdom ensures that this is unlikely to occur.

Incident Analysis

Identification of genuine incidents

All reported incidents are investigated and analysed. Since there is an obligation on factory occupiers to report all apparently excessive doses, the first task is to separate the genuine incidents from the obviously false. Inevitably there is a grey area between the two categories where in spite of all efforts no specific circumstances can be identified to account for the film badge assessment. These doubtful cases, which are few in number, are included with the genuine incidents. Incidents which are found on investigation to be false are discarded from further consideration.

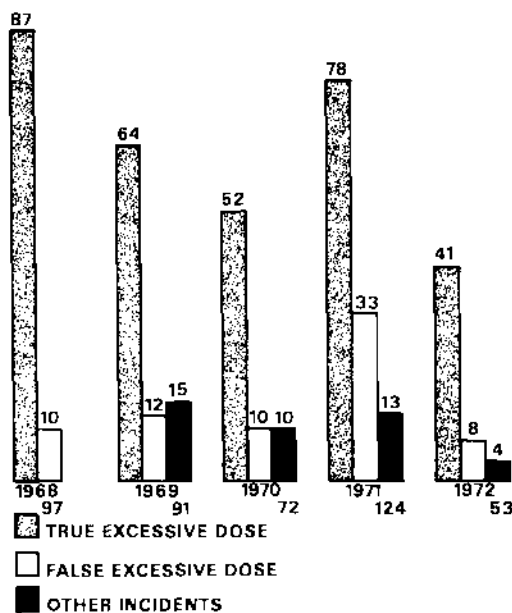


Figure 1  
RADIATION INCIDENTS REPORTED TO H M F I

Figure 1 shows the number of incidents reported to HM Factory Inspectorate in the years from 1968 to 1972. There is no obvious trend in the number of genuine excessive exposures reported and analysed in the years quoted although the number in 1972 was the lowest so far recorded. The number of incidents which proved on investigation to be false remained fairly constant with the exception of 1971 when there was an abnormally large number arising out of two incidents involving twelve personal dosimeters. In one case seven film badges were accidentally irradiated while not being worn when a radiographer was carrying out a radiograph during the night shift. In the other incident five film badges were exposed to organic vapours while being worn.

The other incidents shown in figure 1 refer to breakage or leakage of sealed sources or lost or mislaid sealed sources. All lost sources were eventually recovered.

**Process and location**

An analysis of the place at which incidents occur and the type of work involved is very useful in identifying the areas of risk and in allocating the limited resources available for inspection and enforcement.

Figure 2 shows the location of radiation doses in excess of the maximum permissible reported from 1969 to 1972. Whereas it was reported in 1969 that gamma radiography of pipelines was the major area of risk, the position has changed markedly. From a peak of 54% of the total in 1969, excessive exposures on pipeline sites had fallen to only 17% in 1972. This is partly due to a reduction in the total mileage of pipeline laid during this period and partly to the considerable increase in the use of X-ray Crawlers for pipeline radiography. There was however an increase in the use of sealed sources at engineering construction sites, for example, petro-chemical factories and power stations under construction or repair. Incidents arising from the use of ionising radiations in other processes show no significant pattern over these years.

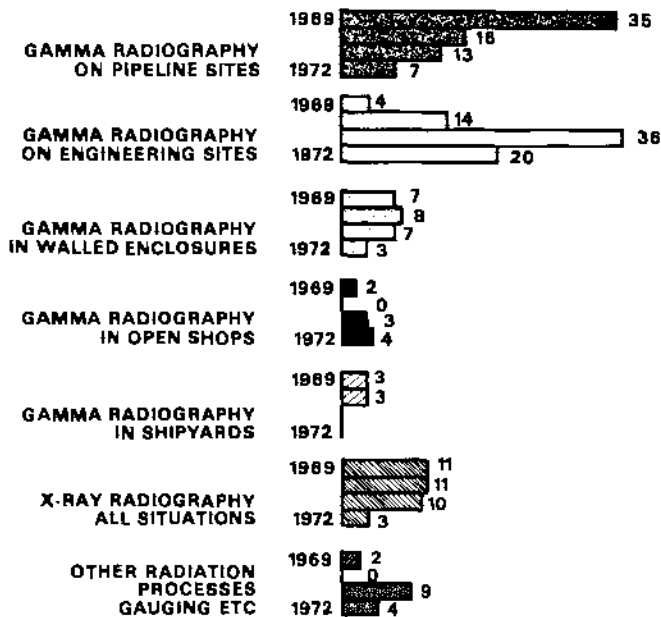


Figure 2  
PROCESS/LOCATION OF EXCESSIVE RADIATION EXPOSURES

The situation for 1972 is summarised in figure 3. It shows that in spite of all efforts in the field of radiography, this process still accounts for 90% of all reported excessive doses. Gamma radiography accounts for the majority although X-ray radiography is widely used. A comparative review of the incidents associated with radiography on construction sites and in factories has been published.<sup>5</sup> That the other categories account for less than four incidents each suggests that a high standard of radiological protection exists in other processes using ionising radiations.

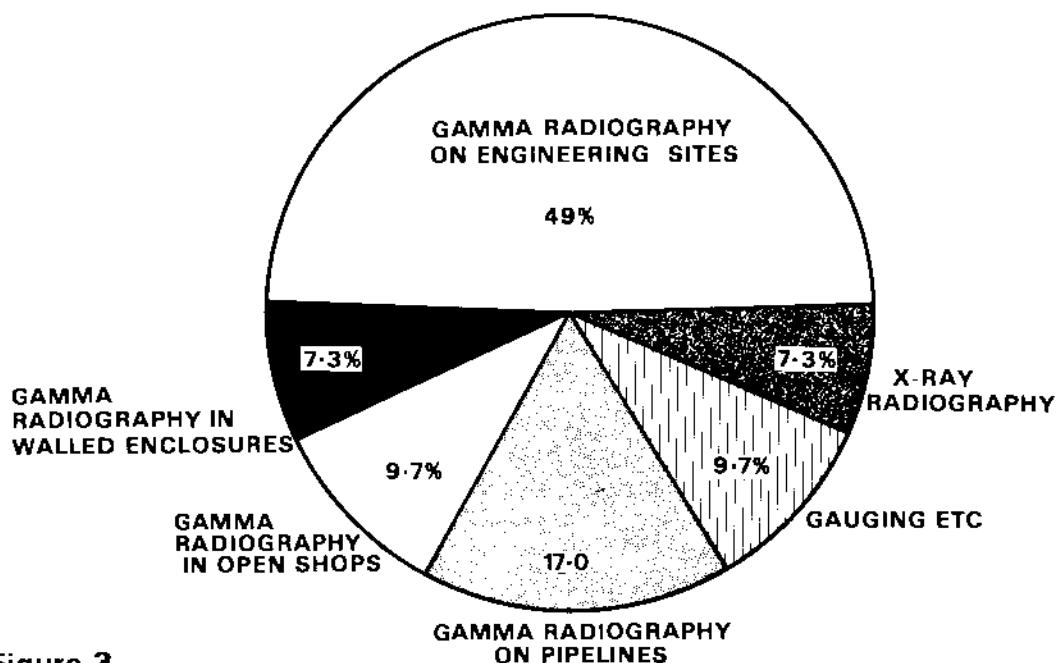


Figure 3  
PROCESS/LOCATION OF EXCESSIVE RADIATION EXPOSURES IN 1972

#### Magnitude of radiation doses

Figure 4 gives an indication of the radiation doses received by the individual in the calendar quarter to which the report relates. These figures are normally those given by film badge dose assessments. However, in some cases a person was exposed to a grossly non uniform radiation field and the film badge did not give a reasonable indication of the dose to that part of the body most affected. In these cases the figures have been amended to show dose assessments to the organ of interest.

The table therefore contains those doses in excess of the maximum permissible to the whole body or to other parts of the body where these doses have exceeded the appropriate maximum permissible doses to that part of the body. The table should therefore be interpreted with care.

70% of the 1972 reports fall in the 3 to 5 rem range. These were usually chronic exposures resulting from a number of frequent increments of small doses accumulated over the calendar quarter giving a cumulative total in excess of the maximum permissible. These cases usually arise from long hours of work, unsuitable equipment, relatively unsatisfactory working conditions or methods of work.

DOSE RECEIVED	1969	1970	1971	1972
3-3.5 REM	11	8	17	13
3.5-5.0 REM	26	24	35	16
5.0-10.0 REM	12	8	11	4
10.0-25.0 REM	8	9	6	4
25-50 REM	1	2	1	2
50-100 REM	4	0	1	0
>100 REM	2	0	5	1
ACCURATE EVALUATION NOT POSSIBLE	0	1	2	1

Figure 4 MAGNITUDE OF EXCESSIVE DOSES

Doses in excess of 5 rems consist in the main of acute doses arising from a single relatively high dose received in one exposure due to accident conditions. All the radiation doses in excess of 100 rems were received either to the head or hands in extremely non uniform radiation fields. There were one or two circumstances over this period where it was not possible to make an accurate evaluation of the radiation dose received by the individual. In most of these cases the individual was not wearing a personal dosimeter.

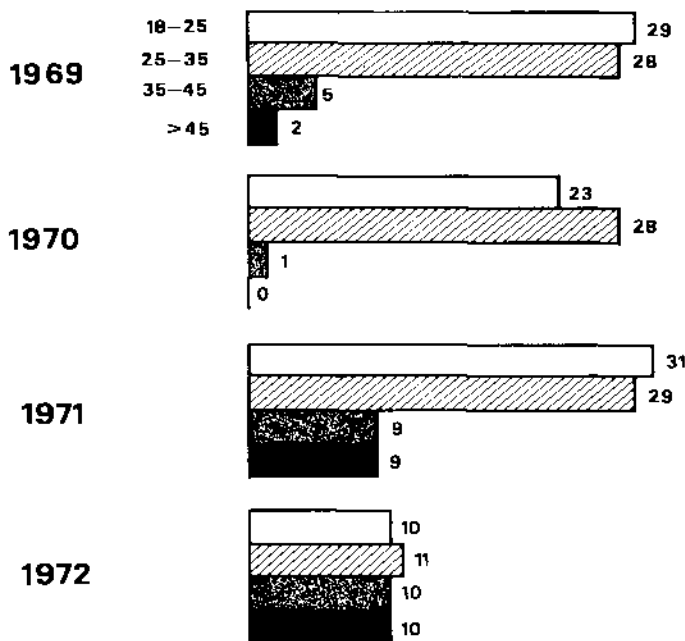


Figure 5 AGE GROUPS OF EXPOSED PERSONS

## Somatic Injuries

Five cases of acute somatic injuries were reported during the period 1968 to 1972. Four involved finger burns as a result of handling sealed sources or source holders without handling tongs. In three cases it was estimated that the doses to the fingers were of the order of several thousand rems over fairly short periods. In the fourth case where the practice was carried out over many months biological dosimetry<sup>6</sup> gave an estimate of 200 rems as the equivalent whole body dose. In the fifth case, the cause of which was not clearly established, a dose of not less than 2,000 rems was received to the chest and it was estimated<sup>7</sup> that the equivalent whole body dose was between 70 and 105 rems.

## Age groups of exposed persons

The proportion of excessive exposure to persons in the most genetically significant age groups, that is those up to twenty-five years old and those between twenty-five and thirty-five years old was considerably lower in 1972. (See figure 5). It is too early to say whether this trend is significant.

## Incident causation

In terms of preventing the recurrence of an incident and to help in the allocation of limited resources available to users and enforcement agencies, causation is the most important factor in incident analysis. Unfortunately the categorisation of causation is a subjective process and is likely to be significantly affected by the attitudes of those involved. Following Catlin's paper<sup>8</sup> we reviewed and reorganised our classification system but we found that it was impracticable to allocate a primary cause to each event. Incidents cannot often be ascribed to a single cause and many are the result of a sequence of events which may involve errors on the part of the management or employee or failure of the equipment, each of which played an important part in the incident. For these reasons, three basic categories were chosen

Management error.  
Operator error.  
Equipment failure or malfunction.

Where more than one error in a particular category (for example, more than one management error) contributed to an incident, the most significant in relation to that incident was chosen.

## Management error

Figure 6 shows that in 78% of the incidents reported in 1972 a management error contributing to the causation sequence was identified.

Equipment 34% of these incidents were ascribed to the provisions of unsuitable equipment. This category primarily relates to gamma radiography exposure containers and ancillary equipment used by itinerant radiographers. It is not always easy to provide operators with an optimum selection of such equipment together with suitable source types and strengths for a protracted series of tasks which may vary widely in scope. Nevertheless in our view a significant improvement in this area should materially reduce the number of incidents.

We have classified gamma radiography exposure containers into three categories; torch type, shutter type and projection type. The torch type

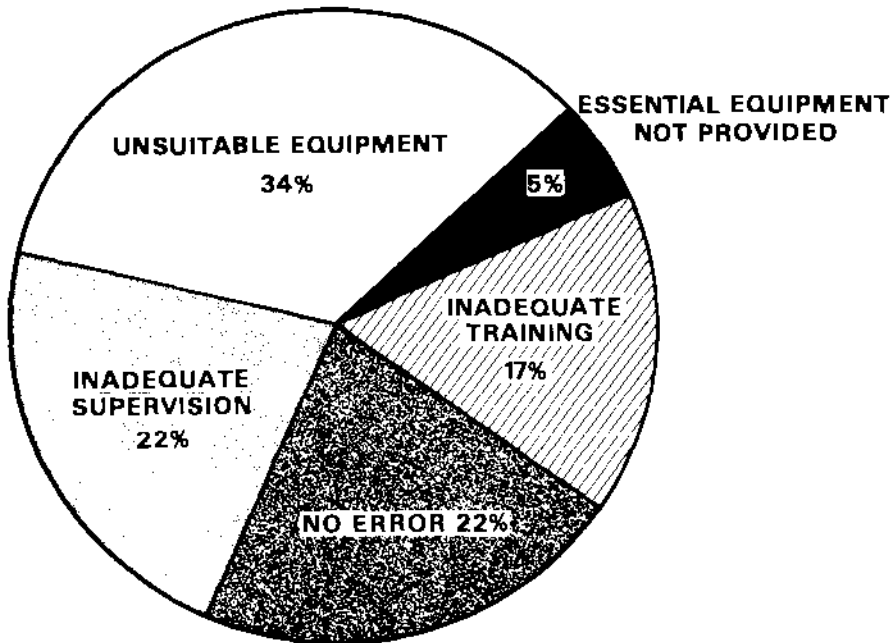


Figure 6 MANAGEMENT ERROR 1972

is the most frequently used due to its advantages of lightness, simplicity, robustness and flexibility in use. It does however suffer from the disadvantage that the operator needs to be relatively close to the source at the time of exposure. Good methods of work are therefore vital. Poor working practice leads to chronic excessive doses.

There have been several instances where insufficient shielding has been provided by the torch/castle combination of a radiography exposure container thus exposing the radiographer to high radiation dose rates. In another case a radiographer was provided with an exposure container which, when he arrived on the site after a long journey, was found to be too large to use in the working space available. In order to avoid delay the source was used without the castle and the radiographer received an excessive dose of radiation. Another common fault was the use of sources with too high an activity for a particular series of radiographic shots. This enables a very large number of shots to be carried out in a short time but invariably results in the radiographer receiving a higher radiation dose.

Shutter type containers are relatively light but slightly less robust since there are more working parts. They are however usually larger than torch type containers and sometimes it is necessary to remove the source from the container for difficult exposures. This is a delicate operation and can lead to chronic excessive exposures or, if the operation is mishandled, to an accident situation.

Remotely controlled projection type exposure containers are more expensive and usually much bigger and heavier. Unfortunately the equipment is often not robust enough for the very arduous conditions of use on site. This results in damage to the projection cable and the source is sometimes left in the projection tube when the operator believes it to have returned to its fully shielded position. This is often not discovered until much later. This accident condition gives rise to acute radiation doses in excess of the maximum permissible.



Figure 7 shows the number and type of exposure containers associated with excessive radiation exposures from 1969 to 1972.

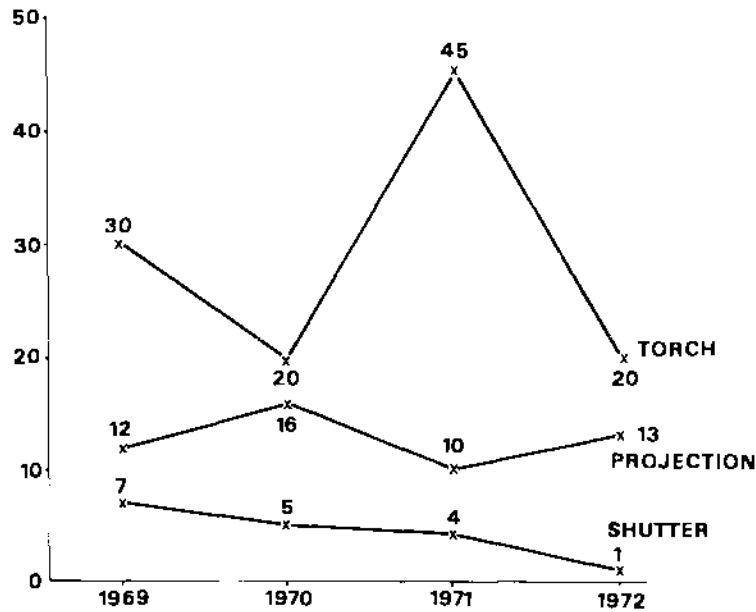


Figure 7 TYPE OF EXPOSURE CONTAINER

As we mentioned in a previous paper<sup>3</sup> it is our view that there is still considerable potential for the development of exposure containers and suitable accessories particularly for situations where access is difficult and clearance is limited. Unfortunately it is by no means easy to develop a performance standard for this equipment due to the variety of conditions in which it is used. Various recommendations by the International Labour Office, ICRP and the British Standards have amongst other criteria, specified the exposure rate at the outside of the closed container. This is useful in limiting the exposure of radiographers while the apparatus is in transit or temporary storage. However when one considers that the exposure rate outside a closed container may only be 20 mR/h while it may be 2,000 mR/h or more when the apparatus is in use it is quite obvious that the latter situation is the critical factor in determining the radiographers exposure.

In our radiation laboratory we are evaluating the protection afforded by a range of exposure containers both under conditions of use and storage with a view to determining parameters for a performance standard. We hope to publish a paper in due course.

Supervision Lack of supervision accounted for 22% of the excessive exposures. In most cases these incidents arose as a result of operators failing to use suitable techniques in the field. It is managements responsibility to ensure that the operators are trained to use approved techniques, and supervision is necessary from time to time to ensure that this is done. A careful and frequent check on radiation dose records will identify cases where special supervision is necessary.

Training Failure to provide adequate training for operators was still an important factor in the area of management errors. It still accounted for 17% of the excessive doses although this is a significant reduction from the 1970 figure of 40%. Some radiography contractors organise excellent training schemes. Others, particularly small firms have no formal training facilities themselves and rely on a combination of "on the job" training sometimes supplemented by external courses. Some of these courses are not orientated towards the detailed process knowledge or the technical equipment used for radiography and fail to demonstrate clearly and in simple terms the techniques of minimising radiation exposure under site conditions. We believe that formal training in radiological protection is essential. It should, nevertheless, be practically based on the equipment used by radiographers. We have sought the advice and help of the National Radiological Protection Board to improve the situation in this field.

Safety equipment The provision of essential safety equipment such as dose rate meters, warning signals and barriers was relatively satisfactory and did not contribute significantly to management errors.

Operator error

Most operator errors such as failure to monitor or the use of unsuitable techniques are related to management errors such as inadequate training or lack of supervision. Only wilful disregard of instructions, failure to use equipment provided and human error are wholly attributable to the operator. These are classified as other causes in figure 8.

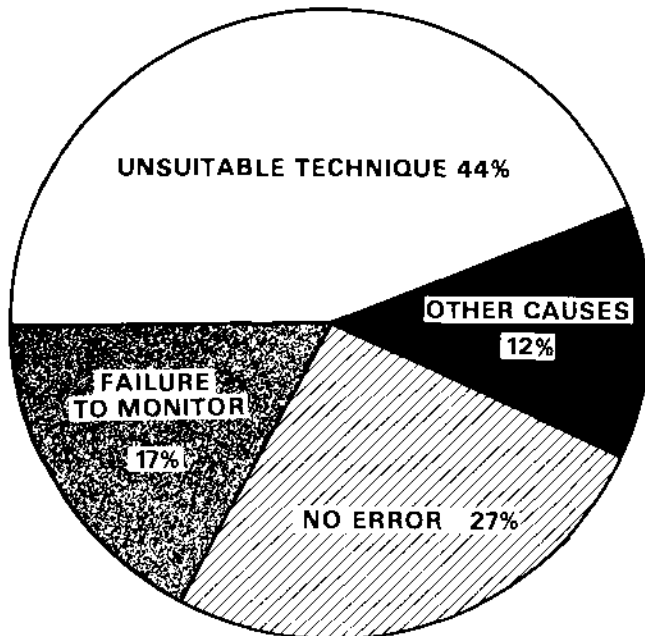


Figure 8 OPERATOR ERROR 1972

Unsuitable or poor techniques increased from 27% in 1971 to 44% in 1972. These were mainly associated with torch type exposure containers, for example, using sources of too high an activity or failing to retire to a safe distance during the exposure

Failure to monitor, usually in connection with a remotely controlled projection type exposure container accounted for seven (17%) of the incidents involving operator error. Two incidents were caused by radiographers failing to lock the exposure containers in the closed position before transportation.

For the first time since 1969 no worker handled a source holder directly.

#### Equipment Failure

Equipment failures or malfunctions are classified in figure 9. Equipment failure was identified as a contributory factor in 30% of the reports.

While these equipment failures were important factors in twelve accidents in 1972, excessive exposure would probably have been avoided in most cases if the operators had been adequately trained and had followed the correct procedures.

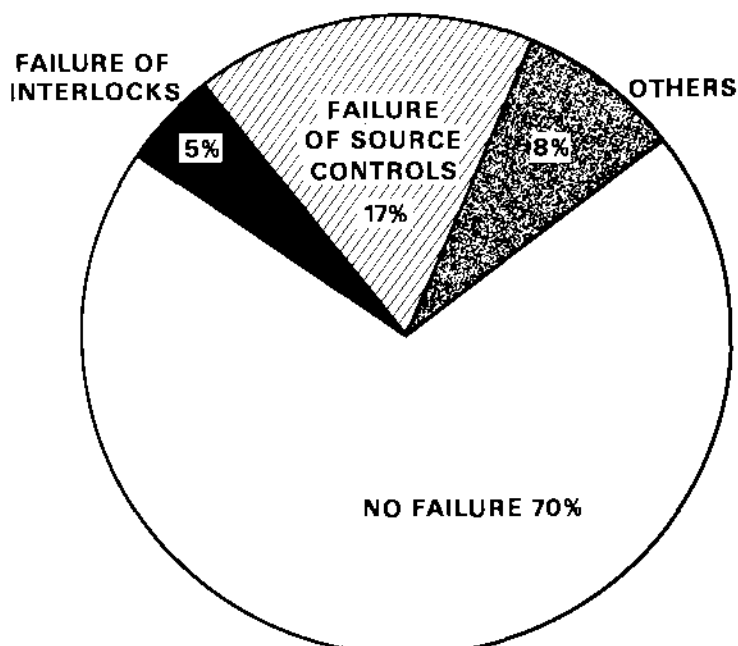


Figure 9 EQUIPMENT FAILURE 1972

Failure of source controls, that is sources becoming disconnected from remote control devices while exposed and shutters failing to close showed little variation at 17% of the total compared with previous years.

Interlock failures at X-ray enclosures contributed to 5% of the incidents in 1972. In terms of actual numbers this must be very small proportion of the total X-ray enclosures in use but this makes them all the more unexpected when they do occur. No failure of warning signals for X-ray enclosures were reported during the year.

### Conclusions

Analysis of industrial radiation incidents in the United Kingdom shows that gamma radiography at engineering construction sites is now the type of work giving rise to most excessive radiation doses. The advent of X-ray crawlers appears to have reduced the risk on pipeline work. The frequency of excessive radiation doses in work other than radiography involving the industrial use of ionising radiations is very low.

Most excessive doses are in the range of 3 to 5 rems in a calendar quarter and are received chronically over the period rather than as a result of accident conditions.

We believe that greater attention needs to be paid by manufacturers to improving the design of gamma radiography equipment to provide better protection for the operator and to prevent failures under the arduous conditions of use. It would be helpful if a performance standard could be developed for this equipment to specify the protection which should be provided under conditions of use.

Users should select their equipment more carefully for the particular conditions of use and ensure that radiographers are adequately trained paying particular attention to minimising exposure under difficult field conditions and the procedures to be followed when accident conditions arise.

### Acknowledgement

Thanks are due to HM Chief Inspector of Factories for permission to publish this work, and also to our colleagues for assistance in collecting and collating the information.

### References

- 1 Ionising Radiations (Sealed Sources) Regulations 1969  
SI 1969 No. 808 HMSO
- 2 Burgess C D Apparent excessive doses from radiation in United Kingdom factories. Proc Sym Handling of Radiation Accidents 1969.  
IAEA SM 119/10.
- 3 Burgess C D Radiological Protection problems in factories.  
J Inst Nuc Eng 1973 14, 51.
- 4 Ionising Radiations (Unsealed Radioactive Substances) Regulations 1968  
SI 1968 No. 708 HMSO.
- 5 Atherton N J An investigation of the radiation doses received by industrial radiographers J Nondes. Test Soc 1973 July 112.
- 6 Dolphin et al. Chromosome aberration dosimetry in a case of overexposure to radiation. Nature 241 69 (1973).
- 7 Purrott et al. NRPB (R)5 (National Radiological Protection Board, Harwell 1972).
- 8 Catlin R J Radiation accident exposure - causes and lessons learned  
Proc Sym Handling of Radiation Accidents 1969 IAEA-SM 119/6.

THE INVESTIGATION AND RECONSTRUCTION OF A SEVERE RADIATION  
INJURY TO AN INDUSTRIAL RADIOGRAPHER IN SCOTLAND

N. T. HARRISON, P. C. ESCOTT  
National Radiological Protection Board, Glasgow  
G. W. DOLPHIN, R. J. PURROTT  
National Radiological Protection Board, Harwell  
J. I. ALEXANDER  
H.M. Factory Inspectorate, Glasgow  
K. E. HAINAN  
Institute of Radiotherapeutics, Glasgow

Abstract

This paper describes a severe radiation injury received by a young industrial radiographer concerned with pipeline radiography. The reconstructions carried out to explain the incident are discussed together with the calculations made to predict the radiation doses at the site of the injury and the effective whole body dose. These results are compared to the clinical estimates of the radiation dose profiles at the injury and the estimates of radiation dose made by reference to the film badge worn by the worker and to the chromosome aberrations detected in the lymphocytes obtained in a blood sample.

The injury was observed on the upper left area of chest wall adjacent to the left nipple, involving a circular area of skin about 100 mm diameter extending in depth 30 to 40 mm to ribs and even heart muscle.

The worker associated this injury with an exposure to an open industrial radiography source housing containing a 25 curie iridium-192 source during a short car journey with the container open on the adjacent front seat and directed towards him. Evidence is presented to discount totally this explanation and the possible alternatives derived from consideration of the clinical estimates of the radiation dose profile are discussed in detail. The radiation doses at the site of the injury were thought to be up to about 20,000 rads. A brief statement is made of the medical treatment given.

Introduction

The purpose of presenting this paper is to report one of the most severe accidents involving exposure of an industrial worker to radiation which has occurred in Great Britain. The paper also brings to light the difficulties in attempting to explain an incident in retrospect when most of the information comes from the person concerned who cannot, or may not wish to, recall the exact circumstances.

### Description of Incident

The young man concerned, aged 20 years, commenced employment on 25th August, 1969 as a trainee radiographer with a large company engaged in non-destructive testing. His initial training was carried out informally in discussion with other more senior radiographers over the first two weeks. Towards the end of the first week he carried out radiography and from the second week onwards he sometimes worked without direct supervision.

On 20th September he was provided with an iridium-192 source, strength 25 curies, contained in an industrial protective source housing, to radiograph a 24 inch gas pipeline. The source housing, figure 1, had been designed by the company themselves to the shielding requirements specified in British Standard, BS.4097, 1966. Provision was made for locking the source in the closed position but on this occasion no padlock was provided.

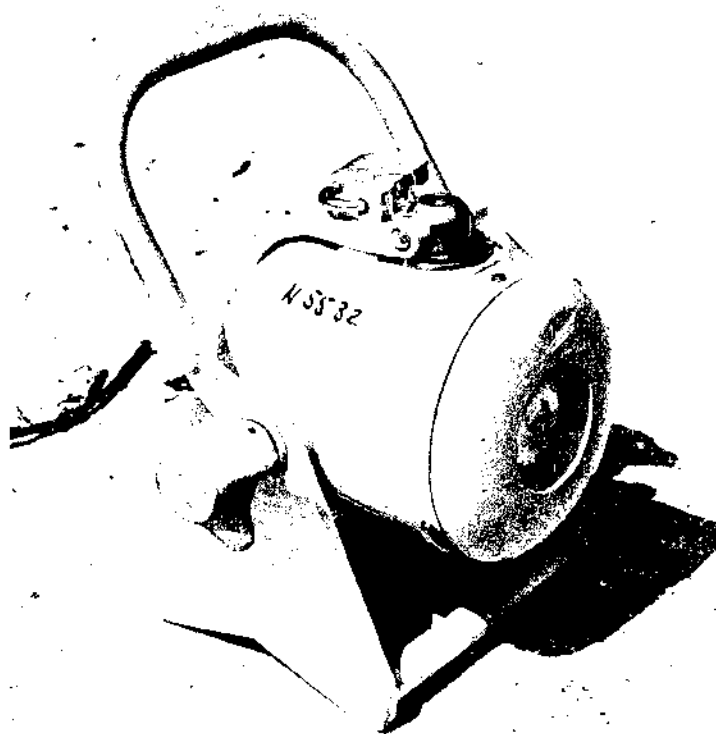


Figure 1. Protective Source Housing used for Industrial Radiography

He was the last radiographer to complete his work and was therefore left entirely on his own at the site. On finishing he loaded the exposed films onto the back seat of his car and placed the source housing on the front passenger seat.

At the completion of his journey a fellow worker noticed that the source housing was in the open position with the radiation beam directed towards the driving seat. It was estimated that he was exposed to the radiation in this situation for three hours. During this time his radiation monitoring film, not in a film holder, was in a wallet in his right hand hip pocket.

He did not work again with radioactive sources from this time until the onset of clinical symptoms.

### Clinical Considerations

The clinical symptoms became apparent on 2nd October, 1969. He noticed for the first time a small white patch on his chest wall just below the left nipple. This patch was surrounded by a circular area 15 mm in diameter which gradually became inflamed. On 5th October he began to feel unwell. The chest area had started to swell, the reddening had extended to an area 100 mm in diameter, the central white patch had spread in size and a blister formed around its edge.

At this stage he sought medical advice. He approached his local general practitioner who, in view of his occupation, referred him to a consultant radiotherapist at the Institute of Radiotherapeutics, Western Infirmary, Glasgow.

By 8th October the injury had assumed an oval shape, bright fiery red in colour, about 100 mm in diameter, with a central spot, deeper and darker red in colour, 20-30 mm in diameter.

Other lesser injuries were also noted. There was a smaller, similar lesion about 20 mm in diameter over the sternum, a small lesion on the inside of the left wrist and blistering of the fingertips of the left hand. During the following week the central parts of both chest lesions vesicated and became ulcerated. Over the next few weeks the erythema decreased around the main injury, but the central ulcer extended to cover an oval area 100 mm x 90 mm developing a deep black scab in the centre, figure 2.



Figure 2. Appearance of main chest injury - December 1969.

The small sternal lesion scabbed and healed completely by the end of January 1970. The deep scab continued to develop over the main lesion however without any healing. He was admitted to Carniesburn Hospital, Glasgow in April 1970 with a view to surgery.

The slough was excised from the lesion revealing an area of necrotic tissue about 50 mm in diameter, which it was also decided to remove. During this

operation, the radionecrosis was found to involve parts of the fourth and fifth ribs. Damage had also occurred to the pericardium and an area of radionecrosis 10 mm in diameter was observed on the heart muscle. The surgery was completed with a thoraco-abdominal flap. The appearance on discharge from Carniesburn Hospital is shown in figure 3.



Figure 3. Appearance of main chest injury after surgery - October 1970.

Some postulations of radiation dose can be made on the basis of the clinical symptoms, their speed of progression and by comparison with tissue damage in radiotherapy. It is reasonable to assume that the injuries were due to single short exposures.

- (1) The area of skin of the main lesion must have received a dose in excess of 2,000 rads but not greater than about 20,000 rads.
- (2) The skin immediately surrounding the lesion where there was no erythema must have received a dose less than 800 rads.
- (3) The area of observed damage to the heart muscle must have received a dose of about 2,000 rads.
- (4) The area of skin of the smaller chest lesion, wrist and fingertips must have received a dose not much less than 1,500 rads.
- (5) The original radiation exposure had occurred about 7 to 10 days prior to the reported onset of symptoms.

#### Physical Dose Estimates

##### Car Incident

Details of the car incident were obtained by careful questioning of the radiographer and a reconstruction carried out in October 1969. A water filled phantom was used to simulate the body to measure the attenuation by tissue for iridium-192 gamma radiation.



The reconstruction showed that the closest distance from the source to the car driver was 0.4 m at the level of the left hip. The estimated three hour exposure time gave an average whole body dose of 45 rads calculated by determining integral dose in body tissue and averaging over the whole body. The maximum dose estimated was 215 rads to the left hip. The absorbed dose calculated at the surface of the right hip gave 7 rads in water. This can be directly compared with the absorbed dose of 7.5 rads in water recorded by the film badge exposed during this incident in a wallet in the radiographer's right hip pocket.

At this early stage the good agreement between the calculated dose at the site of the film badge and that recorded by the film gave a degree of confidence in the reconstruction. Medical opinion had not yet ruled out thermal burns as the cause since the symptoms had not fully developed and the severity of the chest injury was not realised. It was impossible for the incident as described to give rise to the very localised radiation burn on the chest or damage to the wrist and fingers, consequently further radiological investigation was suspended.

### Main Chest Injury

Due to circumstances outwith the control of the authors, medical treatment continued but no further radiological assessment took place until May 1970 by which time it was clear that there had been heavy exposure in addition to that occurring in the car.

After full development of the symptoms, the dimensions and radiation dose profile of the main injury derived from the degree of biological damage are shown in figure 4.

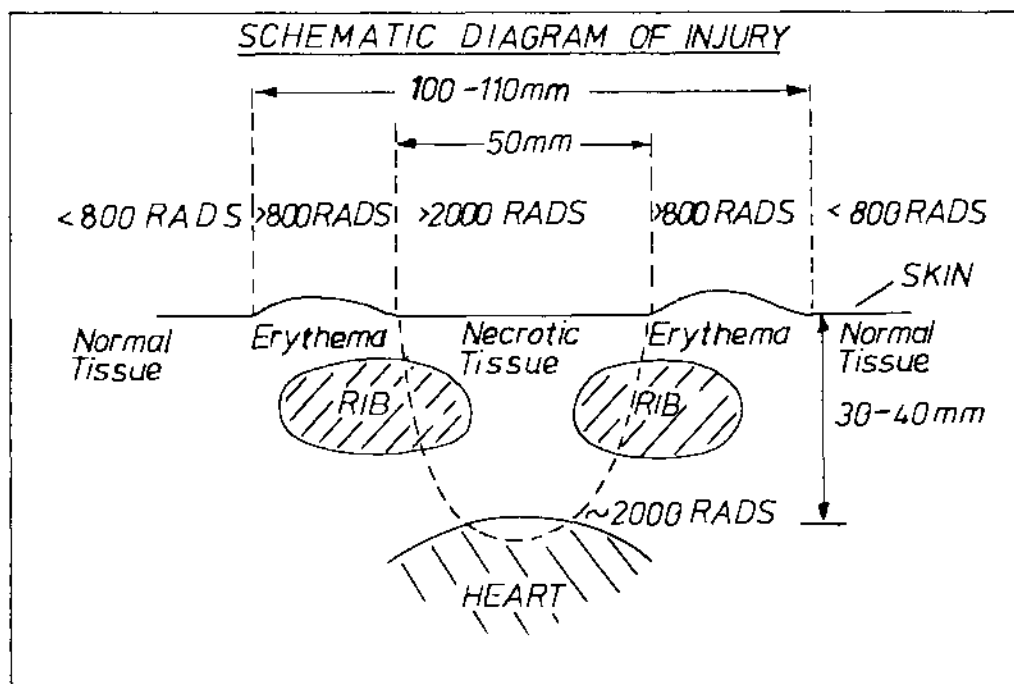


Figure 4. Schematic diagram of main chest injury showing dimensions, and estimates of dose based on the clinical symptoms.

Since the exposure had probably taken place about the time of the car incident, and to be consistent with the availability of sources to the radiographer, the 25 curie iridium-192 source was used to reconstruct the radiation dose profile for the chest injury.

Two possibilities were considered, first that the source had been removed from the source housing and was therefore unshielded and secondly that the source was mounted inside the source housing in the "exposed" position.

Radiation dose profiles were calculated for both cases at a number of source to skin surface distances and were normalised to deliver 2,000 rads at a depth of 30 mm of tissue, i.e. the heart muscle and are shown in figures 5 and 6.

Comparison of these calculated dose profiles with the estimated doses required to cause the observed biological damage of the injury, shows that the necessary conditions of exposure are met either with the unshielded source at a distance of 10 mm from the surface of the skin for about 12 minutes or the collimated source in the source housing, with the housing in contact or close to the surface of the skin for about 19 minutes.

In both cases at greater source skin distances, the radiation dose profiles are too flat to have caused the observed damage. For distances closer than 10 mm from the unshielded source, the dose to the centre of the lesion would have been too high to be consistent with the speed of development of the injury, or if the exposure time was reduced the dimensions of the injury would have been smaller than observed.

The average whole body dose was calculated at 15 rads for both cases.

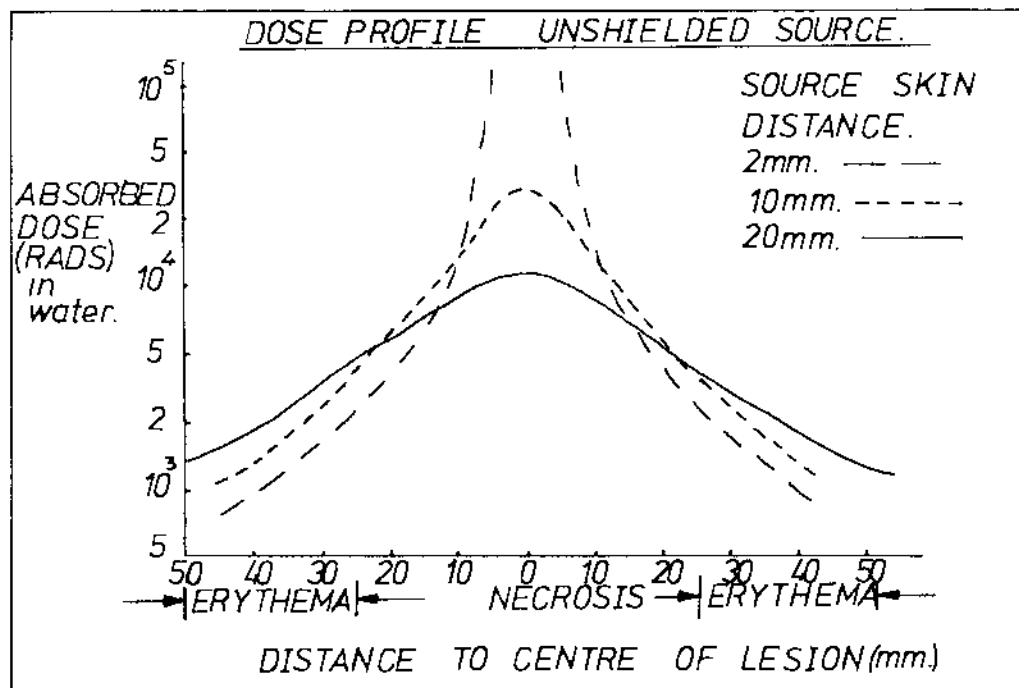


Figure 5. Surface dose profiles for the unshielded source normalised to a dose of 2,000 rads at a depth of 30 mm of tissue in the centre of the lesion.

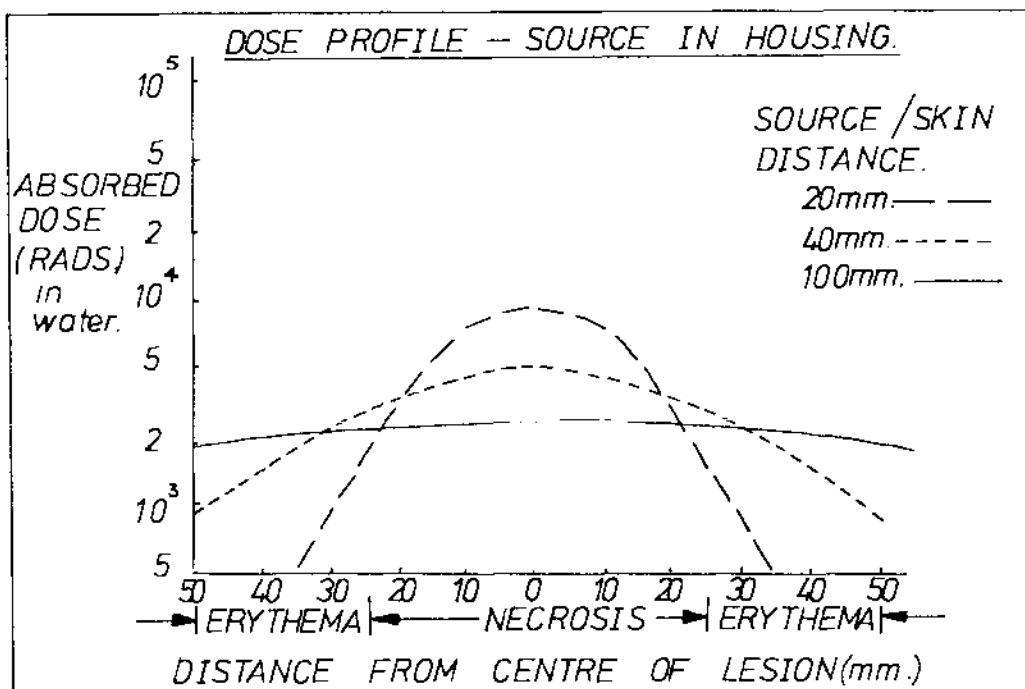


Figure 6. Surface dose profiles for collimated source in the source housing normalised to a dose of 2,000 rads at a depth of 30 mm of tissue in the centre of the lesion.

#### Biological Dose Estimates

Blood samples were taken in Glasgow and were sent in heparinized tubes to the National Radiological Protection Board, Harwell for chromosome aberration analysis. Lymphocytes from these samples were cultured for 48 hours by the mini-culture method<sup>1</sup>. Slides were prepared and 500 cells scored from each of three separate blood samples. The chromosome aberration yield is shown in table 1.

Date	Cells Scored	Damaged Cells	Dicentric	Centric Rings	Acentric Aberration
December 1969	500	43	50	2	19

Table 1. Chromosome aberration levels in blood samples.

Only cells with 46 centromeres are included in these data<sup>1</sup> and in cells where one or more dicentric were found associated fragments were also present.

The estimate of equivalent whole body dose based on the dicentric yield for this sample was 90 rads. This value was obtained from unpublished data on the dicentric yield following acute and chronic exposure of blood samples to cobalt-60 gamma radiation.

As the exposure must have been extremely non-uniform the distribution of dicentric among cells was examined as suggested by Dolphin<sup>2</sup> and is given in table 2.

Date	Cells Scored	Normal	Distribution of Dicentric				
			0	1	2	3	4
December 1969	500	457	476	20	3	0	1

Table 2. Distribution of dicentric among scored cells.

In the first sample one cell was found with four dicentric which is an unexpected finding at low dose levels. In calibration experiments a mean yield of four dicentric per cell denotes a dose of about 1,000 rads and the observation of one cell in 500 with this amount of damage suggests that a small fraction of the body's lymphocyte population received a particularly high dose.

#### Discussion

Detailed questioning of the radiographer failed to reveal any suggestions as to the possible causes of the chest injury. Any intimate contact with a sealed source either in or removed from the protective source housing was categorically denied. It was therefore necessary to consider as a theoretical exercise what sources have been available to him and which of these could have been misused so as to cause the injury in question.

These considerations led to the two possibilities mentioned earlier concerning the large radiography source exposed either directly to the chest wall or with the source in the housing in contact or close to the chest wall. The calculations and observed damage did not allow any distinction to be made between these alternatives.

It is possible to make suggestions as to how these exposures might have taken place. These range from the unshielded source held close to but not in contact with the chest, or in the top breast pocket of a loose fitting shirt, to holding the source housing close to the chest or even lying down beside the housing.

The reconstruction of the car incident indicated good agreement between the expected film badge dose and the actual dose recorded lending a degree of confidence to the reconstruction and therefore in the estimate of average whole body dose of 45 rads. The contribution to this dose from the chest injury was about 15 rads yielding a total of 60 rads.

The estimate of whole body dose may be directly compared with that made by analysis of chromosome aberrations at 90 rads. This higher value may be expected due to the effect on the yield of dicentric when small volumes of tissue are irradiated at higher doses.

#### Conclusions

The car incident undoubtedly occurred, contributing about 45 rads to the average whole body exposure. The chest injury probably added a further whole body exposure of about 15 rads.

The physical dimensions of the main injury appear to restrict the possible explanations to those discussed in this paper. All of the suggestions for the mechanisms of the exposure were completely denied by the radiographer.

The conclusions are somewhat unsatisfactory in that it is not possible to offer a firm explanation agreed by all parties.

Under these circumstances it is difficult to say what lessons can be learnt from this accident. Large radiography sources can be exceedingly dangerous if incorrectly used. It is unwise to make such sources freely available to untrained, unsuspecting persons and leave them almost completely unsupervised. Adequate instruction is vital if radiation injury is to be avoided and some form of certification of competence would go a long way towards solving this problem.

#### Acknowledgements

Very sincere thanks are due to many medical and scientific staff who helped in the reconstruction and management of this difficult and taxing case. The authors would like to mention particularly Mr. T. Gibson, Mr. G. C. Jardine and Mrs. Barbara Stephenson.

#### References

1. Parrott, R.J. and Lloyd, D.C.  
The Study of Chromosome Aberration Yield in Human Lymphocyte as an Indicator of Radiation Dose. 1 - Techniques.  
NRPB-R2, National Radiological Protection Board, Harwell.
2. Dolphin, G.W.  
Biological Dosimetry with Particular Reference to Chromosome Aberration Analysis.  
In: Handling of Radiation Accidents, p.215, I.A.E.A. Vienna 1969.

## LARGE RADIATION EXPOSURE

Vidya Bhushan  
Department of Medical Physics  
Safdarjang Hospital, New Delhi-16, India

### ABSTRACT

A case of large inadvertent radiation exposure to a person concerned with the replacement of a decayed  $^{60}\text{Co}$  source by a new source in a teletherapy unit is described. An estimated exposure of 800 rads was received by the person while pushing the source in the source head of the unit with the hand in the vicinity of the source itself. The case history is discussed.

### INTRODUCTION

A high radiation exposure took place in May 1967 at Safdarjang Hospital, New Delhi due to a planned deviation from an accepted procedure. This involved an acute local exposure to the hand far beyond the limits of maximum permissible doses to the hand and the skin<sup>1</sup>.

### $^{60}\text{Co}$ SOURCE TRANSFER AND EXPOSURE

The loading, shipping and replacing sources by means of the Atomic Energy of Canada Ltd. 'Drawer' system ensures adequate simplicity and complete safety in all phases of source handling. The source shipping and transfer container has two drawers known as source drawer and dummy drawer. The cobalt beam therapy source is locked in the source drawer in a cavity in the center of a solid lead or lead-tungsten alloy drawer during the shipping and the transfer phase. The source head and the transfer container are aligned and the decayed source is withdrawn in the slot of the shipping container in place of the dummy drawer. Immediately after this, the new source is inserted as per usual procedure in the source head of the equipment.

However, it was observed that the source was not sliding into the source head probably due to disturbed alignment. It was at this time that a worker planned specially to insert the source manually by wearing lead gloves in hands as the only measure for radiation protection. The person who received large exposure had to hold the source drawer for a short time with his right hand palm in contact with the unshielded side of the source fixed in drawer itself. Thus the source in the source drawer was manually inserted in the source head without involving any other accident. It appears that the

hand of the person with complaints of the radiation burn received a heavy instantaneous localised exposure of 8000 rads during the span of 10 seconds.

#### ESTIMATION OF DOSE

We are interested in the estimation of the dose in the vicinity of the source which has to be therefore of finite size and the decrease of intensity is to be less rapid with distance in comparison to the case for a point source. As the irradiated hand has been covered with lead gloves and kept in contact with heavy solid medium of high atomic number and the surface of the sealed source, it appears therefore that the uniformity of the dose in the palm skin and its vicinity will be limited by inherent characteristics of the radiation source and the medium in contact with the skin.

Exposure to the hand in the geometrical confine of the beam approximates to the case of uniform irradiation of class A<sup>2</sup>. The portion of the hand irradiated by the direct beam specifically in the geometrical confine of the beam should have received a uniform dosage mainly due to high quantum energy of the cobalt gamma radiation and due to thinness of the object irradiated. We may approximately calculate exposure at the mid point of the thickness of the irradiated site according to the inverse square law if we do not consider contributions from the scattered photons or the bremsstrahlung from the source and its surroundings. Exact amount of dose in the vicinity of the source would largely depend upon the duration of exposure and the distance of the hand from the source. An approximate evaluation of the dose rate at a distance of 3 cm from the source surface to the center of the hand along the axis of the beam is as follows:

Exposure rate in air at one meter  
from the source = 45 R/min

Dose rate in air to small mass of  
soft tissue at 3 cm from the source = 800 rads/sec

A detailed and accurate distribution of the dose in the confines of the beam and around it is under study and would be reported later on.

#### BRIEF POST-EXPOSURE HISTORY OF THE CASE

The person having received high exposure during the source transfer, complained about radiation burn within two weeks of the incident. He complained just after the source transfer a feeling of burning in the affected right hand which disappeared by the same day. Later his exposed hand did not give any feeling of inconvenience for about one week. Suddenly, after twelve days of exposure, he complained about tremendous burning sensation, itching and pain in the palm of the affected hand. He had been therefore hospitalised for about two days where his hand was given hot water fomentation. Immediately after discharge from the hospital, he developed a big boil of reddish colour at the place of the exposure giving unbearable pain and burning sensation. Some temporary relief was obtained when the boil was opened. However, thereafter his hand had been under various types of treatments at various places which involved plastic surgery and physiotherapy.

## RADIATION EFFECTS

In the present case, there appears to be negligible exposure to the gonads and as such we will not consider hazards from a genetic stand point. As is well known the radiation effects can be classified into several categories depending upon the magnitude of the dose, extent of exposure, etc. They can be acute, chronic or acute-chronic effects.

The case under reference appears to have received therefore an acute and chronic exposure. In the above case, it is interesting to note that acute radiation effects would be pronounced firstly due to short period of time involved and secondly due to extremely large amount of local exposure which is even far beyond the normal tolerance level of skin and subcutaneous tissues of human beings. This influence of dose rate applies particularly to radiations of low LET which are commonly encountered in occupational exposure at the present time.

### RELATIVE INTEGRAL DOSE

We have shown earlier<sup>3</sup> that relative integral dose (to be referred subsequently as R.I.D.) instead of the whole body integral dose could be a better choice for explaining patient's reaction in a course of radiotherapy. On similar grounds, we can define R.I.D. in the present case as follows:

$$\text{R.I.D.} = \frac{\text{Integral absorbed dose delivered to the whole body of a worker for a given set of conditions}}{\text{Integral absorbed dose to the tissues in the geometrical confine of the radiation beam for the same set of conditions.}}$$

The R.I.D. in the present case, would approximate to unity because whole body exposure is negligible in comparison to the local high exposure. This is the minimum possible value of the R.I.D. as per its definition in any case of localised exposure. It corresponds to the minimum general clinical reactions as a result of local exposure. We have earlier encountered values of R.I.D. to be around 6 in the treatment of carcinoma oesophagus by <sup>60</sup>Co teletherapy by two pairs of opposing fields at right angles to one another planned for a tumour lethal dose of 6000 rads in six weeks.

Hence local acute effects are mainly due to high exposure at a high dose rate. This acute somatic injury is therefore related to a causative radiation exposure.

### CONCLUSIONS

It appears to be a case of acute and chronic exposure. The manifestation of acute effects is such that the relationship to the causative radiation exposure is obvious. The low value of R.I.D. confirms absence of general clinical reactions as a result of exposure. The severity of local effects is likely to show up also in terms of late effects.



It would be interesting to study chromosomal aberration frequency in order to confirm the present estimate of dose. In man, the extrapolation of chromosome aberration to dose can be made from the peripheral blood lymphocyte/leucocyte system<sup>4,5</sup>.

#### ACKNOWLEDGEMENTS

The author is grateful to Dr.A.R.Gopal-Ayengar for taking keen interest and useful discussions. Thanks are also due to Dr.A.K.Ganguly and Dr.K.G.Vohra for their kind interest and encouragement. It is a pleasure to acknowledge for the official support provided by the Medical Superintendent, Safdarjang Hospital, New Delhi for presentation of this paper.

#### REFERENCES

1. Recommendation of the International Commission on Radiological Protection(Adopted September 17,1965), I.C.R.P.Publication 9.
2. Recommendation of the International Commission on Radiological Units and Measurements, Report 10 e, 1962. Handbook 88, Nat.Bur. Standards, U.S.A.
3. Bushan, V.: Integral Dose in Radiotherapeutic Practice, Indian J. Radiol., 1967, 21, 48-53.
4. M.Babu, K.P.George, K.Sundaram and A.R.Gopal-Ayengar. Radiosensitization by Iodoacetic acid on human peripheral blood lymphocytes.HHL/WP. 13. WHO Meeting of Investigations on chromosome aberration analysis as a biological indicator of environmental effects, December 4-8,1972, Mol. Belgium.
5. K.P.George, M.Babu, K.Sundaram and A.R.Gopal-Ayengar. Radiation induced chromosome aberrations in human peripheral blood lymphocytes in culture and the effect of some chemicals in altering its sensitivity., IARP, First Annual Conference April 12-14,1973, Bombay (Abstract).

CLINICAL COURSE AND DOSIMETRY OF ACUTE HAND INJURIES TO INDUSTRIAL  
RADIOGRAPHERS FROM MULTICURIE SEALED GAMMA SOURCES

Eugene L. Saenger,  
James G. Kereiakes  
University of Cincinnati College of Medicine  
Cincinnati, Ohio

Neil Wald  
School of Public Health, University of Pittsburgh  
Pittsburgh, Pennsylvania

George E. Thoma  
St. Louis University Medical School  
St. Louis, Missouri

Abstract

In the past several years there have been a number of incidents in which industrial radiographers have received severe radiation injuries to the fingers from inadvertent contact with sealed sources of such nuclides as  $^{192}\text{Ir}$  and  $^{60}\text{Co}$ . These injuries are characterized by an initial severe reaction progressing from erythema and edema to bulla formations at sites of contact with relatively little reaction at distances of 1-3 cm. from the point of contact. Pain is relatively little in the absence of infection. There is often spontaneous resolution of the lesion over a period of 6-8 weeks. Plastic surgical repair depends on the severity of the lesion.

Dosimetric measurements and calculations indicate that doses at the surface are in the kilo R range per Ci and at 1 cm. are 0.1 to 0.01 of these surface doses. The unusually high surface dose is due in part to electrodeposition in the capsule wall. The gamma absorption in tissue is 5-10% per cm. The inverse square law is the principal factor accounting for the relative lack of effect below 2 cm.

Introduction

During the past decade, there have been a number of severe injuries to the fingers and hands of industrial radiographers resulting from inadvertent exposure and sometimes direct contact with multicurie sealed sources. The frequency of such injuries is very low in relation to the widespread use of industrial radiography using radionuclide sources. When these injuries do

occur, they can result in interference with or termination of the worker's specific duties as a radiographer. Because of the peculiar geometrical relationships resulting from direct contact with these sources, the nature and course of the injury is somewhat different from that customarily encountered with x- and gamma-rays in brachytherapy, external beam therapy or in radiation accidents where the source to subject distance is greater.

There are several reasons for the occurrence of these hand injuries. Occasionally, the sealed source becomes detached from the cable which returns it to the housing. At times the cable fails to return the source to the housing. Or, the technician, neglecting to use his survey meter, may forget that the source is not in its housing at the time he is changing film or positioning the cable. In these instances, there seems to be a break in the previously well understood routine of the radiographer thus leading to the injury.

A useful classification of skin and hand injury<sup>1</sup> is as follows:

Type I. Erythema (only) is equivalent to a first degree thermal burn or a mild sunburn. Some time after exposure, a sensation of warmth or itching may occur; the redness, however, can appear as late as 2-3 weeks after exposure, the length of the symptomless interval depending on the dose. Medical care is not necessary and ability to work is not usually impaired. Dry desquamation (scaling) occurs.

Type II. Transepidermal injury (wet desquamation) is equivalent to the injury seen in a thermal burn of the second degree. After the erythema develops, bullae form and break open leaving open, painful wounds vulnerable to infection. After exposure, early itching is noted. Subsequently, tenderness and swelling are experienced. The hand becomes stiff and finger motion is often painful. The symptom-free latent period is shorter than in the Type I lesions, and blisters appear within 1-3 weeks depending on the dose. Recognizable injury of this grade requires a skin dose above 1,000 rads. These lesions usually heal with conservative treatment. The new skin is usually thin and easily injured, and the fingers are tender to pressure and rapid and marked temperature change.

Type III. Dermal radionecrosis is a more serious degree of the Type II lesion caused by doses of radiation above 2,000 rads. This level of injury has been observed in persons after accidental exposure of hands to the direct beam of an electron accelerator and in those who handled multicurie sealed sources, fresh fission product material or targets in which radioactivity was induced during laboratory experiments by neutron or electron bombardment. The lesions resemble those caused by a severe scalding or chemical burn. Pain and edema occur within hours or a few days and is intense. Medical abatement of pain is urgently needed. The skin doses responsible for burns of this severity are probably in the range of 3,000 rads or greater.

Type IV. Frequently repeated or continuous exposure of the skin to x-rays, gamma rays, or beta rays over a period of months to years causes an eczema-like condition. Once it has developed, it seldom heals completely and ulceration frequently occurs. Skin cancer occurs in a large (but unknown) proportion of such cases of chronic radiation dermatitis.

Our interest was drawn to this group of patients because of the appearance of severe superficial changes and a relative absence of the deep tissue changes noted with the previously encountered injuries resulting from x- and gamma radiation produced by external radiation sources. Nine cases form the basis of

this report. In all cases, there have been exposure to or during the fabrication of sealed sources either emitting primarily gamma radiation or, in two cases, beta radiation.

The circumstances of exposure have been somewhat varied. In 6 cases, there has been a single acute exposure recognized at the time of occurrence. In 3 cases, the exposures have been chronic and unrecognized until the eventual development of clinical changes.

Table 1 presents the dose estimates based on calculations by one of us (JGK) and published originally in NCRP Report 40.<sup>2</sup> Subsequently, values were measured near discrete sources using TLD chips and these observations indicate that the previously published ones should be reduced by about 20% (See Appendix I). Admittedly, the estimate of exposure time even in the acutely exposed individuals is difficult to estimate. In some cases, re-enactment of the exposure incident provided some clarification (cases 1, 2, 4, 6, 7). In the other cases, estimate both of doses and times have been adjusted in large part on the basis of rate of change of the clinical manifestations. Because of the very rapid change in dose with distance, the doses are at best approximations of the many variations in exposure to the different parts of the hand. Yet, as can be seen from Table 2, it is possible to group these few cases in order of severity.

Table 2 further demonstrates the progression of the radiation injury by indicating the days on which particular manifestations of injury were first noted either by the patient or physician. These patterns reveal a mild degree of inconsistency characteristic of human data, particularly in radiation exposure cases. Nevertheless, the most severely injured patients show an earlier onset, more changes, and more rapid progression of lesions as would be expected. Also, in these cases, the required treatment is more elaborate.

An important facet of these cases is the absence of systemic effects of the acute radiation syndrome. In spite of the intense local injury, there has been no clinical or laboratory evidence from peripheral blood counts of the acute radiation syndrome. Five of the patients (cases 1, 3, 4, 5 and 6) have had chromosome cultures of peripheral blood lymphocytes. In patient 1, the data suggested a whole body dose ranging between 70 and 100 rad from one culture at 6 months. Hematological findings had been quite normal from the 14th day on. In the other cases (cases 3, 4, 5 and 6), chromosome cultures were normal or suggested subclinical exposure.

### Clinical Observations

The initial symptoms in the acutely exposed cases expectedly varied somewhat. The most frequent complaint was a superficial feeling of irritation, tenderness or itching which was sometimes transient. Restricted motion or stiffness of the finger was noted early in a few instances. Erythema, edema and diminished sensation were noted subsequently. In the absence of ulceration and infection and with immobilization, pain was minimal or absent. The development of vesicles, bullae and frank ulceration was associated with increased pain in proportion to the size and depth of the lesions and the presence of secondary infection. An important and characteristic observation is the rather sharp demarcation of bullae and ulcers indicating the specifically high local doses unique to these sources. Frequently, the unirradiated opposing surface

of the extremity appeared entirely normal at least in the acute stage. Epilation, a valuable dosimeter, commonly observed after acute radiation doses of 300 - 1,000 rad of beta or gamma radiation, was not noted in any of these cases and has not been mentioned in the literature on this specific site of injury. Dry epidermitis or wet epithelitis usually developed 2 - 3 weeks after exposures of 1,000 - 4,000 rads. Restricted motion was an important finding in the acutely exposed being present (cases 1, 2, 4 and 5). It was associated with marked local edema and possibly with some reaction of tendon and synovial surfaces. In some cases, the finding persisted and worsened even after recession of the edema.

The initial findings and course in these patients have been remarkably similar to other scattered case reports. Evans and Orr<sup>3</sup> reported a case of localized radiation injury from an unidentified beta-gamma source with an estimated exposure of 1,200 - 1,400 rads due to operator error. There was initial discomfort and numbness at 8 days and subsequent erythema and bulla formation at 13 days and eventual spontaneous healing. Maxfield and Porter<sup>4</sup> reported one case of localized hand damage from a 70 Ci iridium source with a clinical course similar to that of case 3 above. No finger doses were estimated although bullae formed at 18 days and healing began at day 27. Treatment was conservative.

Several patients receiving very high doses to the hands have been reported in the literature. The patient with four-extremity radiation necrosis from an accelerator accident<sup>5</sup> developed mild erythema beginning on day 1 progressing to bullae formation on day 10 to complete superficial desquamation and gangrene at day 32 and with continuing progression necessitating bilateral serial amputation between 3 and 17 months. In the report of case 1 receiving 20,000 - 40,000 rem at Los Alamos,<sup>6</sup> brawny edema began in 30 minutes and both upper extremities were severely involved within 36 hours. Death occurred on the 25th day. In the Argentinian case involving the loss of both lower extremities from radiation necrosis,<sup>7</sup> the hand doses were probably of the order of 1,000 - 3,000 rads. The patients reported in this paper represent less serious injury.

The effect of dose rate in these patients may be compared to the changes suggested from Strandqvist plots produced by orthovoltage x-rays<sup>8</sup> (See Table 3). Necrosis occurs with doses of 3,000 r given in a single dose, 3,800 r divided into two doses, and 4,400 r divided into 3 doses at daily intervals. Low Beer<sup>9</sup> (See Table 4), using plaques impregnated with <sup>32</sup>P, gives values for threshold erythema of 143 r, dry desquamation of 1,700 r and bullous epidermitis of 7,200 r. In our series, except for case 3, the observed changes were undoubtedly affected to a considerable degree by the extremely high superficial dose rates (Table 1).

### Treatment

Initial treatment has been conservative utilizing standard principles of surgical cleanliness and avoidance of additional trauma. Manual work and radiation exposure should be avoided. Opinion has varied in regard to evacuation of bullae. Some surgeons have evacuated them; others have not done so. As long as no infection develops, it would seem immaterial as to which course is followed. In the presence of frank ulceration and necrosis, various local antibiotic and steroid containing cremes have been applied. Covering and immobilization of clean surfaces has resulted in abatement of pain. In the

presence of progressive ulceration and necrosis, the use of split thickness grafts as advocated by Krizek and Ariyan<sup>10</sup> has relieved pain and hastened healing. As noted by them and in case 1, there was remarkable cessation of pain at the completion of this procedure. In cases 2 and 3, appropriate skin grafting resolved the late tissue breakdown developing many months after exposure.

Late effects which may be expected are those resulting from insidious but progressive tissue atrophy, fibrosis and chronic radiodermatitis with tissue breakdown. The hands and fingers may exhibit increased sensitivity to temperature changes and to certain roughened or metallic surfaces. There may be cartilagenous atrophy and also other changes of joint spaces and tendon sheaths leading to progressive limitation of motion.

Finally, the development of skin cancer must always be watched for. Radiation-induced cancer has not been observed in the absence of radiodermatitis.<sup>11</sup> If such dermatitis is severe or progressive, skin grafting should be seriously considered. Amputation is needed only with necrosis and failure of skin grafting.

Since the clinical course of this peculiar type of radiation injury may occasionally be obscure, the clinical evaluation must be carefully planned. Table 5 presents the studies found to be most useful. Routine hematological tests and chromosome analysis will estimate whole body doses. The serial ophthalmologic evaluations with slit lamp are suggested to allay apprehension concerning effects on the lens of the eye. As in all cases of radiation injury, the availability of prompt and continuing medical care and a well organized plan of rehabilitation will do much to allay the obvious concerns of the injured man and his family during a difficult period.

Prevention of this type of injury can only be attained by continuing education of industrial radiographers. This group of workers usually have only a high school or technical school education and no trade journal or society providing national intercommunication. The responsibility for prevention rests almost exclusively with plant management, radiation safety officers and health physicists to provide the necessary continual educational stimulus. In some of our cases it would seem that continuing personal contact with the individual radiographer is desirable since there is a tendency for these accidents to occur when the operator becomes excessively preoccupied with his personal emotional problems or interests at which times his concern for radiation safety should be increased. The diligent use of self-reading pocket dosimeters, film badges or TLD monitors, and portable survey meters are essential for adequate protection.

Equal attention in regard to radiation safety need be paid to uninstructed helpers usually obtained from unskilled labor pools. Such workers should not participate in radiographic work unless constantly attended and supervised by the qualified radiographer and wearing appropriate dosimeters.

### Summary

Radiation injury from sealed sources of multicurie level are found chiefly in industrial radiography. The lesions are characterized by intense superficial reactions and a very rapid fall off in dose with increase in tissue depth.

It is possible for the lesion to heal satisfactorily if it is only superficial; in other respects, the treatment is similar to the more usual radiation lesion where the radiation dose is more uniform to a greater depth.

#### Acknowledgment

Permission of Dr. Joseph W. Kraut, Santa Clara Valley Medical Center, Radiation Therapy Center, San Jose, California, to use case 1 is gratefully acknowledged.

#### References

1. Saenger, E. L. (ed): Medical Aspects of Radiation Accidents. A Handbook for Physicians, Health Physicists and Industrial Hygienists. United States Atomic Energy Commission, 1969.
2. National Council on Radiation Protection and Measurements: Protection Against Radiation from Brachytherapy Sources. NCRP Report 40, March 1, 1972.
3. Evans, J. C. and Orr, R. G.: Report on over-irradiation of a research worker's hand: Physical and clinical dosimetry. In Handling of Radiation Accidents, proceedings of a symposium, Vienna, 19-23 May 1969, International Atomic Energy Agency, SM-119/6, pgs. 431-435 (abstract), 1969.
4. Maxfield, W. S. and Porter, G. H.: Accidental radiation exposure from iridium-192 camera. In Handling of Radiation Accidents, proceedings of a symposium, Vienna, 19-23 May 1969, International Atomic Energy Agency, SM-119/64, pgs. 459-467 (abstract), 1969.
5. Schenck, R. R. and Gilberti, M. V.: Four-extremity radiation necrosis. Surgical management. Arch Surg. 100:729-734, June, 1970.
6. Hempelmann, L. H., Lisco, H. and Hoffman, J. G.: The acute radiation syndrome: A study of nine cases and a review of the problem. Ann Int Med. 36:2, part 1, February, 1952.
7. Beninson, D., Placer, A. and Vander Elst, E.: Estudio de un caso de irradiacion humana accidental. In Handling of Radiation Accidents, proceedings of a symposium, Vienna, 19-23 May 1969, International Atomic Energy Agency, SM-119/35, pgs. 415-429 (abstract), 1969.
8. Strandqvist, M.: Studien uber die kumulative wirkung der rontgenstrahlen bei fraktionierung. Acta Radiol. Suppl. 55:1-300, 1944.
9. Low Beer, B. V. A.: External therapeutic use of radioactive phosphorus. I. Erythema studies. Radiology 47:213-222, 1946.
10. Krizek, T. J. and Ariyan, S.: Severe acute radiation injuries of the hands. Report of two cases. Plastic and Reconstructive Surgery. 51:1, 14-22, January, 1973.
11. National Academy of Sciences - National Research Council: The Effects on Populations of Exposure to Low Levels of Ionizing Radiation. A report of the Advisory Committee on the Biological Effects of Ionizing Radiations, Division of Medical Sciences, November, 1972.

Table 1

## DOSE ESTIMATES

Subject	Radionuclide	Activity	Location	Dose Rate rad/min*	Estimated Time of Exposure	Range of Estimated Doses rads**	
1	$^{192}\text{I}$	38 Ci	surface	23850	1 - 3 min	23850 - 71550	
			1 cm tissue	1262			1262 - 3786
			3 cm tissue	162			162 - 486
2	$^{60}\text{Co}$	48 Ci	surface	76891	3 sec	4000	
			1 cm tissue	4224			222
			3 cm tissue	593			31
3	$^{60}\text{Co}$	10 mCi	surface	20.0	6 - 29 mos a few minutes per day	5200	
			1 cm tissue	1.1			266
			3 cm tissue	0.15			39
4	$^{192}\text{I}$	83 Ci	surface	52093	10 - 20 sec	7800 - 16000	
			1 cm tissue	2755			411 - 84.4
			3 cm tissue	353			52 - 107
5	$^{192}\text{I}$	70 Ci	surface	43934	< 2 wks acute and protracted	<4000	
			1 cm tissue	2324			211
			3 cm tissue	298			29
6	$^{192}\text{I}$	93 Ci	surface	58370	1.5 - 2.5 sec	1460 - 2432	
			1 cm tissue	3087			77 - 129
			3 cm tissue	395			9.9 - 16.5
7	$^{60}\text{Co}$	48 Ci	surface	76891	< 1 sec	1000	
			1 cm tissue	4224			55
			3 cm tissue	593			8
8	$^{90}\text{Sr}$	?	[Unknown exposure 5 years before			>1000	
9	$^{90}\text{Sr}$	?	clinical changes]			>1000	

\* Values from Appendix I

\*\*These values are derived from estimates of time by patient, re-enactment of the episodes and comparisons of clinical manifestations by the authors. Since the doses to the parts of the hands vary so greatly, these ranges embrace maximal and mean doses.



Table 2

## CLINICAL OBSERVATIONS

(in approximate order of decreasing severity of manifestation)

Pt.	Highest Dose (rads)	Cause	1st Symptom	Superficial Irritation, tenderness, Itching	Erythema	Swelling, Edema	Decreased Sensation	Dry Epidermitis, Scaling	Wet Epithelitis, Vesiculation, Bullae	Restricted Motion	Ulceration	Therapy	Time of Grafting
1	23850-71550	Operator error	7	7	14	10	14	17	14	17	14	graft	49 days
2	4000	Camera mal-function	4	15	15	10		16	16	5	15	graft	13 months
3	5200	Mal-function and operator error	30*	30*	30		30		33		15 months	graft	15 months
4	7800-16000	Operator error	0	11	0(?)	11			18	11		conser-vative	
5	<4000	Mal-function and operator error	7*	15	15	12			16	11		conser-vative	
6	1460-2432	Operator error	1	15	15	19	14		15			conser-vative	
7	1000	Camera Mal-function	15	15	15	-						conser-vative	
8	1000	Operator error	<150	<150	-	-					5 years	conser-vative	
9	1000	Operator error	<150	<150	-	-					5 years	conser-vative	

Day 0 is the day of exposure

\*Because of protracted exposures and one or more acute episodes, the estimate of the day of occurrence is not exact.

Table 3  
 CHANGES PRODUCED BY FRACTIONATION OF X-RAY DOSES  
 ACCORDING TO STRANDQVIST<sup>8</sup>

Effect	Dose Needed If Given Over The Noted Interval To Produce The Same Effect		
	Single Dose	2 Doses in 2 Days	3 Doses in 3 Days
Erythema	1000	1280	1490
Dry Epidermitis	1600	2000	2700
Healing of Skin Cancer	2450	2800	3250
Necrosis	3000	3800	4400

Table 4  
 EFFECT OF <sup>32</sup>P BETA RADIATION PRODUCED BY 5 CM. DIAMETER PLAQUES  
 ON HUMAN SKIN<sup>9</sup>

Effect	Dose Rate (uCi hr./sq. cm.)	Total Dose (R)
Threshold Erythema	34	143
Dry Desquamation	2000	1700
Bullous Epidermitis	4400	7200

Table 5  
 USEFUL STEPS IN CLINICAL EVALUATION OF HAND INJURY

- History and Physical Examination
- Serial Blood Counts
- Chromosome Analysis
- Re-enactment of Accident
- Frequent Color Photographs
- Baseline Extremity X-rays
- Ophthalmologic Examination with Slit Lamp

Appendix I

APPROXIMATE GAMMA-RAY DOSE RATES TO THE HAND FOR 1 CURIE IN A SEALED SOURCE<sup>a,b</sup>

Nuclide	Beta max (principal) MeV	Gamma (principal) MeV	R/mCi-h at 1 cm	Surface Dose Rate <sup>c</sup> R/min	Dose Rate at 1 cm tissue depth R/min	Dose Rate at 3 cm tissue depth R/min
<sup>137</sup> Cs	0.51, 1.2	0.662	3.26	396	22	2.9
<sup>60</sup> Co	0.31	1.17, 1.33	13.00	1602	88	12.4
<sup>192</sup> Ir	0.67	0.468	4.80	628	33	4.2
<sup>226</sup> Ra	0.4-3.2	0.047-2.4	8.25	1011	56	7.5

<sup>a</sup>Industrial source housings are usually of stainless steel and for the purpose of the calculations, the activity is considered to be a point source. In considering these dose estimates, there is assumed a capsule of outside diameter 1/4 inch, with a wall of stainless steel (type 304) which is 1/32 inch thick.

<sup>b</sup>Modified from Table 6, Appendix B, NCRP Report 40<sup>2</sup> based on TLD measurements

<sup>c</sup>The total surface dose rate for the <sup>226</sup>Ra source is 1900 R/min based on a 45 percent increase due to electron production in the stainless steel wall. For the other nuclides given in the table, the increase in surface dose rate due to electron production in the stainless steel wall is estimated to be between 25-45 percent.

EVALUATION AND DOSIMETRY OF ACCIDENTAL  
EXPOSURES TO AN X-RAY ANALYSIS BEAM.

I.S. Leith and W.A. Cornelius

Australian Radiation Laboratory  
Melbourne, Australia.

(Accidental exposure of two research workers to an X-ray analysis beam during two days resulted in large area burns to the arms and chest. The dosimetry performed was later corroborated by the clinical findings.)

---

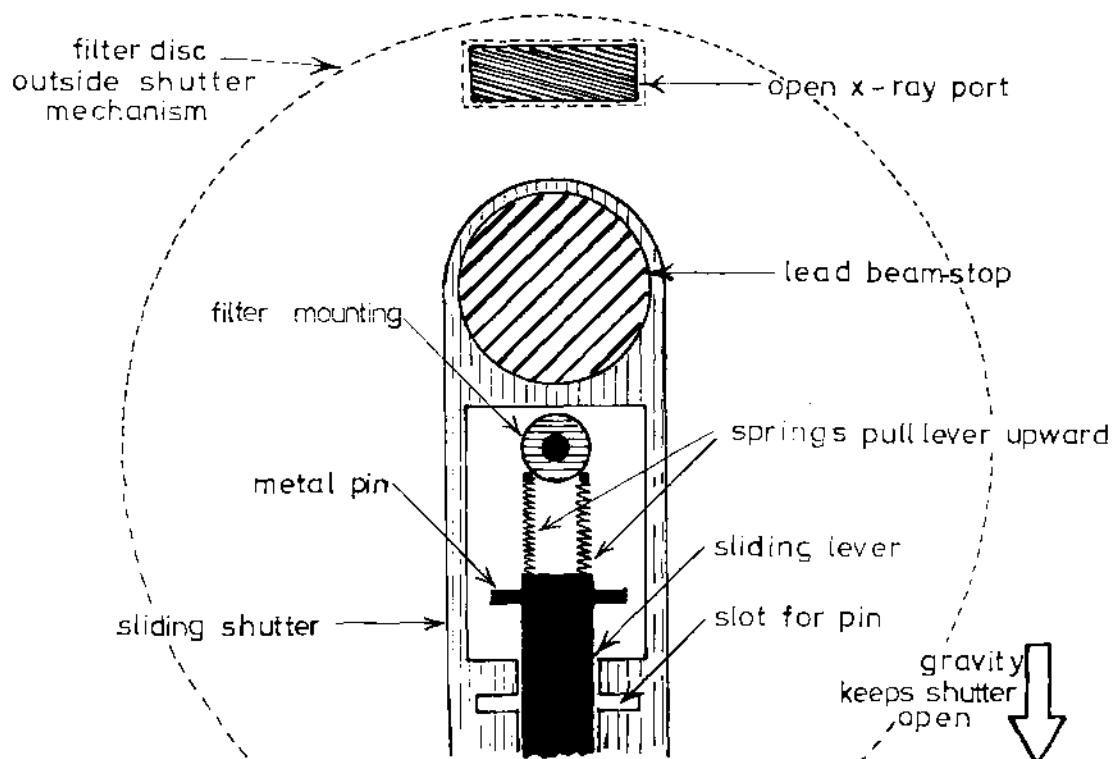
Introduction

During the period 23rd to 25th June 1970, three persons were inadvertently exposed to low-energy radiation in a direct X-ray beam from a mis-assembled X-ray analysis unit at an established research institution. Late on 25th June 1970, the assistance of the Australian Radiation Laboratory was sought following report of an erythema by one of the exposed persons and discovery of the X-ray beam by another. Measurements made by the authors on the following day indicated that three persons may have received radiation doses sufficient to lead to injury. The Director of the Australian Radiation Laboratory arranged for examination of the three persons by a medical specialist in therapeutic radiology. Two of the exposed persons exhibited extensive erythema over a period of several weeks which was consistent with the doses estimated by the authors. The third exposed person showed no erythema and this also was consistent with the estimated doses.

Conditions of the Accident

The accidental exposures occurred over a period of almost three days. The exposure conditions varied during this period, which complicated the dose estimates. Investigation disclosed that three persons only could have been exposed to the X-ray beam. These persons were an experienced post-doctoral research worker, an instrument maker and a technician. Some eighteen months prior to the accident the X-ray analysis unit had been modified but subsequently had been operated on one occasion only until the days of the accident. On this single other occasion the unit was operated for 12 to 20 hours, but was largely unattended. The modification had been made to the tube housing of the X-ray unit (Philips Model PW1016 with copper target) to permit use with diffraction cameras previously used with an older X-ray unit. The X-ray tube housing had been dis-assembled and two opposite shutter mechanisms had been modified by removal of the solenoid-operated shutter opening devices and conversion of the shutters to manual operation. The remaining two opposite shutters were not modified and the standard shutter-closing mechanism was allowed to operate. This mechanism operates by means of closing springs which act on a lever which engages the shutter by means of a

metal pin (see Fig. 1). However, one of these shutter mechanisms had been dis-assembled and re-assembled incorrectly so that the lever no longer engaged the shutter. The shutter then remained permanently open under the force of gravity as illustrated in Figure 1.



**FIGURE 1. FAULTY SHUTTER MECHANISM**

The microswitches operated by each solenoid were disconnected and a warning light was connected to switches operated by the two modified shutter mechanisms only. It was possible to see that the shutter which remained open due to mis-assembly was in the open position, as the filter disc was in a no-filter position, but this was not observed.

During the accidental exposures the X-ray unit was operated with a diffraction camera at the modified port immediately to the right of the open shutter. The camera was undergoing alignment and adjustment by the research worker during the days of the accidental exposures. It proved a difficult operation for him, and he enlisted the assistance of the instrument maker who was working on modification of the same camera. The difficulty encountered by the research worker may have been due to poor eyesight. The X-ray beam was viewed by means of a hand-held fluorescent screen mounted on a stick approximately 20 centimetres long. The camera was open, allowing access to the beam path inside it.

The X-ray analysis unit incorporated a camera table below the level of the X-ray ports which were at waist level. The X-ray beams from the open shutters are emitted almost horizontally.

During the afternoon of Tuesday 23rd June, 1970, the research worker attempted to align the camera from 2 p.m. till 4 p.m. then left the room after requesting the instrument maker to continue the attempt. This the instrument maker did from 4 p.m. till 4.30 p.m., when he switched the unit off. During

these (unsuccessful) attempts to align the camera, the X-ray tube was operated at 35 kilovolts with a tube current of 10 milliamperes. In Figure 2, a diagram of the room and equipment is given, indicating the positions occupied by the persons involved in the accidental exposure.

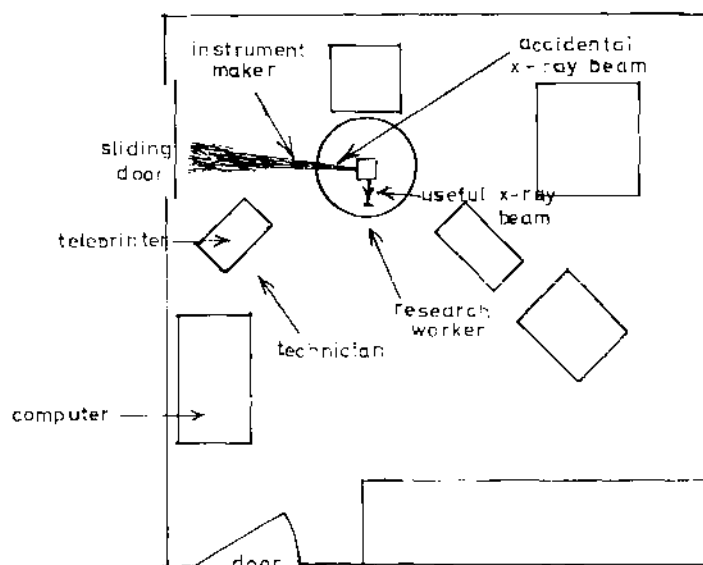


FIGURE 2. DIAGRAM OF X-RAY ROOM

The research worker took up a position in the line of the X-ray beam with which he was working during alignment, being protected from that beam by a beam stop in the camera. In this position he was not exposed to the X-ray beam from the mis-assembled shutter to his left except on repeated brief occasions when his left arm was in that vicinity. However, when his arm was exposed, it may have been fairly close to the open shutter, inside the perimeter of the camera table. During this time, he observed a faint fluorescence on the screen not caused by the beam which he was aligning but did not investigate its source.

The instrument maker, however, took up a position to the left of the camera he worked with and in this position was directly in line with the beam from the unintentionally open shutter. The beam irradiated his abdomen outside the line of the camera table at a distance of 40 centimetres or more from the open shutter throughout the period in which he worked on the camera. During the following morning, the X-ray unit was energised, using the same operating factors as before, and the instrument maker again attempted to align the camera, from 9 a.m. till 10 a.m. Due to a change of position, he was in the accidental beam for approximately half this period. The X-ray unit was then switched off.

At 5.15 p.m. on the same day, Wednesday 24th June, the research worker switched the unit on again and worked until 6.00 p.m. He took up positions mainly out of the accidental beam and was probably exposed for a maximum time of 5 minutes, again to the left arm.

On the evening of Wednesday 24th, the instrument maker observed an erythema on the skin of his abdomen in the region of the diaphragm but did not

associate this with an X-ray burn.

On Thursday 25th June, the X-ray unit was operated at increased tube factors of 45 kilovolts and 15 milliamperes, to provide higher X-ray intensities to assist in viewing the beam. The same camera as before was again worked on by the instrument maker from 9 a.m. to 10.30 a.m. but he was aligning an eyepiece and stood in line with the beam with which he was working. In this position he received little or no exposure to the accidental beam. The research worker was in the vicinity of the unit for approximately 15 minutes during this period and may have been exposed to the accidental beam for a maximum of 5 minutes. At 11.15 a.m. the instrument maker returned to the unit and worked until 12.30 p.m. on the camera mechanism. During approximately 30 minutes of this period he took up a position in the accidental X-ray beam. At 12.30 p.m. it was noticed that the phosphor screen continued to glow brightly when removed to the left from the useful X-ray beam in the camera. The accidental X-ray beam and faulty shutter mechanism were then discovered. The technician, who had occupied a position in the room, but away from the X-ray unit and not in line with the accidental beam until this time, then exposed his fingers to the accidental beam for approximately 15 seconds while holding the phosphor screen. At approximately 1.00 p.m. the X-ray unit was switched off. During this period of discovery the instrument maker recalled the erythema he had observed and associated it with the accidental beam.

#### Investigational Dosimetry

Late on the afternoon of Thursday 25th June, the Australian Radiation Laboratory was notified of the accident and its advice sought. Early on the following morning, the authors assembled radiation monitors and travelled to the scene of the accident to investigate and perform dosimetry. Our object was to assess the likelihood of injury to the exposed persons and provide dose estimates to assist the medical specialist who would examine the exposed persons. The research worker reported a slight erythema on his left forearm and upper arm at this time.

Measurements of the direct beam were made with a Victoreen Model 555 Radocon II Integrating Ratemeter with Timer-Trip Module. A Victoreen Model 555-100 LA low energy probe was used for the measurements. This probe is calibrated by the manufacturer for the energy range of 6 keV to 30 keV. The energy correction factors provided by the manufacturer for this probe vary from 1.02 at 8 keV to 1.05 at 20 keV. This monitoring equipment had been earlier purchased for the specific role of dosimetry of X-ray analysis beams because of the very high intensity limit (100,000 R/min ) and flat low-energy response of the 555-100 LA probe and the accuracy of the meter. The intensity limit is not sufficiently high to permit accurate measurement close to shutters under all circumstances but due to the estimated distances to the heavily exposed persons in this accident, accurate dosimetry was possible.

Measurements of scattered radiation around the room and direct radiation at large distances from the open shutter were made using a Berthold TOL/D-HF radio-frequency shielded X-ray monitor. This monitor uses probes employing gas amplification, permitting calibration over a very wide range of exposure-rate by adjustment of the high tension voltage applied to the probe. The radio-frequency shielded probe (purchased for other monitoring situations) has a more energy-dependent response at very low energies than the unshielded probes by this manufacturer, requiring a correction factor of 1.7 to be applied to readings at 8 keV.

The mis-assembled X-ray shutter produced a rectangular beam 18 centimetres wide by 5 centimetres high at a distance of 40 centimetres from the shutter. While working with the X-ray unit, the instrument maker was wearing a cotton laboratory coat, a woollen sweater, a woollen shirt and a cotton vest. Measurements were made of the exposure-rate in the middle of the accidental beam at 40 centimetres from the tube shutter with and without attenuation by material used to closely simulate the instrument maker's clothing. These measurements are given in Table 1.

TABLE 1  
Exposure-Rates Measured at 40 cm. from  
X-ray Tube Shutter

X-ray Tube Factors	Exposure Rate in Air R/min	Exposure Rate Under Clothing R/min
35 kilovolts, 10 mA	420	178
45 kilovolts, 15 mA	785	336

In Figure 3, the scattered radiation exposure-rates measured around the X-ray room are shown.

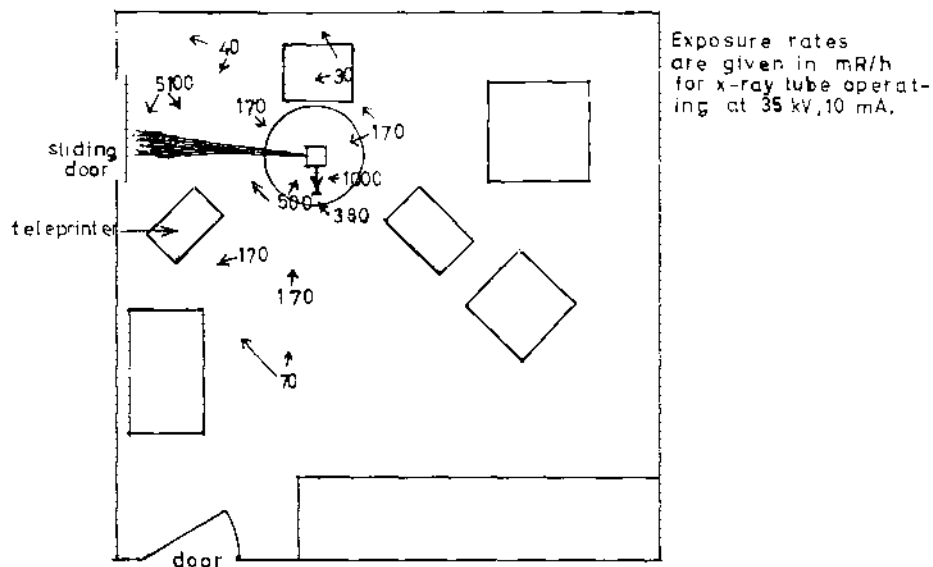


FIGURE 3. EXPOSURE RATES FROM SCATTERED RADIATION

From the measurements of attenuation in clothing and in air, an approximate calculation of the energy of the radiation in the accidental beam was made. The estimate of half-value thickness was 0.055 millimetres of aluminium, corresponding to approximately 8 keV effective. It was expected that the effective energy of the X-ray beam would not be significantly different from that of the characteristic radiation from the copper target



(i.e. approximately 8 keV) because of the thin window of the X-ray tube.

### Dose Estimates

The accuracy of estimates of dose in this accident is largely dependent on estimates of exposure time and distance. Distances to exposed persons were fairly constant and mostly relatively large. Persons were exposed mainly in regions where exposure-rate is not a strong function of distance. Errors due to distance uncertainty are thus not of major importance. However, errors due to exposure time uncertainty are potentially very large. In this case, significant reduction of this uncertainty was shown to be possible and the value of patient and tactful enquiry, retracing the accident history several times to improve recall, was proven. As a result, initial estimates of duration of exposure made by the exposed persons were very much reduced. For example, initially periods of exposure were estimated as starting or stopping at the official hours of duty. These estimates were reduced firstly by time and motion study to determine the probable proportion of working time during which persons were exposed to the accidental beam. In the next stage, patient questioning and better recall by the exposed persons indicated longer than usual tea-breaks and early departures from duty, leading to the final estimates of exposure time given in this paper.

Estimates of doses at the skin surface were made based on exposure-rates measured using the Victoreen Model 555. Account was taken of the attenuation of clothing. However, the critical tissue lies at a depth of approximately 1 to 2 millimetres below the skin surface in the stratum germinativum or replicating layer. Estimates of dose at this depth were derived using a tenth-value thickness in skin of 1.80 millimetres. This value was derived for monochromatic radiation at 8 keV using the data for muscle given in I.C.R.U. Report No. 17. This coincides closely with a value of 1.85 derived by extrapolation of the data of Lubenau et al<sup>2</sup>. Thus the dose at the critical tissue is estimated to be in the range 0.28 to 0.077 of the dose at the surface. In table 2, the estimates of total exposure time and doses at the surface and at the critical tissue are given. The dose estimates are based on maximum estimated exposure times at a distance of 40 centimetres. It was believed that the estimates of doses to the research worker may have been only a small fraction of the true doses as exposure to his arm may have occurred much closer to the X-ray shutter than 40 centimetres. The dose estimates for the research worker and instrument maker were derived from exposure-rates measured after attenuation by clothing and would need to be increased by a factor greater than 2 if bare skin was exposed. The dose estimates for the technician were derived from exposure-rates measured without attenuation by clothing. The vertical width of the erythema visible on the abdomen of the instrument maker was approximately 5 centimetres, which suggested that he received most of his dose at a distance of 40 centimetres.

TABLE 2  
Estimated Exposure Times and Doses

Person	Total Exposure Time (min.)	Dose at Surface (rem)	Dose at Critical Tissue (rem)
Instrument Maker	90	19,200	1500 - 5400
Research Worker	30	5,500	400 - 1500
Technician	0.25	180	14 - 50

## Clinical Findings

The clinical examinations confirmed that the erythema which appeared on the abdomen of the instrument maker was consistent with a dose of 1500 to 2000 rem to the replicating layer of the skin. Dry desquamation appeared at 3 to 4 weeks after exposure on the abdomen and on the backs of the hands. These clinical findings indicated a dose to the hands of approximately 2000 rem.

The examination of the research worker revealed no later sequelae to the arm (which is consistent with the estimated dose) but at 3-4 weeks dry desquamation appeared on the hands and this was estimated to be equivalent to a dose of approximately 1500 rem. The specialist also noted some evidence of earlier facial injury, which he attributed to exposure to soft x radiation.

The technician showed no clinical signs of exposure, as expected.

The exposure to the hands of the instrument maker and research worker was not anticipated by their memory of their actions during the exposures. It probably occurred as a result of several brief exposures in close proximity to the open shutter but may have been due to exposure to the useful beam with which they were working. Dose estimates would be meaningless due to uncertainty of distance and exposure time and were not attempted.

## Conclusions

The conditions of the accidental exposure described in this paper lead to a number of useful conclusions. Firstly, the conditions were such that much higher doses and severe injury may well have resulted and the workers concerned were fortunate to escape severe effects. Secondly, the need for many of the principles of good practice in the use of X-ray analysis units is clearly illustrated in this accident.

The adoption of one such principle, that of adequate radiation monitoring of facilities whenever modified, would have led to early discovery of the mis-assembled shutter. The safe principles and practices had been formalised and published in Australia in the previous year, 1969 as a code of practice. This Code, entitled "Code of Practice for the Safe Use of X-ray Analysis Equipment"<sup>3</sup> was published by the National Health and Medical Research Council of Australia in its series of publications and codes of practice on radiation health. One of the authors (I.S.L.) worked on the drafting of this Code of practice. The code details general working practices and equipment features essential to ensure radiation safety with all X-ray analysis units. However, the code recognises the great variety of X-ray analysis equipment in use and in particular, the improvement in design for safety which has proceeded for many years. While proceeding from the premise that in-built safety should be relied on in preference to relying on workers to maintain good practice, the code recognises that it is unpractical to modify many older X-ray analysis units and some units used in particular configurations to the extent that working rules are not required. Hence, the code categorises X-ray analysis units according to their degree of in-built protection and grades working rules accordingly. Nevertheless, it was recognised that some units in use do not meet the minimum requirements of any of the categories of unit specified. A separate set of very thorough and restrictive working rules is laid down for such units, in an attempt to ensure safety in their use and to act as a strong incentive to improve the built-in safety of these units. The X-ray analysis unit described in this paper was a unit of this type and several of the special working rules provided would have served to prevent the accidental exposures. These include not operating the unit if a person not essential to its operation occupies the room (this would apply to the instrument maker and technician)

and not making alterations to the unit while it is operating (this would apply to the work on the camera performed by the instrument maker). Other rules include monthly radiation surveys, weekly examination of the unit for hazards and the requirement for an experienced observer to be present at all times when persons work with the unit to warn of hazards. Another rule requires that the operations of alignment or adjustment shall not be carried out by inexperienced persons unless under the direct supervision of an experienced worker. This rule would apply to the periods of exposure of the instrument maker, who had no appreciation of the radiation hazards involved. Each of these rules would have reduced or prevented the accidental exposures in this case.

There were many other elements of good practice and in-built safety not applied during this radiation incident, but certain examples of bad practice are more notable. The absence of any form of radiation survey monitoring or close examination of the unit after modification, the modification of the warning light system, the secondment of an instrument maker to work of a hazardous nature with which he was unfamiliar and in which he was not supervised and the siting of a small computer and console in the X-ray room are all good examples. We hesitate to assign such simple labels as "carelessness" to the cause of accident but a change of attitude of the research worker would appear to be essential in this case. This worker had been in the field for many years and was accustomed to working with open cameras and unshielded beams particularly when equipment design was in an early phase. However a change of attitude appears to be necessary, especially if this worker had been overexposed on one or more earlier occasions.

Another matter is illustrated in this incident. The initial dismantling of the unit was done to remove automatic shutter-opening mechanisms not provided by the manufacturer on earlier units, as these interfered with the use of cameras previously used with the older units. The same situation arises frequently when cameras from one manufacturer are used with X-ray units of another. Frequently, the built-in safety features are removed rather than construct a satisfactory part for inter-connection of the camera and X-ray unit while preserving the safety features. This illustrates an inadequate approach by many workers with these units but also points to a serious lack of planning and standardisation by manufacturers. Another question of design in this case is the particular shutter configuration used in the X-ray unit. The shutter was designed in such a way that mis-assembly was easily possible and represents a fundamental cause of the accident.

---

#### References

1. Radiation Dosimetry: X-rays Generated at Potentials of 5 to 150 kV. I.C.R.U. Report 17, International Commission on Radiation Units and Measurements. Washington, 1970.
2. Analytical X-ray hazards: a continuing problem. Lubenau J.O., Davis J.S., McDonald D.J. and Gerusky, T.M. Health Physics Vol. 12, No. 6, June, 1969 pp. 739-746.
3. Code of practice for the safe use of X-ray analysis equipment. Reprinted from the report of the 68th Session of the National Health and Medical Research Council, May 1969, Government Printer, Canberra, 1970.

## FACTS, FABLES AND FOLLIES IN MEDICAL RADIOLOGY

Richard E. Chamberlain, M.D.  
Chairman and Professor  
Department of Radiology  
Hospital of the University of Pennsylvania  
3400 Spruce Street  
Philadelphia, Pennsylvania 19104

It is repeatedly said that medical radiology is the largest single man-made source of radiation exposure of human beings. Not only is this true, justifying a considerable emphasis on medical radiology by all members of the International Radiation Protection Association, but it would be surprising if it were not true. Medical radiology is the only legitimate situation in which radiation exposure is purposefully given to human beings for their own benefit. It would be more remarkable, if not alarming, if any other man-made source exceeded it in human exposure.

As radiation protection experts, all of us should be well informed and have an accurate perspective of many of the aspects of medical radiology. We should also be able clearly to distinguish between facts, on the one hand, and the frequent fables and follies circulated and committed in the name of medical radiology, on the other. Radiation protection experts should have reasonably complete and accurate answers to such questions as: How much good is diagnostic radiology in medical practice? What is it particularly good for? How available is it? How well is it provided in different circumstances? What does it cost? How safe is its use and what can reasonably be done to make it even safer? Finally, how can radiation protection experts help to improve medical radiology in truly important ways?

I prepared this paper before the Congress began. It seems even more relevant after what you heard at the opening session and what was reported and emphasized in the news media from Mr. Lapp's presentation. You were told a large number of facts and subjected to much clever reasoning on radiation perspective in the United States of America.

Less in anger than in sadness, I must also say that you were subjected to some unfortunate fables, based on a series of "assumptions," and some dangerous follies were proposed, when Mr. Lapp strayed from his area of competence into pronouncements on medical radiation and what to do about it.

My viewpoint and attitudes are those of a physician-radiologist who has devoted a major part of his professional energies for over 35 years to the use of radiation in medicine. I have had three special interests: first, personal involvement in the medical care of individual patients, particularly in radiological diagnosis; secondly, appreciation of what is known of human radiobiological reactions, both somatic and genetic; thirdly, the constantly increasing applications of developments in physics, electronics and engineering to the production and control of x-ray beams, and to the improvement of methods of obtaining images of human structures. These interests have led

me to major involvement and commitment in two action programs. One is promotion of the principles and methods of expert and efficient use of radiation, commonly called radiation protection, which I prefer to call the thrifty use of radiation for medical diagnosis. It seems to me that the word thrifty more nearly expresses the concept of a proper balance between risk and benefit than does the word protection. No doubt my preference for the old fashioned term is influenced by the tradition of Benjamin Franklin and my Philadelphia environment. The second action program has been a dedication to the delivery of fine and appropriate radiological care to population groups large and small, both in the United States and abroad, and especially in disadvantaged and developing countries and areas.

What are some of the facts about the value of diagnostic radiology in medical management? In economically advantaged situations, where sophisticated medical radiology has been fully developed, experienced clinicians largely agree that from one-third to one-half of all the truly important decisions that they make for individual patients are based on radiological studies. These decisions are not only for the original discovery or diagnosis of disease and abnormality but also for intelligent and effective follow-up of the disease process and its treatment.

Three general levels of use can be related to a division of the types of diagnostic radiology into basic, advanced, and special examinations. As illustrated in Table I, basic examinations include reasonably simple studies of the chest, extremities, abdomen, head and spine. Advanced examinations frequently require the introduction of radio-opaque contrast materials and include studies of the stomach, small intestine, gallbladder, colon and kidneys. Special studies include the more elaborate examinations of the vascular system generally referred to as angiography, air studies such as pneumoencephalograms, etc.

Table I

Types of Diagnostic Radiology

1. BASIC: Chest, Extremities, Abdomen  
Head, Spine.
2. ADVANCED: Stomach, Small Intestine, Gall-  
bladder, Colon, Kidneys.
3. SPECIAL: Angiography, Air Studies, etc.

Even though the full spectrum of examinations is available in fully advanced medical installations, it is interesting that the simpler basic studies are not only more easily made available but also have higher yield of benefit for most patients. The true value of a major tool in medical care, such as diagnostic radiology, is not to be measured only in mortality statistics or any other oversimplified criterion. The quality of life and the sense of well-being and health of the individual is of primary concern, although we all hope also for longevity. Medical statistics are not a group commodity but are made up of the contributions of each personal unit. In economically disadvantaged areas, the well-being of the individual patient reflects on the family and national welfare to an even more critical extent than in advanced countries, concerned as we are with such things as the rising cost of medical care in all nations. The patient with a broken bone, or active tuberculosis, or a hemoglobin of 7 or 8 grams due to multiple intestinal

parasites or malaria is not only sick himself but may be a critical detriment to his family and to the national productivity. The restoration of such an individual to productive life can be triply effective by converting him from being a burden, through neutrality, to making a positive contribution to the family and national economy.

Many fables are in circulation concerning the use of medical radiological diagnosis. Some persons seem to believe that the present use is excessive, that it ought to be limited to extreme urgency and that rigid rules are justifiable in prohibiting the small doses needed for diagnostic purposes, especially in potentially pregnant women and in the case of the fetus. I would not wish to give the impression of condoning promiscuous and random waste of radiation exposures. As a physician, however, I would like to emphasize that rigid rules are almost never sensible in clinical practice, that in a high proportion of radiological examinations one cannot anticipate the beneficial yield until the examination has been performed, and that the omission of a vital examination can lead to a disaster far more important than a possible or statistical risk from the radiation exposure. I wholeheartedly urge that considered judgment be used in each individual decision for radiation use. I view as follies the emphasis on expensive, elaborate and essentially useless programs of national recordkeeping of medical exposures, simplistic rules as to the justification of abortion on a radiation basis and undue emphasis on radiation protection programs in areas which should be using those same energies to furnish more medical radiological diagnosis for their people within their extremely limited national economy.

The availability of medical radiology in advantaged countries is documented by such surveys as the XES 1964 and the XES 1970 studies in the United States of America. These studies show that diagnostic examinations average to half of the population per year and are available essentially equally to different economic groups. Less reliable is a precise documentation of the availability of diagnostic radiology and, indeed, of medical care generally to developing areas. It seems likely, however, that of the approximately 3.5 billion of the world's population, as many as 2 to 2 1/2 billion have essentially no modern medical care, including medical radiology. It is a fable to imagine that the full spectrum of highly developed radiological diagnostic equipment and service can be transplanted directly to developing countries. With gross national product figures of the order of \$200 to \$300 per capita per annum, with no facilities for maintenance, totally inadequate educational systems and with far more pressing needs for basic medical control of infectious diseases, sanitation, and nutrition, they face stark reality in choice of emphases. Well meaning though the intent may be, it is also folly to imagine that the furnishing of yearly monetary aid for immediate consumption, or the sending out of small groups of physicians from advantaged countries for service can solve these problems. Aid in the form of money and people can, however, be helpful and effective if applied to assistance of developing nations to help themselves. I quote, from an unknown source:

"If you give a man food  
You feed him for a day,  
But if you teach him how to grow his own,  
He can feed himself for life."

We also have some facts as to the current levels of the safety of the use of diagnostic radiology and of the costs. The full spectrum of modern diagnostic radiology can be enormously expensive; the increasing sophistication of the application of modern developments in electronics and physics, as well as the

general inflationary trends, are constantly raising those costs. Most of them, however, apply to the advanced and special studies which, desirable though they are for the individual patient, are generally less crucial in medical conditions which have a highly favorable prognosis. An impartial view would lead me also to propose that the associated radiation exposure, in even the most advanced nations, is acceptable both in terms of the actual radiobiological probabilities of somatic and genetic damage and in relation to the stark and unfulfilled health care needs and the variations within natural background. Fables are also in circulation concerning the safety of medical diagnostic radiation and, largely through misunderstanding or misquotation, have resulted in headlines attributing tens of thousands of deaths to diagnostic medical radiology. I simply do not believe, nor is there real scientific proof, that we are "killing" people with diagnostic radiology, even in those nations where it is being most widely used. I hasten to add, however, that I do not condone truly wasted radiation and believe that careful study and major attention should constantly be paid to both confining it to the area being examined and striving for higher efficiency. This brings us, however, to the difficulties and even the dilemmas of establishing cost effectiveness. It is always possible to use somewhat less radiation, no matter how low the exposure may be in any particular situation. This can be done either by much more elaborate pre-studies and identification of what is thought to be the precise problem or by the use of much more elaborate apparatus. In practice, however, there comes a point at which a very low dose simply does not justify the expenditure of excessive effort and cost to reduce it a tiny fraction further. A loss of perspective in this regard has led to what I consider to be a folly in our own nation, the proposal of regulations with undue emphasis on the percentage variation allowable in x-ray generator tolerances, and on elaborate feed-back control of collimation, most likely highly prone to greatly increased service problems. I believe that my opinion is supported by the XES 1970 study, which shows that the greater portion of the improvement likely to be achieved in collimation has already occurred between 1964 and 1970, without governmental regulation, but rather by education and voluntary cooperation.

I would like to make a plea that we refrain from playing the "numbers game" of extrapolated pseudo-statistics, which seems so popular these days, especially when human health and life are at stake as in clinical medicine. Fables are not converted to established facts by assumptions! We can sometimes excuse fables if they are harmless exercises of the imagination. We cannot excuse resultant follies if they stem from ignorance of elementary understanding of medicine, health, and the nature of diagnostic decisions for individual patients. Then they truly endanger the welfare of almost everyone--for all of us will eventually encounter illness and come to death.

Let me try to illustrate the cruelty of such folly. Mr. Lapp "calculated" 50,000 iatrogenic cancer deaths from diagnostic medical radiation in a little over 25 years. I reiterate that this was based on many "assumptions" and is not scientific fact. It divides into approximately 2,000 cases per year, however.

In the three hospitals for which I am the Director of Radiology, my staff and I perform a little over one thousandth of all the diagnostic radiological examinations per year in the United States. Even though we try to use as superb techniques as are available and practical within our financial limitations (and I think this may be better than the national average), we also have a high proportion of difficult, elaborate, and comprehensive examinations, with associated radiation doses that are higher than average.

If what Mr. Lapp contended were true, we could be producing 2 or more iatrogenic deaths per year. But let us now look at the real perspective in medical radiology. For this supposed risk, my staff and I are doing nearly 200,000 diagnostic radiological examinations per year. We not only interpret the films and do the associated procedures and fluoroscopies, but also know why the patients are being studied. In a large proportion of the cases, we personally consult with the other physicians involved regarding the original problem and its follow-up.

Beyond any shadow of doubt, our examinations are either the vital factor, or a major one, in saving tens of thousands of lives per year. I extend to Mr. Lapp, and to any of you, an invitation to come and see what really goes on in medical radiology -- and I am sure that great numbers of my radiologist colleagues would do the same.

To propose, as Mr. Lapp is reported to have said, that these examinations or their exposures could and should be reduced by 50% is not only folly and arrant nonsense, but would literally be condemning tens of thousands of patients to misery and untimely death, and for a highly dubious hypothesis of exaggerated emphasis. There are much higher priorities for intense attention.

I have dedicated a major part of my life to the cause of what I have referred to as "thrift" in the use of radiation. I can tell you that it is not easy or cheap to cut procedures or exposures in half. It may well cost triple or more for the same diagnostic information. This may well deny its accessibility to thousands of patients. Furthermore, if you had the choice of applying the same effort and cost to other directions which might save hundreds or thousands of lives, which would you have us choose?

The greatest cruelty is when, through the way in which it is reported, credulous people may be so confused or fearful that they forego needed radiological examination for their health and life.

For many years I have been seeking solutions to the need for simplified systems of equipment, associated with simplified education of technologists and physicians, and simplified service and supplies. In recent years this has resulted in a concept that I have called the "Technematic System" of Radiology. It has been developed on principles of systems engineering to satisfy not only these aims, but also to bypass the difficulties of deficient and unstable electrical power supplies, to produce superb and reproducible quality of films, to have optimum characteristics of radiation safety, and to perform all of the basic category of examinations as well as the radiographic portions of some advanced studies. Hopefully, at least in large volume production, such apparatus is within economic feasibility and the education requirements are attainable. In four years of practical field trials with the units, in conjunction with the University of Saigon School of Medicine, my staff agree that it "performs as hoped for." I have also gained some other experience with its use in an Emergency Clinic and, as applicable, to various examinations in a large teaching hospital.

Figure 1 shows an overall view of the stand design which features fixed distance from tube to film plane, fixed axial centering, a high quality, fine-line stationary grid, an adequate range of vertical movement of the tube-film carrier C-arm, and a wheeled litter which acts as a poor-man's floating top. Without the litter it can be used for extremities. Rotated to a horizontal beam, it is applicable to erect chest examinations, as in Figure 2, or erect skull studies. Only 2 sizes of films, in appropriate cassettes, are used, 14" x 17" filling the film slot and 10" x 12" centered with a spacer-tray. A simple two-position rotating-box double collimator ensures precise collimation to the two film formats (Figure 3). The x-ray beam is furnished by a



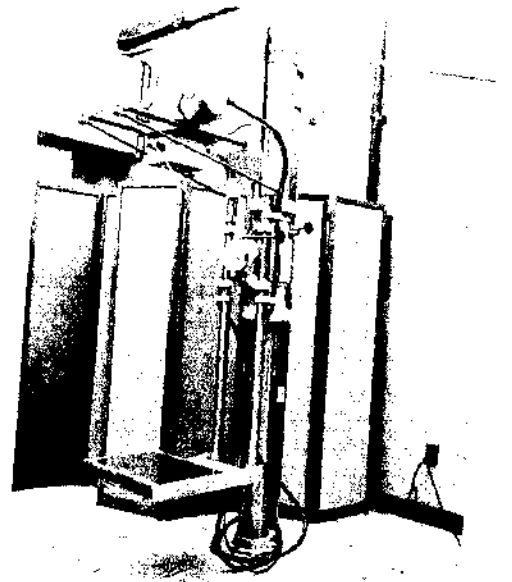


Figure 1.

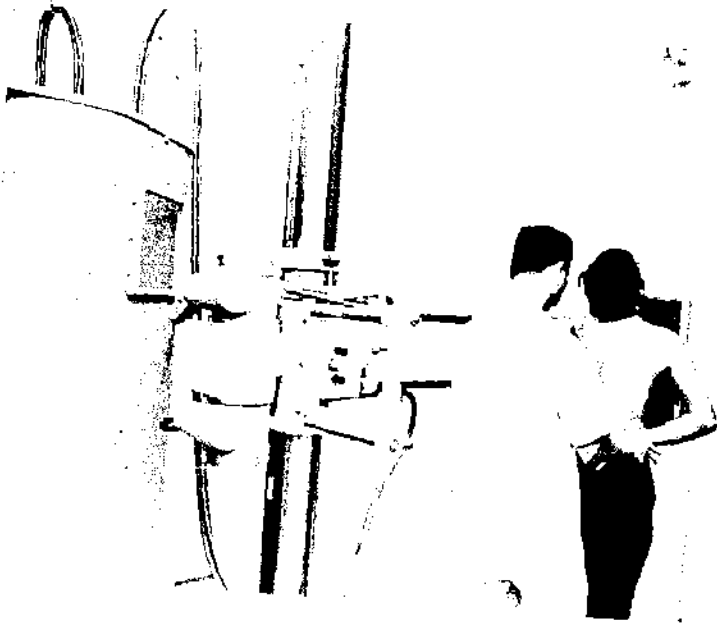


Figure 2.

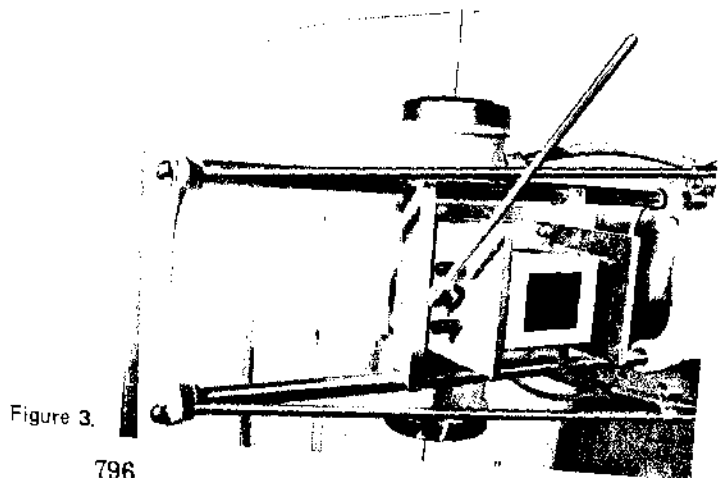


Figure 3.

500 Hz inverter at 100 milliamperes and 110 kV maximum, energized by high capacity nickel-cadmium batteries which also furnish filament and anode-rotor power. The controls are quite simple. High temperature hand-tank film developing has also been provided. The scope of examinations attainable is surprisingly complete and is shown in Table II. Simplified manuals for education of technologists and for daily use, applied specifically to this system, allow excellent performance after as little as 6 to 8 weeks training. They are easily translated into local languages. Tens of thousands of superb films so made are of such high quality that accurate diagnoses of fractures, pulmonary tuberculosis, and regionally prevalent disease are possible by physicians with only a few months radiological education. The good films also are suitable for referral to radiological specialists for consultative opinions. Service and supply problems are minimal compared to those posed by more complex apparatus, of various designs, and with less direct simplicity.

Obviously, many of these features have been dictated by the special needs and limitations of developing countries, but comparable advantages are also apparent when these concepts have been applied to small towns, inner city health centers, and special areas such as emergency clinics in hospitals in "developed" countries.

This example hopefully illustrates how practical, balanced, efficient, and deliverable solutions may be found to satisfy the multiple demands imposed by medical needs, poor financial and educational resources, and yet with due attention to high film quality and good radiation protection practice.

In conclusion, may I re-emphasize five points as follows:

1. There is a great need, for much of the world an unaccessible need, for the benefits of medical radiology for the good of mankind.
2. These needs can be met with due regard to acceptable radiation safety.
3. Emphasis on regulatory methods to ensure radiation safety is likely to be largely futile and much less desirable than emphasis on education and improved engineering.
4. Great future advances in the service of mankind are attainable through sound concepts of systems engineering which fits solutions to needs.
5. Radiation protection experts can exert great influence for good by discouraging narrow viewpoints and distorted emphases, and promoting balanced programs based on larger perspectives.

Mr. Lapp closed with a plea for you, in the International Radiation Protection Association, to assume individual responsibility to persuade the medical profession to reduce the diagnostic dose. I have also closed with a plea for you to work for dose reduction, but to work for it in areas where you have knowledge, experience, and understanding. May I also suggest that you be sure that you know the facts before leaping into areas beyond your expertise.

My favorite motto to post on the wall of scientific laboratories and workshops as well as in clinical areas is: "Are you working on the Answer; or are you part of the Problem."

TABLE II  
DIAGNOSTIC RADIOLOGY PROCEDURES WITH  
THE TECHNICOMATIC SYSTEM

NEST	PA & Lateral	MANDIBLE	AP & Oblique
INGERS	PA & Lateral	SINUSES	Waters PA & Lateral
AND	PA & Oblique	CERVICAL SPINE	AP & Lateral Lateral flexion/extension
RIST	PA & Lateral Ulnar Deviation	RIBS	AP & Oblique
OREARM	PA & Lateral	THORACIC SPINE	AP & Oblique
BOW	AP + Prone/Supine Lateral + Prone/Supine	LUMBAR SPINE	AP & Lateral Lateral L/S junction
MERUS	AP & Lateral	CLAVICLE	AP
HOULDER	AP Internal & external rotation	ABDOMEN	AP Supine and Erect Lateral Right and Left Decubitus
OT	AP & Oblique Calcaneus lateral	UROGRAM	AP Supine and Erect AP Pelvis
IKLE	AP & Lateral	GB	AP & Oblique Decubitus
IG	AP & Lateral	PELVIS	AP
EE	AP & Lateral	PREGNANCY	PA Erect Lateral
MUR	AP & Lateral		
P	AP Regular & Frogleg Lateral		
ULL	AP & PA Lateral right and left Special - Waters, Towne		

## OPERATIONAL HEALTH PHYSICS - 1

### OPERATIONAL HEALTH PHYSICS DURING DISMANTLING OF THE ELK RIVER REACTOR

Dan McConnon  
United Power Association  
Elk River, Minnesota

#### Abstract

The Elk River Reactor (ERR), a 58 MW (th) boiling water reactor, was operated for four years and shut down in 1968 for economic reasons. The facility is now being dismantled such that the reactor site will be returned to approximately the condition that existed prior to installation of the reactor with all vestiges of the reactor plant (except for subgrade foundations) having been removed and disposed of. This is the first time that decommissioning of a power reactor has involved complete removal and disposal of large radioactive structures, rather than in-place entombment.

Removal operations and radiological conditions during dismantling of the highly radioactive portions of the reactor are described. The removal sequence of these structures started with the reactor internals and inner thermal shield, progressed through the pressure vessel and outer thermal shield and ended with the inner sections of the biological shield. Segmentation of the structures, either by mechanical or hot-cutting techniques, and packaging for transportation were done remotely to control personnel exposure with the addition, at times, of temporary shielding. To prevent the spread of contamination to work areas, operations were carried out underwater, or where this was not possible, in a specially designed containment structure around the work area.

#### Introduction

The Elk River Reactor (Figure 1), an indirect-cycle, natural circulation, boiling water reactor, was constructed by Allis Chalmers under a United States Atomic Energy Commission contract as part of the Commission's second round power reactor demonstration program and was operated by the United Power Association of Elk River, Minnesota under contract to the Commission. The total thermal capacity of the plant was 58.2 megawatts with a rated net electrical output of 22.5 megawatts. The reactor plant was constructed adjacent to an existing conventional electrical steam generating plant on land owned by the United Power Association and leased to the Atomic Energy Commission.

Initial reactor criticality was achieved in November, 1962, with power operation commencing in July, 1964. The plant was operated by the United Power Association from June, 1965 until final shutdown in January, 1968. Since that time, the Elk River Reactor has been maintained in a shutdown status with a limited number of systems being operated. All fuel was removed from the reactor and shipped from the site in 1968 and 1969.

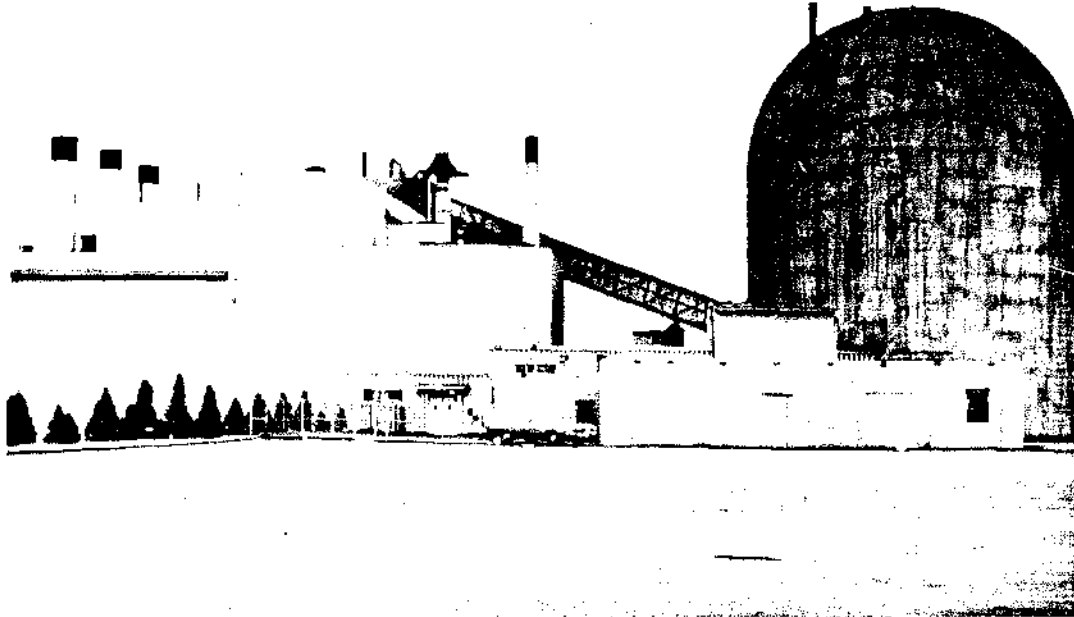


Figure 1: General View of the Elk River Reactor Site and Electrical Steam Generating Plant

Upon expiration of the contract with the Atomic Energy Commission for operation of the reactor, the United Power Association, for economic reasons, waived its option to purchase the plant. Pursuant to the terms of a modification to the operating contract, the Elk River Reactor Plant is being dismantled with all structures being removed approximately to grade level. Upon completion of the dismantling operation which is estimated to require approximately 2-1/2 years, the site will be available on an unrestricted basis to the United Power Association for installation of facilities associated with its conventional steam-electrical generation plant.

Planning of dismantling operations began in March, 1971 and a document summarizing planned operations<sup>(1)</sup> was submitted for approval to the Atomic Energy Commission in August, 1971. An Environmental Impact Statement on the project<sup>(2)</sup> was issued by the Commission in May, 1972. An order to proceed with dismantling of the facility was received from the Commission in June, 1972 when dismantling operations began. To date, all highly radioactive structures including the reactor internals, pressure vessel and outer thermal shield have been removed and disposed of. The current phase of dismantling involves removal and disposal of the concrete biological shield.

## Removal Operations

The general arrangement of reactor internal structures is shown in Figure 2. The removal sequence of highly radioactive structures proceeded from the inside out. That is, the first structures removed were the components contained within the reactor pressure vessel, then the pressure vessel itself, and finally the outer thermal shield which was a cylindrical lead and steel sandwich providing a radiation barrier and a thermal barrier between the pressure vessel and the biological shield concrete. Estimates of the total inventory of long-lived radioactivity in each of the structures and measurements of the resultant maximum contact radiation levels are shown in Table 1. The total inventory of radioactive material in the facility at the start of dismantling was estimated to be 10,000 curies of which greater than 99% was contained in the structures listed in Table 1. These structures are those which have been removed and disposed of thus far in the dismantling program. Contact radiation levels as measured underwater varied from 1 to 8000 R/hr.

All internal structures were attached each to the other either mechanically by means of machine screws or bolts or they were welded in place. Specially designed remote tools were used to separate those components which were mechanically united while a remotely-operated plasma arc cutting torch was used to separate components welded together. Separation of all components was done underwater. With the exception of the core and shroud support plate, components were transferred in-air (Figure 3) from the reactor cavity to a fuel element storage well located near the reactor cavity where preparation for shipment was completed. Because of high radiation levels, the core and shroud support plate was transferred underwater through the fuel

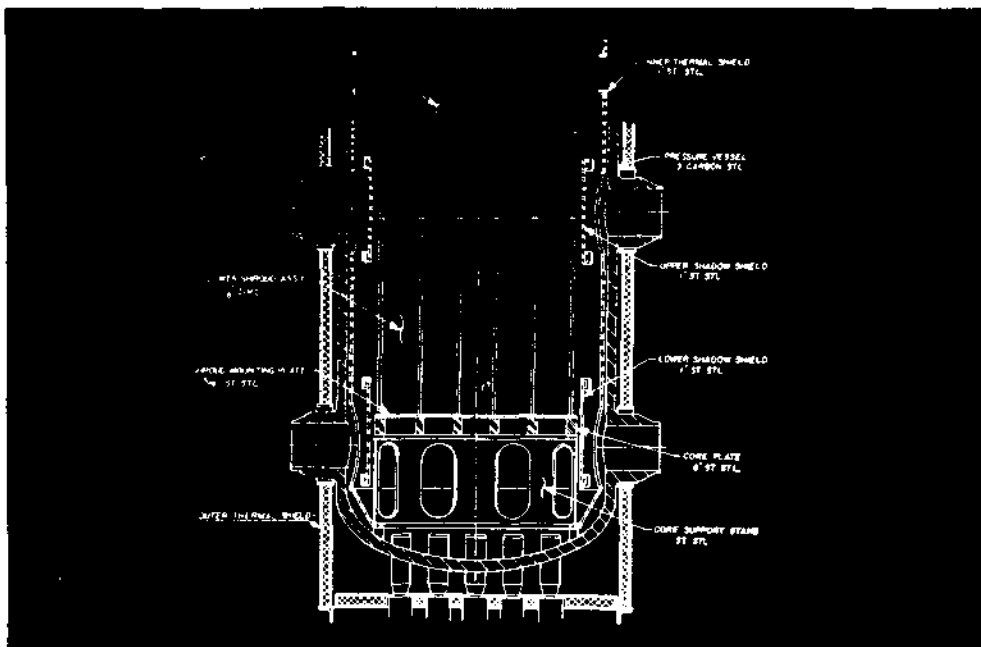


Figure 2: Elk River Reactor Pressure Vessel Internals and Outer Thermal Shield



transfer canal to the fuel element storage well. The packaging and disposal of the reactor internals depended upon the size of the component and its external radiation level. Components were segmented, if necessary, and loaded underwater (Figure 4) into various types of steel or lead and steel shielded cask liners (Figure 5) which in turn were then transferred in-air (Figure 6) to one of two shipping containers for transportation to the burial facility. The LL-50-100 container (Figure 7) weighs 50,000 pounds and has an internal volume of 100 cubic feet. Shielding is provided by 3-1/2 inches of lead and 2-1/4 inches of steel. The BC-48-220 container (Figure 8) weighs 48,000 pounds, has an internal volume of 220 cubic feet and has 7 inches of concrete and 2-1/2 inches of steel for shielding.

Table 1

Radioactive Inventory and Maximum Contact  
Radiation Levels by Reactor Component

<u>Component</u>	<u>Inventory (Curies)</u>	<u>Maximum Contact Radiation Level - R/hr</u>
Internals		
Upper Shroud	770	2800
Lower Shroud	35	175
Core & Shroud Plate	2370	8000
Core Support Stand	100	150
Inner Thermal Shield	3090	1000
Shadow Shields	2330	3000
Feedwater Distribution Ring	75	60
Pressure Vessel	1110	115
Outer Thermal Shield	75	1
	<u>9955*</u>	
Total		

\*As of April 30, 1971

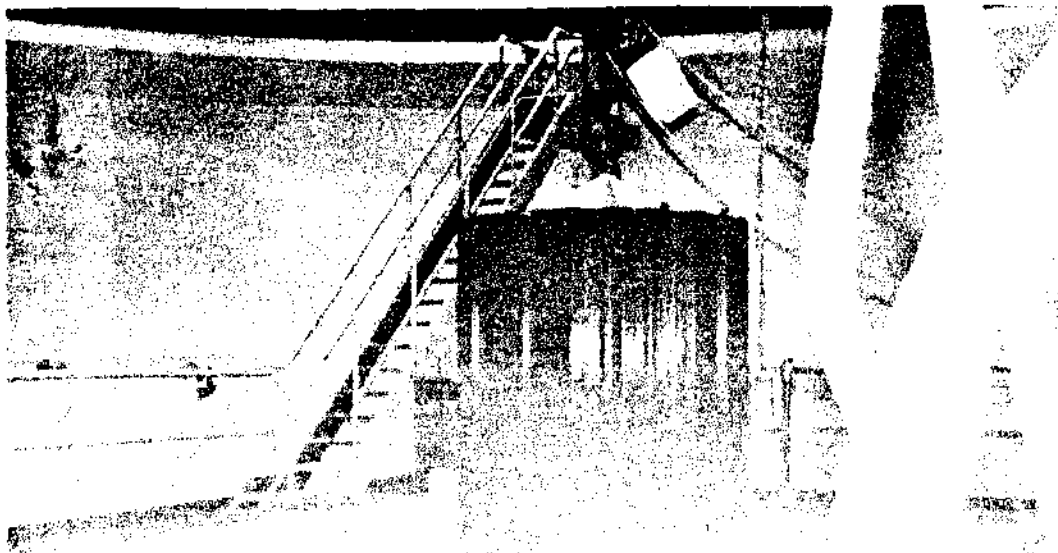
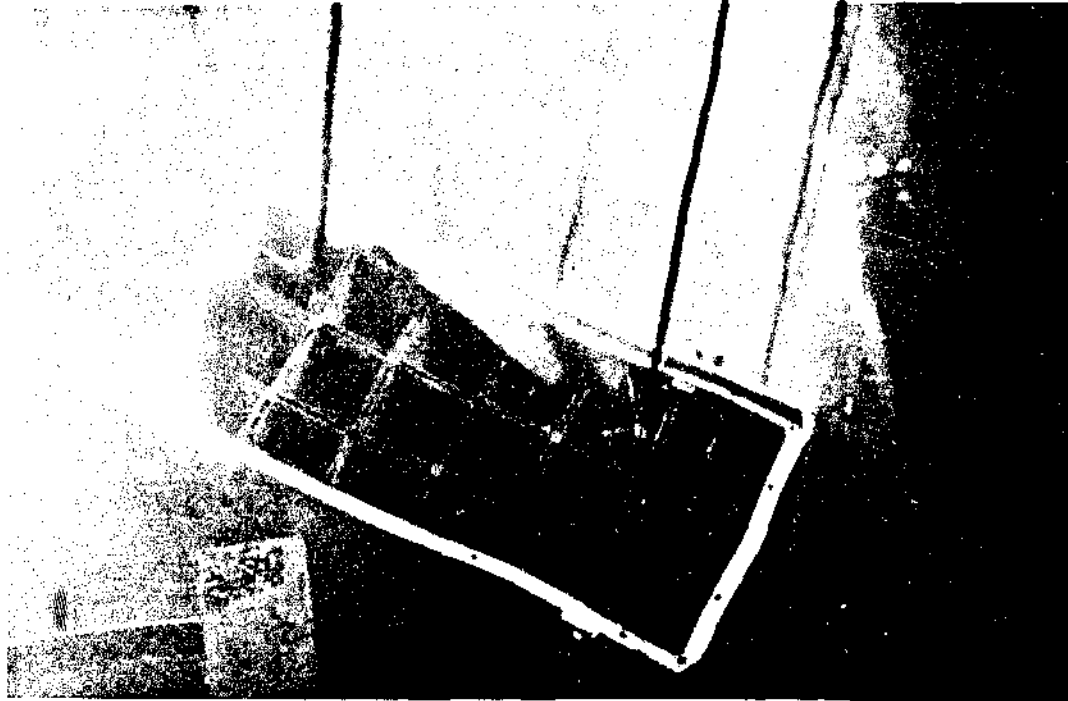
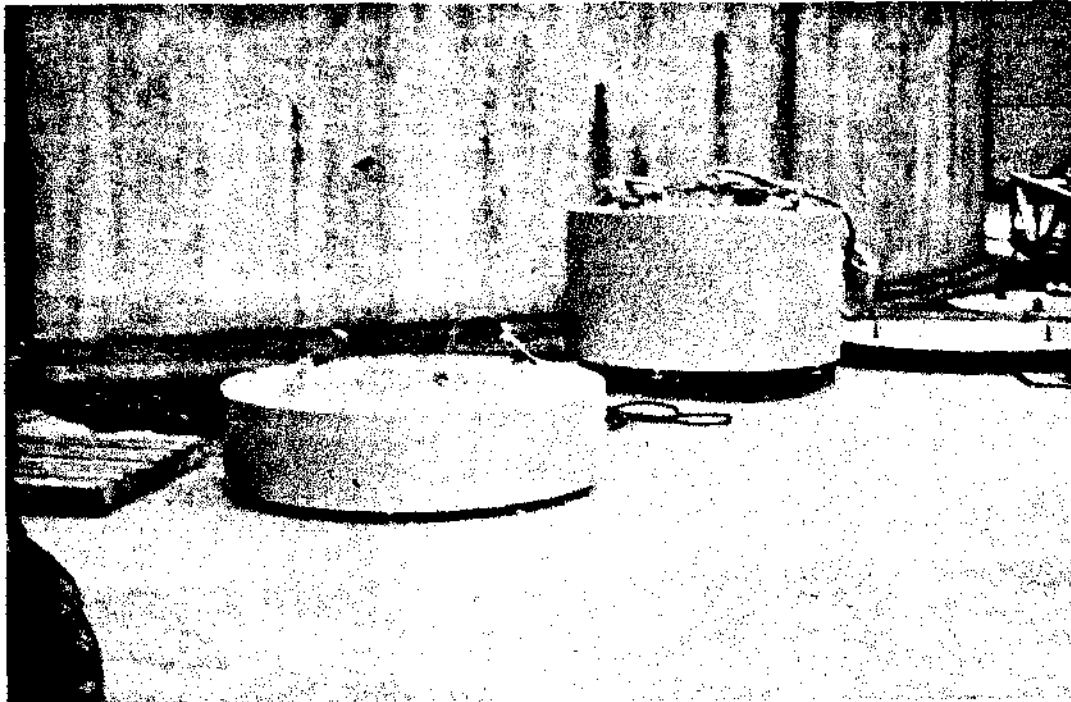


Figure 3: In-Air Transfer of the Reactor Upper Shroud Assembly



**Figure 4: Segmented Upper Shroud Assembly in Shielded Cask Liner Underwater**



**Figure 5: Typical Shielded Cask Liners Used for Packaging of Reactor Internal Components**



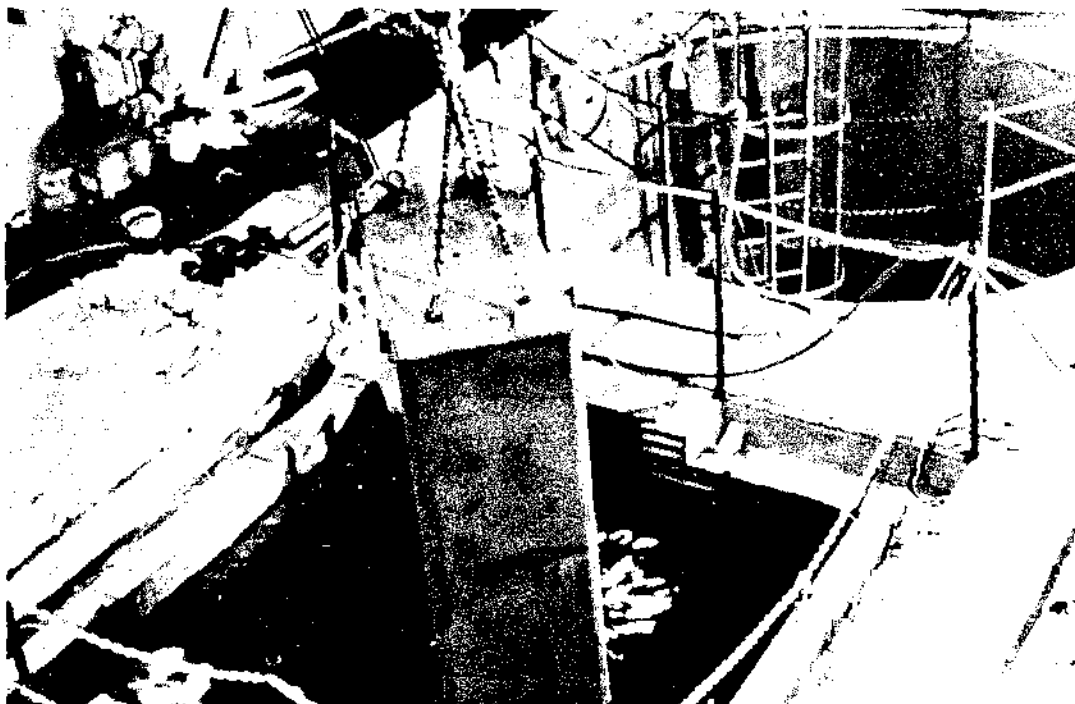


Figure 6: In-Air Transfer of a Shielded Cask Liner Containing a Segment of the Upper Shroud Assembly

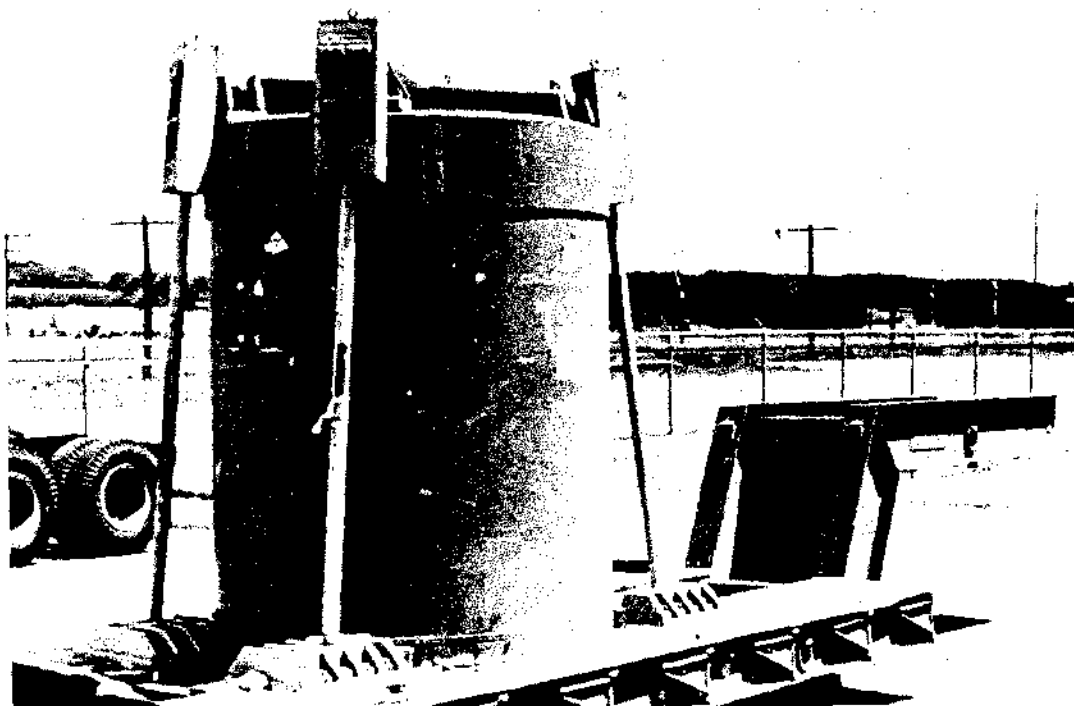


Figure 7: The LL-50-100 Shipping Container

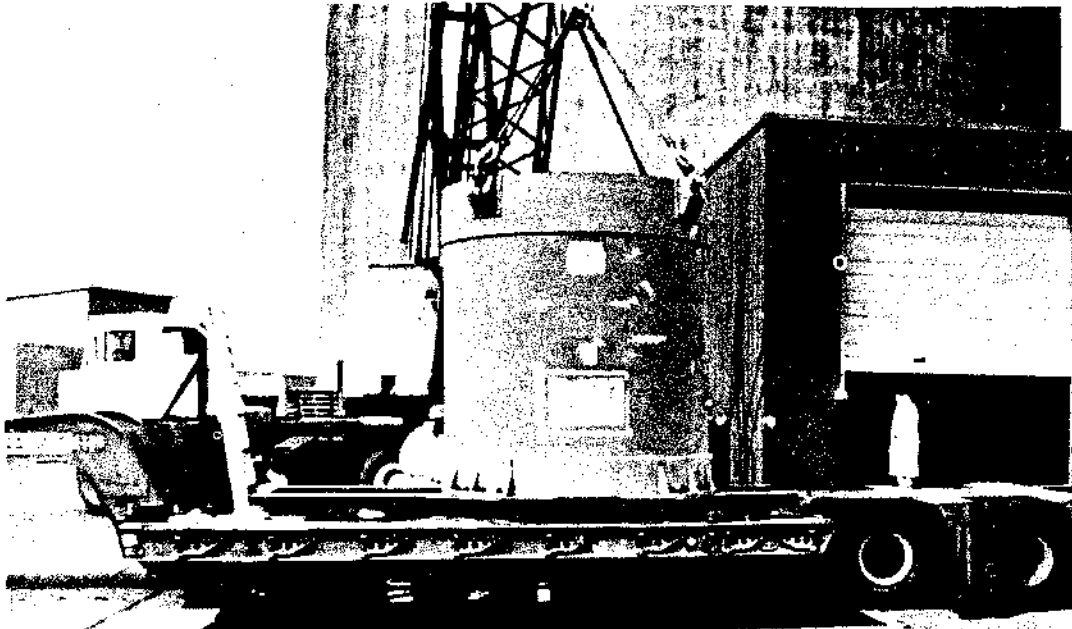


Figure 8: The BC-48-220 Shipping Container

During in-air movements of radioactive components or loaded liners, all unnecessary personnel were cleared from the reactor building and those engaged in the transfer were located in a shielded area for the control of radiation exposure. During underwater operations, personnel exposure rates usually varied between 5 and 15 mR/hr while in-air movements caused exposure rates of from 100-200 mR/hr to personnel in the shielded area. During loading and preparation of the shipping container, personnel exposure rates as high as 2 R/hr were experienced for short periods of time.

Segmentation of the pressure vessel and inner and outer thermal shields was done in place. The inner thermal shield was segmented with a remotely-operated plasma arc cutting torch designed for underwater operation. The pressure vessel and the steel portions of the outer thermal shield sandwich were segmented remotely in-air, again with a plasma arc torch. Lead was melted away from the outer thermal shield with an acetylene torch. All of these operations were done within a containment enclosure formed by the side walls of the reactor cavity. All penetrations into the cavity were sealed and a metal cover containing a filtered air inlet was placed over the cavity (Figure 9). The containment enclosure had a separate exhaust fan and filtering system (Figure 10) containing a prefilter and a high efficiency particulate filter capable of removing most of the airborne contamination generated during cutting operations. Samples of airborne contamination from within the containment enclosure taken during cutting operations indicated concentrations on the order of  $10^{-7}$   $\mu\text{Ci/cc}$  of gross beta activity. On the other hand, samples of reactor building air outside the containment structure rarely exceeded  $10^{-11}$   $\mu\text{Ci/cc}$  indicating the effectiveness of the exhaust filtering system. Following segmentation, the pressure vessel and outer thermal shield segments were packaged for shipping as outlined previously (Figure 11 and 12).

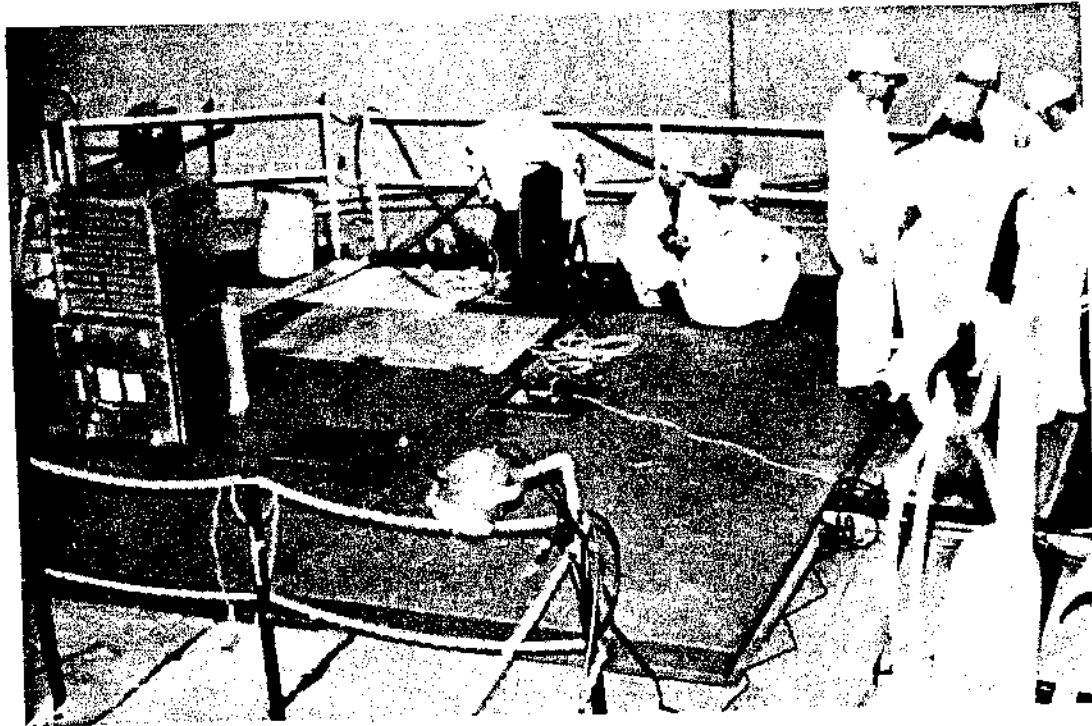


Figure 9: Metal Cover over the Containment Enclosure Showing Filtered Air Inlet, Viewing Windows and Access Plate

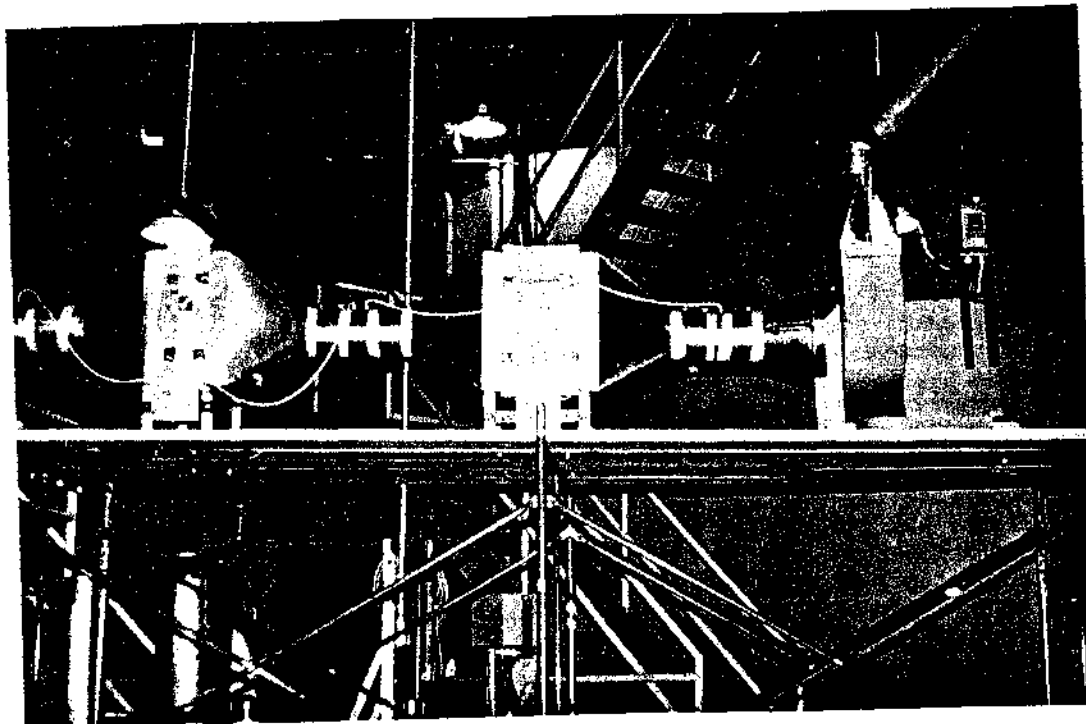
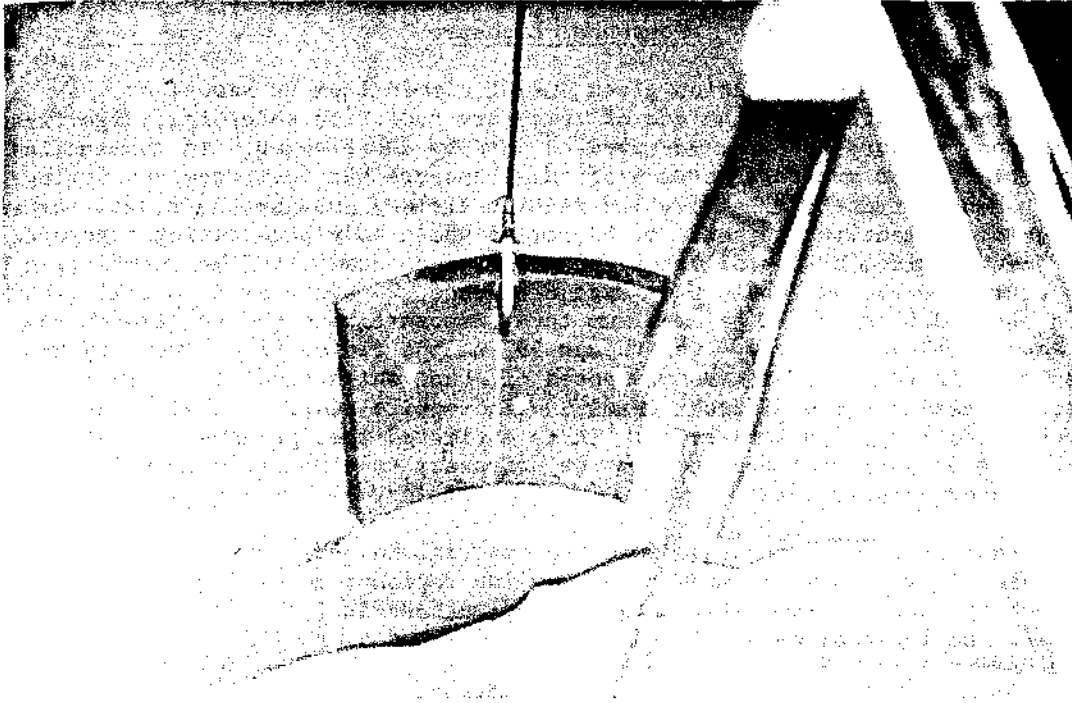
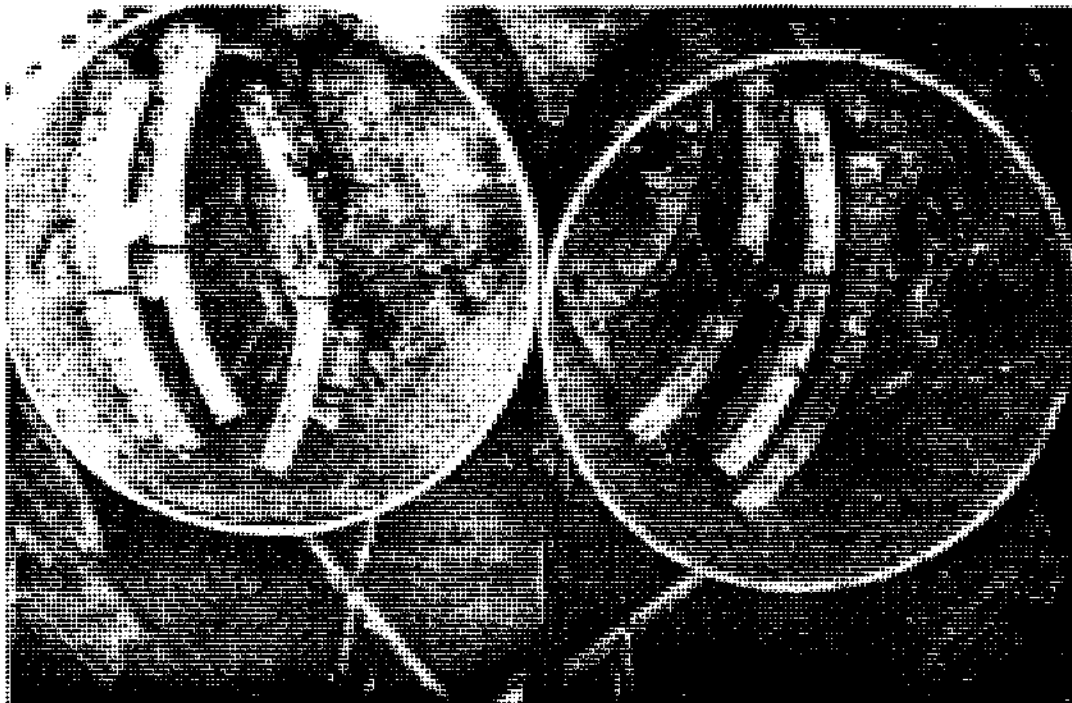


Figure 10: Containment Enclosure Exhaust System Containing Pre-Filter, Absolute Filter and Blower



**Figure 11: In-Air Transfer of a Pressure Vessel Segment**



**Figure 12: Pressure Vessel Segments in Cask Liners during Preparation for Shipment and Disposal**

### Personnel Exposure

In spite of the attention given during dismantling to remote or underwater operations or the use of temporary radiation shielding, significant radiation exposure has been received to date by those engaged in dismantling activities. The greatest portion of this exposure has been received during almost constant work in low level radiation fields. During the first fourteen months of dismantling, a total of 62 rem of whole body penetrating exposure has been received by approximately 80 people connected with the dismantling project. The average whole body exposure since the start of dismantling has been about 0.8 rem while the maximum total exposure received to date by any workman has been 4.8 rem. Even though 99% of the radioactive inventory has been removed from the site and disposed of, that which remains, primarily concentrated in the biological shield, will require additional chronic exposure to low radiation levels. It is estimated that perhaps another 20-30 rem of personnel exposure may be necessary to complete dismantling of radioactive structures.

Internal deposition of radioactive material in personnel is estimated periodically with a whole body counter. The radionuclides identified by whole body counter examination have been almost exclusively  $^{137}\text{Cs}$  and  $^{60}\text{Co}$ . To date, no internal deposition has exceeded 1% of a maximum permissible body burden.

### Liquid and Airborne Waste Management

The restraints placed upon the dismantling project with respect to the release of liquid and airborne wastes to the environment were outlined in the project's Environmental Impact Statement. A commitment was made to control waste releases below the numerical guidelines proposed in Appendix I of Title 10, Part 50 of the Code of Federal Regulations as shown in Table 2. The annual average concentration of liquid effluents at the site boundary is not to exceed 20 pCi/l of gross beta activity and 5000 pCi/l of tritium. The annual average airborne emissions are not to exceed the numerical values contained in Appendix B, Table II, Column I of 10 CFR 20 divided by a factor of 100,000. For the radionuclides estimated to be present in Elk River Reactor structures and given the meteorological conditions of the area and site layout, the annual average concentration at the worst location of the site boundary is not to exceed  $7.5 \times 10^{-3}$  pCi/m<sup>3</sup>. The intent of these numerical guidelines is to limit public exposure in the vicinity of the project to less than 5 mrem per year. Since June, 1972, the average concentration at the site boundary of liquid effluents has been 5.5 pCi/l of gross beta activity and 350 pCi/l of tritium while airborne waste concentrations has averaged 4% of the applicable limit. Using very pessimistic assumptions, it is estimated that the maximum dose received by an individual in the vicinity of the site from effluents generated during dismantling operations has been 0.4 mrem.

Table 2

Proposed Numerical Guidelines of  
Appendix I, 10 CFR 50 for Release  
of Radioactive Waste Products to the Environment

<u>Type of Waste</u>	<u>Annual Average Concentration</u>
Liquid	20 pCi/l 5000 pCi/l for Tritium
Airborne	<u>Appendix B, Table II, 10 CFR 20</u> 100,000
(For Elk River Dismantling)	0.0075 pCi/m <sup>3</sup>

Operations to be Completed

Since the United Power Association's contract with the Atomic Energy Commission calls for complete removal and disposal of the reactor building including all structures contained therein, there is a considerable amount of dismantling yet to be completed. However, all highly radioactive structures have been removed from the site. From a radiological standpoint, the only structures yet to be removed which contain significant quantities of radioactive material are the concrete biological shield and certain reactor systems such as the liquid waste disposal system. It is estimated that removal of these structures will take approximately six months following which demolition of remaining structures by conventional methods will begin.

References:

- (1) AEC Elk River Reactor Dismantling Plan (Rural Cooperative Power Association (UPA) and Gulf United Nuclear Fuels Corporation), SS-836, August 27, 1971.
- (2) Environmental Statement, Elk River Reactor Dismantling (U. S. Atomic Energy Commission), WASH-1516, May, 1972.

СПЕЦИФИЧЕСКИЕ ВОПРОСЫ РАДИАЦИОННОЙ БЕЗОПАСНОСТИ И  
САНИТАРНО-ДОЗИМЕТРИЧЕСКОГО КОНТРОЛЯ  
ПРИ РАБОТАХ С ТРАНСПЛУТОНИЕВЫМИ ЭЛЕМЕНТАМИ

М.С. Егоров, В.В. Копьев, Л.Н. Коржов,  
Г.М. Пархоменко, В.А. Сарычев

Институт биофизики Министерства здравоохранения СССР  
Москва, Союз Советских Социалистических Республик

Abstract

The report shows that the factors of radiation exposure at operations with transplutonium elements are in quantitative dependence on the physico-chemical properties, the time of target irradiation in the reactors and the degree of their purification from the fragment fission products. The doses of external and internal irradiation have been estimated.

В процессе радиохимической переработки облученных мишеней при получении тяжелых изотопов трансплутониевых элементов (ТПЭ) радиационная обстановка характеризуется как внешними потоками ионизирующих излучений, так и газоаэрозольным загрязнением воздушной среды радиотоксичными веществами.

Особенность работ с ТПЭ состоит в том, что приходится сталкиваться с одновременным действием практически всех типов ионизирующих излучений. Вследствие этого требуется дифференцированный подход к комплектации защитного оборудования в зависимости от рода проводимых работ, изотопного состава тяжелых элементов, их весовых количеств и степени очистки от продуктов деления. Данные положения могут быть иллюстрированы материалом, представленным в табл. 1 и 2.

Из табл. 1 видно, что при работах с малыми количествами ТПЭ, когда используются защитные экраны малой толщины, важное значение приобретает степень очистки ТПЭ от изотопов осколочного происхождения.

Таблица 1

Относительный вклад в суммарную мощность дозы  
различных типов ионизирующих излучений  
смеси оксидов америция с кюрием

Тип излучений	Относительный вклад в %	
	Без защиты	Защита из чугуна 15 см
Нейтроны спонтанного деления	5,2	77,0
Нейтроны ( $\alpha$ , n)-реакции	0,2	3,0
Собственное гамма-излучение ТПЭ	13,3	-
Собственное гамма-излучение продуктов деления	21,7	19,5
Тормозное излучение	59,6	0,5
ВСЕГО	100,0	100,0

Таблица 2

Относительный вклад в суммарную мощность дозы  
различных типов ионизирующих излучений  
оксида калифорния

Тип излучений	Относительный вклад в %	
	Без защиты	Защита из чугуна 15 см
Нейтроны спонтанного деления	95,5	99,9
Собственное гамма-излучение калифорния	3,5	0,1
Собственное гамма-излучение продуктов деления	0,2	-
Тормозное излучение	0,8	-
ВСЕГО	100,0	100,0



При работах с калифорнием практически вся радиация формируется нейтронами спонтанного деления.

Одновременно с изучением уровней радиации за защитой была проведена оценка индивидуальных квартальных доз от нейтронного и гамма-излучений при работах с изотопами америция и кюрия. Для операторов величины доз внешнего гамма-облучения составляли  $0,05 \pm 0,15$  рентген, а дозы от нейтронов - до  $0,05$  бэр. Это указывает на то, что работы проводились, главным образом, с небольшими количествами ТПЭ в боксах с легкой защитой.

Введение в работу изотопов калифорния повышает дозы нейтронного облучения до  $0,36$  бэр за квартал. При этом доза гамма-облучения остается без изменений.

Представляло интерес оценить также для операторов горячей лаборатории примерную величину инкорпорированной активности, накопленную за время работы с ТПЭ. Характеристика воздушной среды рабочих помещений определялась нижеследующими данными.

Распределение среднемесячных концентраций радиоактивных аэрозолей ТПЭ при выполнении различных операций соответствовало логарифмически нормальному закону с параметрами: медиана -  $1 \cdot 10^{-15}$  кюри/литр, стандартное геометрическое отклонение -  $2,4$ . С вероятностью 95% все значения концентраций находились в интервале  $1 \cdot 10^{-16} + 1,4 \cdot 10^{-14}$  кюри/литр.

Характерный изотопный состав аэрозолей: америций-241 - 10%, америций-243 - 5%, кюрий-244 - 75%, калифорний-252 - 10%. Как правило, получаемые соли были окислами ТПЭ и относились к классу нерастворимых соединений.

Дисперсность аэрозолей, определяемая радиографическим методом с использованием жидких ядерных эмульсий, характеризовалась параметрами: СМАД =  $3 \pm 5$  мкм,  $\sigma_g = 1,5 \pm 2$ , АМАД =  $13 \pm 20$  мкм.

Величины накопленной активности и доз внутреннего облучения различных областей дыхательного тракта (носоглоточной, трахеобронхиальной, легочной и лимфоузлов) при непрерывном поступлении аэрозолей ТПЭ в течение 5 лет работы приведены в табл. 3.

Таблица 3

Нагрузки и дозы для различных областей дыхательного тракта

Изотоп	Накопленная активность в ликокюри		Дозы облучения в бэрах			
	Р	Л	Н-Р	Т-В	Р	Л
Америций-241	4	3	$2,4 \cdot 10^{-3}$	$< 1 \cdot 10^{-3}$	0,04	0,2
Америций-243	2	1,5	$1,2 \cdot 10^{-3}$	$< 1 \cdot 10^{-3}$	0,02	0,1
Кюрий-244	27	23	$19 \cdot 10^{-3}$	$< 1 \cdot 10^{-3}$	0,3	1,7
Калифорний-252	3	3	$2,5 \cdot 10^{-3}$	$< 1 \cdot 10^{-3}$	0,04	0,2
ВСЕГО	36	30,5	0,025	$1 \cdot 10^{-3}$	0,4	2,2

Оценка проводилась согласно динамической модели легких<sup>1</sup> с использованием расчетных формул и номограмм из работы<sup>2</sup>. Коэффициент использования средств индивидуальной защиты был принят равным 90%. Рассчитанные дозы внутреннего облучения легких за

5 лет соизмеримы с дозами, полученными за счет внешнего облучения в течение этого же времени.

При переработке реакторных мишеней, когда необходимо выделить ценные короткоживущие изотопы типа кюрия-242, берклия-249, эйнштейния-253, облученный материал поступает с малым временем выдержки. Вследствие этого значительную роль начинают играть изотопы осколочного происхождения. Следует учитывать возможный выдох в воздух радиоактивных газов и аэрозолей продуктов деления и, в первую очередь, йода-131. Проведена оценка вкладов газовой и аэрозольной фаз йода-131 при некоторых способах переработки мишеней (табл. 4).

Таблица 4

Относительный вклад аэрозольной и газовой фаз  
в суммарную активность йода-131 в воздухе горячей камеры

Вид переработки мишени	Относительный вклад, %	
	Аэрозольная фаза	Газовая фаза
Механическая резка	99,8	0,2
Щелочное растворение	81,0	19,0
Кислотное растворение	48,0	52,0

Из табл. 4 видно, что при кислотном растворении существенно возрастает процент газовой фазы, которая играет ведущую роль в загрязнении воздушной среды радиоактивным йодом-131 и увеличивает вероятность проникновения его в зону пребывания персонала.

Выявляемая специфичность радиационных условий при работах с ТПЭ требует дифференцированного подхода к решению задач санитарно-дозиметрического и медицинского контроля персонала.

#### Литература

1. Morrow P. E. et al. Health Phys., 12, 173 (1966).
2. Kotrappa P. Health Phys., 17, 429 (1969).

ENSURING RADIATION SAFETY WHEN USING LARGE ISOTOPIC  
SOURCES OF IONIZING RADIATIONS FOR BRINGING ABOUT  
RADIATION PROCESSES.

Chistov E.D., Larichev A.V., Terman A.V.,  
Zaraeyev O.M.

( USSR )

ABSTRACT.

Results of an investigations series for ensuring personnel radiation safety when bringing about various radiation processes are presented. The investigations include: studying radiation penetration regularities in large isotopic plants shielding structures such as labyrinths, technological channels etc. with design formulae; studying environment contamination mechanisms formation using the design formulae which allows to prognosticate the foreseen wasnes amount. Analysis of the accidents occurred during large isotopic plants operation and proper procedures developed to prevent their occurrence are given. The All-Union standards and methodical rules regulating radiation safety conditions in design, construction and operation of large radiation apparatus on the basis of the authors investigations are considered. The many-years experience showed that the observance of the standards developed by the autors reduces and in a number of cases completely excludes the possibility of the personnel over-exposure, the environment and irradiated products contamination.

ENSURING RADIATION SAFETY WHEN USING LARGE ISOTOPIC  
SOURCES OF IONIZING RADIATIONS FOR BRINGING ABOUT  
RADIATION PROCESSES.

Chistov E.D., Larichev A.V., Ternan A.V.,  
Zaraeyev O.M.

( Central Research Institute of Labour Protection of All-Union  
Central Council of Trade Unions,  
Branch of the Karpov Physicochemical Institute,  
Central Institute of Advancement Training of Physicians, USSR)

Ensuring radiation safety when using large isotopic sources is a necessary condition for bringing about any radiochemical and other investigations and processes (1). The urgency of studying such matters is caused by the necessity of solving engineer and technological problems in developing radiation plants and processes excluding any possibility of undesirable consequences of the over-exposure of the plants personnel, the neighbouring area population and irradiated products consumers (2,3). The development of a series of such plants which are in full accord with the present day radiation protection demands became possible due to fundamental and experimental investigations including (4):

- protection from external radiation - studying radiation penetration regularities in large isotopic plants shielding structures and developing design methods;
- protection from internal radiation - studying the environment contamination mechanisms' formation;
- radiation processes safety - studying the conditions of accident-free radiation processes;
- formulation of standards and methodical rules - development of sanitary rules for radiation plants design, construction and operation.

The following hazard factors arising from the use of large ionization sources in radiation technology were considered:

- a) radiation hazards defined by the physical nature, activity and type of radioactive substances and the character of their technological use; quantitatively these hazard factors are defined by the doses of the personnel external exposure and internal exposure stipulated by the radioactive substances entering the organism of workers;
- b) non-radiation hazards characterized by the quantity and type

of toxic substances intaken from irradiated objects or formed by irradiation processes, or as a result of a fire or explosion or by aggressive media formation. Sudden or gradual destruction of hermetic shells in the sources, shieldings and apparatus may be observed afterwards (5).

Transportable and movable units may have biological shield in the form of a cylinder or a sphere. The information about the larger efficiency of such type shielding in comparison with flat shielding having the same thickness could be found only in single publications (6-9). However the physical nature of this phenomenon was not analyzed. The authors have shown that for a point on the shielding surface the share of the scattered radiation from the flat-type shielding is larger than from the spherical shielding of the same thickness. Energy distribution studies of scattered gamma-radiation intensity showed the maximum the energy of which was approximately equal to the energy of gamma-quanta single scattered at an angle  $\theta$ . Calculation and experimental results allowed to determine that the dependence of the angle distribution of the intensity ( $I_\theta$ ) of gamma-quanta scattered in a spherical or flat shielding from the angle  $\theta$  is of exponential character and can be given in the considered angles range (with an error  $\pm 10\%$ ) as follows:

$$I_\theta = A_{fl(sph)} \cdot \exp\left(-\frac{\theta}{\theta_{fl(sph)}}\right) \quad (1)$$

where:  $A_{fl}$ ;  $A_{sph}$  - are the proportionality coefficients for flat and spherical shieldings respectively;

$\theta_{fl/sph}$  - the constants of the angle distribution of the intensity of gamma-quanta scattered in flat and spherical shieldings respectively.

Moreover the decrease of the shielding material atomic number and gamma-quanta energy leads to the increase of the difference  $\delta^{(s)}(A)$  in the energy  $B_E$  and dose  $B_D$  build-up factors:

$$\delta^{(s)}(A) = \frac{B_E^{fl} - B_E^{sph}(A)}{B_E^{sph}(A)} \cdot 100\% \quad (2)$$

Shielding geometry effect becomes more essential for gamma-quanta energy less than 1 Mev and the shielding made from the low atomic number materials (10,11). The physical characteristics studies of multiply scattered gamma-quanta in the lead and steel shielding in the standard many-elbowed channel of the serial agricultural units showed that the curve from of gamma-ra-

radiation intensity reduction (I) along the axis of the channel is slightly dependent on the initial energy of radiation and can be given as the following dependence:

$$I(l) = I(l_0) \cdot C \cdot e^{-K} \quad (3)$$

where: C and K are the empirical coefficients equal to 58,0 and 2,6; 22,0 and 3,0 for steel and lead respectively. It is necessary to note that K is slightly dependent on the shielding material;  $l$  - is the geometrical parameter of the many-elbowed channel.

As the investigation results of spectral composition of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  gamma-radiation passed through the many-elbowed channel show there is the maximum in the energy range of 150 keV. When the channel is filled by water-equivalent material the spectra maximum is changed to the energy range of 100-120 keV, whereas the portion of multiply scattered gamma-quanta increases, radiation intensity for  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  decreases on 20-30% accordingly. The obtained data on scattered gamma-radiation spectral composition allowed to recommend technological channels schemes and choose detectors with corresponding characteristics for biological dosimetry.

The studies of the field of the exposure dose rates in a labyrinth and its spectral distribution gave a formula (4) that is of great importance not only for constructing labyrinths but also for the evaluation of exposure doses during the anticipated accidents (12,13):

$$P = 8,4 \cdot A \left[ \frac{e^{-\mu x} \cdot B(h\nu, \mu x, z)}{R^2} + \frac{2}{(20)^n R_1^2} \prod_{i=1}^n \frac{S_i \cos \theta_i \alpha_i(\theta, E)}{R_{i+1}^2} \right] \quad (4)$$

where: A - the irradiator gamma-equivalent, mg-equiv.Ra;

$\theta$  - the normal/ $R_1$  angle;

$R_1$  - the distance (cm) from the centre of the irradiator to the centre of the area  $S_1$  ( $\text{cm}^2$ );

$R_{i+1}$  - the distance from the area  $S_i$  where  $i$  - reflection occurs to the corresponding calculated point (or to the next area  $S_{i+1}$ );

$\mu$  - the linear coefficient of gamma radiation decrease,  $\text{cm}^{-1}$ ;

x - the concrete shielding thickness, cm;

R - the irradiator - to calculated point distance, cm;  
 $d_i(\theta, E)$  - the albedo of the dose from concrete (from  
i-area);

$B(\mu, x, z)$  - the dose build-up factor.

The formula (4) is just under the following assumptions; the irradiator is of point type, linear dimensions of the areas are comparatively small to the distance  $R_1$ .

Realizing the future prospects of using beta-radiation sources in the large radiation plants construction the authors have experimentally studied beta- and gamma-radiation penetration through the shielding non-homogeneities peculiar to any plant. As a result they got empirical dependence which allowed to evaluate mixed radiation dose from technological slits and openings. The dependence received takes into account the slit depth (t), its height (g), and the outer edge of the slit-to-detecting point distance (x). The formula connecting the dose rate ( $P_{\beta+\gamma}$ ,  $\text{mrad}\cdot\text{sec}^{-1}$ ) of beta-gamma-radiation at the outlet of the technological slit with its geometry,  $^{90}\text{Sr} + ^{90}\text{Y}$  irradiator activity (Q, curie) and the effective atomic number (z) of the material from which the inner lining of the irradiation chamber is made, is the following:

$$P_{\beta+\gamma} = \frac{K \cdot g \cdot (\bar{Z})^{0,27} \cdot Q}{t^2} (4e^{-0,6x} + e^{-0,25x}) \quad (5)$$

It can be assumed with sufficient accuracy for practical aims that  $K (\bar{Z})^{0,27} = 3,4 \cdot 10^2$  (k - the proportionality coefficient) under the following conditions: the irradiator is flat and parallel to the technological slit; the irradiator and the shielding are made of aluminium, stainless steel, lead. In cases corresponding to the given assumptions made on the basis of analyzing the technological schemes of beta-irradiators use, the calculated by the formula (5) radiation dose rate exceeds the measured one on 15 + 20% (14).

Studies of the ways of radioactive contaminants formation during the operation of the so-called "closed" radiation sources were made. It was caused by the facts of the unsealment of the radiation sources used in large gamma-installations that was confirmed by a number of other papers.

According to the technological scheme of reciprocal movements of an irradiator and irradiated object large isotopic radiation

installations may have fixed and mobile irradiators. By the type of shielding we can classify them as the installations with dry, liquid (water) and mixed shielding, that allowed detailing the mechanisms of radioactive contaminants formation during the operation of different installations.

The radioactive contaminants at the gamma-installations with dry shielding can appear as a result of the source unsealment (plants with the fixed irradiator) or residual radioactive contaminant dispersion from the sources shell (plants with mobile and immobile irradiator). In this case the dispersion coefficient of surface contamination ( $\mathbf{E}$ ,  $\text{m}^{-1}$ ) can be used as a qualitative characteristic of aerosol formation intensity. As a result of model and production research we have got an expression for defining total activity of wastes  $Q$  (curie) formed at the installations with dry shielding for the time  $t$  (15,16):

$$Q = \mathbf{E} \cdot t \cdot V \left\{ (n - n^1) \bar{C} + \left[ \sum_i C_i S_i + a_0 e^{-\frac{0,693 \cdot t^1}{T_{1/2}}} \cdot \frac{S}{S_0} \right] n^1 S_0^{-1} \right\} \quad (6)$$

where:  $\bar{C}$  - the average surface contamination of one sealed source, curie/ $\text{m}^2$ ;

$n$  - the total number of sources in the installation, pieces;

$n^1$  - the number of unsealed sources, pieces;

$V$  - the volumetrical rate of the cooling air,  $\text{m}^3 \cdot \text{h}^{-1}$ ;

$S_0$  - the surface area of the source,  $\text{m}^2$ ;

$C_i$  - the radioactive contamination of separate areas of the upper and lower surface parts ( $i=1,2$ ) and side surface of the sources ( $i=3$ ), curie. $\text{m}^{-2}$ ;

$S_i$  - the area of the upper and lower parts ( $i=1,2$ ) and side surface of the sources ( $i=3$ ),  $\text{m}^2$ ;

$a_0$  - the initial activity of one source, curie;

$T_{1/2}$  - the half-life period of an isotope,  $\text{h}^{-1}$ ;

$t^1$  - the time, which has passed from the moment of the source initial activity definition, h;

$S$  - the defect area in the source shell,  $\text{m}^2$ .

By means of experiments the dispersion coefficient  $\mathbf{E} = C_0 / C$  was found, where  $C_0$  - the quantity of the radioactive material, blown from the source, curie. $\text{m}^{-3}$ ;  $C$  - the radioactive contamination of the radiation source surface shell, curie. $\text{m}^{-2}$ . The authors have shown that this coefficient value is equal to  $1,5 \pm 0,9 \cdot 10^{-6}$ ,  $\text{m}^{-1}$ . Moreover it was found that practically this coefficient depends neither on the surface contamination of the sources nor on



the air flux speed above the surface.

For the gamma-installations with water shielding the transferred to water activity value  $Q$  (curie), can be defined from the relation  $Q=b.K.s.t$ , where  $b$  - the specific activity of the radioactive material, curie/g;  $K$  - the coefficient characterizing the solution of radioactive contaminants in the liquid from the surface unit for the time unit,  $g/cm^2.day$  (paper review shows that for large gamma-installations this coefficient is accepted to be  $5 \cdot 10^{-6} g/cm^2.day$ );  $s$  - the defect value in the source shell,  $m^2$ ;  $t$  - the time of the source contact with the water medium, day.

While prognosticating the quantity of the radioactive material released into the environment it is possible to define beforehand the value of the defects. For preliminary calculations the value of a defect can be accepted in the range of  $0,5 + 1,0 mm^2$ , that approximately corresponds to the pin-hole area of the sources under testing and is similar to real conditions connected with the moment of detecting the considerable leakage of radioactive material into environment caused by the local corrosion of the source shell.

Our investigation results caused stopping the serial production of  $^{60}Co$ ,  $^{137}Cs$  radioactive isotopes and beta-sources in aluminium shells and initiated their production in stainless steel. The use of highly active sources in hermetic stainless steel shells excluded environmental radioactive contamination.

As many-years experience of using large radioisotopic installations showed our recommendations concerning organizational-technical procedures and shielding constructed in accordance with our calculations provide bringing down irradiation levels to the safe values under the normal mode of radiation plants operation. However, the accidents caused by various reasons can occur during the plants operation. By the analysis it was stated that accident reasons can be the following: the radiation sources unsealment, the shut down of the devices for the sources movement, design and construction drawbacks of the shielding and braking the safe operating procedures. Very often combination of the two or more above-mentioned reasons can be the cause of an accident (17).

Taking into account that the shut down of the blocking and signalization systems would cause radiation accidents with serious consequences, the system of blocking the entrance door "according

to the dose" was developed. This system uses automatic commutation of the dosimetry instruments sensors (18) and is designed for large isotopic installations. The proposed blocking system excludes the possibility of the personnel entering the irradiation chamber when the sources are under working conditions.

But still the existence of the smallest probability of the blocking and signalization system shut down made us to develop the dose evaluation express-method for the case of an accident exposure of the large radioisotopic installations personnel that is necessary for the timely and proper medical protection actions (19). Probability evaluation of the average and limit accident occurrence during the operation of one or a group of large radiation installations having the same constructional peculiarities showed that the most reliable are the installations with dry shielding and fixed irradiator and the installations with mechanical devices for irradiator movements; whereas the less reliable are the installations with pneumatic devices for the sources movements.

One of the possible causes of non-radiation accidents at the large isotopic installations is the personnel poisoning by the radiolysis products (ozone and nitrogen oxides) or by toxic substances either from irradiated objects or formed as a result of irradiation process. Production and model research conducted allowed to give recommendations concerning the arrangement, size and construction of inflow and outflow vent holes, and rational ventilation modes of operation in the working area of large stationary gamma-installations, that exclude the possibility of such type accidents occurrence (20).

Our investigations results for ensuring radiation safety when using large radiation sources were used by the USSR Ministry of Health which exercises state sanitary monitoring in our country.

The sanitary monitoring system adopted in the USSR supposes the development of the All-Union normative documents regulating the radiation safety conditions when carrying out design, construction and operation of radiation-hazardous objects including large radiation installations. Such regulatory documents developed for the first time in the world practice are: "Sanitary rules of the construction and operation of large isotopic gamma-installations", "Sanitary rules of the construction and operation of the reactor primary circuits", "Sanitary rules of the construction

and operation of large isotopic gamma-installations with fixed irradiator", "Sanitary rules of the construction and operation of large beta-installations", "Methodical instructions on carrying out radiation monitoring when loading, adding to load and replacing the ionizing sources of large isotopic installations". All these documents developed with our help are approved by the USSR Ministry of Health and thus became a sanitary law for all the institutions and enterprises connected with the development and operation of large radiation installations.

Such unification of demands to the large radiation installations allows not only to use optimal solutions received by research and development but to provide the united system of sanitary radiation monitoring of radiation workers, employed at these installations. The collected dosimetry data including those received by the use of the various methods of personal monitoring and counters for determining whole-body exposure allow to evaluate annual exposure doses to be in the range up to 0,5 rem/year which envisages the high reliability of the developed radiation protection system and shielding of the large isotopic installations.

#### REFERENCES

1. Bykhovski A.V., Larichev A.V., Chistov E.D. Problems of Shielding from Ionizing Radiations in Radiation Chemistry. M., Atomizdat, 1970.
2. Proceedings of the 2nd All-Union Meeting on Radiation Chemistry. USSR Academy of Sciences Publishing House, 1962.
3. Radiation Chemistry, M., Atomizdat, 1972.
4. Chistov E.D. Materials of the 1st Scientific-Practical Conference on Radiation Safety. M., 1968.
5. Chistov E.D., Terman A.V., Laricnev A.V. Scientific Papers of the Institutes of Labour Protection of the All-Union Central Council of Trade Unions. M., Profizdat, 83, 1973.
6. Arefyeva Z.S., Bochkarev B.B., Mikhailov L.M., Timofeyev L.M. "Atomnaya energiya", vol. 11, n. 2, 186, 1961.
7. Abrams I.A., Veberis O.E., Godkalne A.K., Kalis Kh.E. "Izvestiya of the Academy of Sciences of the Latvian SSR", Series of Physical and Technical Sciences, No 5, 26, 1965.
8. J. Umeda "Nuclear Science and Technology". 1, 37 (1964).

9. H.Oho, A.Tsuruo "Nuclear Science and Technology", 2 , 229 (1965).
10. Larichev A.V., Partolin O.F., Chistov E.D. "Atomnaya energiya", No 2, 26 , 155 (1967).
11. Partolin O.F., Larichev A.V., Chistov E.D. "Atomnaya energiya", No 1, 26 , 78 (1969).
12. Breger A.Kh., Gurvits S.S., Pozdnyakova L.A., Chistov E.D. "Scientific Papers of the Institutes of Labour Protection of the All-Union Central Council of Trade Unions", m., Profizdat, No4, 12 , (1960).
13. Mamin E.B., Terman A.V., Chistov E.D. "Gigiyana i sanitariya", No 4, 39 (1971).
14. Chistov E.D., Larichev A.V. "Scientific Papers of the Institutes of Labour Protection of the All-Union Central Council of Trade Unions", M., Profizdat, issue 3 , 29,49 (1964).
15. Chistov E.D., Zarayev O.M., Sprygayev I.F. Ibid issue 68,38 (1971).
16. Sprygayev I.F., Chistov E.D., Terman A.V. Ibid issue 74,56 (1971).
17. Chistov E.D., Sprygayev I.F., Terman A.V., Larichev A.V. Ibid issue 66, 28 (1970).
18. Goldin V.A., Voropayev Yu.V., Shalyapin N.K., Chistov E.D. Ibid issue 1(33), 54 (1965).
19. Chistov E.D., Sprygayev I.F., Korenkov I.P., Terman A.V., Sedov A.V. "Atomnaya energiya", issue 5, 30, 460 (1971).
20. Seredin Yu.V., Chistov E.D. "Scientific Papers of the Institutes of Labour Protection of the All-Union Central Council of Trade Unions",M.,Profizdat, issue 60, 52 (1969).

К ОЦЕНКЕ ВЛИЯНИЯ АБСОРБИРОВАННОЙ ФАЗЫ ЗАГРЯЗНЕНИЯ  
ЗАЩИТНЫХ ПОКРЫТИЙ И СРЕДСТВ ИНДИВИДУАЛЬНОЙ ЗАЩИТЫ  
НА РАДИАЦИОННУЮ ОБСТАНОВКУ В ПОМЕЩЕНИЯХ

Dr. A.L. Kononovitch  
Biophysics Institute  
USSR Health Ministry  
Moscow, USSR

ABSTRACT

This report gives a method for accounting dependence of radioactive contamination hazard on the function type of its distribution in the depth of material. The method is based on describing time dependence of distribution functions by Green's operators. The proposed procedure is simple enough and can be used by the personnel of dosimetric service which has not special mathematical training.

В ряде случаев основным фактором, определяющим опасность, которую представляют поверхности защитных покрытий и средств индивидуальной защиты, загрязненные радиоактивными веществами, является поступление этих веществ в воздух помещения. С этой точки зрения загрязнение, проинфильтрованное в глубь материалов, может считаться фиксированным. Но течением времени, выделяясь на поверхности оно создает опасность рецидива загрязнения. Вероятность перехода абсорбированного вещества в адсорбированное состояние существенно зависит от того, как это вещество распределено в объеме покрытия. В данной работе предлагается способ количественного определения зависимости степени опасности абсорбированного загрязнения от вида функции его распределения по глубине материала.

Эта задача представляет особый интерес при работе с некоторыми альфа-излучателями. С аналогичными вопросами приходится встречаться, когда имеют дело с нерадиоактивными токсичными веществами, об-

ладающими высшей проникаемостью. Следует отметить, что функция распределения альфа-активных излучателей по глубине загрязненного объекта может быть измерена по спектру альфа-излучения у поверхности, причем исследуемая загрязненная поверхность не подвергается какой-либо специальной обработке и в процессе измерения не разрушается<sup>1;2</sup>.

В данной работе использованы следующие определения. Под поверхностью понимается часть материала, доступная непосредственному воздействию моющих средств или контакту с воздухом помещения.

Считается, что токсичное вещество, сосредоточенное на поверхности или перешедшее с нее в воздух помещения, находится в опасном состоянии или опасной фазе. Токсичное вещество, недоступное непосредственному контакту с внешней средой, мы будем называть квазификсированным загрязнением.

Для оценки степени опасности требуется по заданному распределению квазификсированного вещества определить вероятность рецидива в течение времени  $t$ . В связи с прикладным характером задачи в данной работе использован приближенный полуэмпирический способ описания процесса, позволяющий на конечном этапе избежать громоздких вычислений.

Обозначим распределение квазификсированного загрязнения в начальный момент времени через  $\rho_0(x)$ , где  $x$  — пространственная координата, направленная вглубь загрязненного объекта. Пусть процессы массопереноса и обмена веществом между опасным и квазификсированным состоянием являются линейными. В этом случае зависимость функции распределения абсорбированного вещества от времени может быть представлена как результат действия оператора Грина на первоначальную функцию распределения.

Практически в результате измерения функция  $\rho_0(x)$  получается в форме набора дискретных чисел, определяющих количество активного

активного вещества, сосредоточенное в различных слоях материала. Поэтому операторы записываются в матричной форме.

$$[\rho(t)] = [G] * [\rho_0] \quad (1)$$

Необходимо подчеркнуть, что матрица  $[G]$  может быть получена непосредственным измерением или полуэмпирическим путем. В реальных условиях невозможно учесть все факторы, влияющие на перераспределение вещества в объеме материала и выделение его во внешнюю среду. Поэтому для оценки степени опасности используется оператор  $[G]$ , который описывает не реальное развитие абсорбированной фазы, а некоторый фиктивный критический процесс, а именно, при его построении полагают, что все неопределенные составляющие процесса протекают таким образом, что они максимально способствуют выделению вещества в опасную фазу. Аналогичным приемом можно учесть также и нелинейные процессы.

Количество токсичного вещества, перешедшего в опасное состояние, определяется разностью поверхностных плотностей квазификсированного загрязнения в начальный и заданный моменты времени. Оно находится суммированием всех компонент вектора  $[\rho]$  (см. выражение (1)). Изменив порядок суммирования, результат можно представить в виде произведения двух векторов.

Окончательно: Если в начальный момент времени  $t = 0$ , в материале имеется квазификсированное вещество, распределенное по закону  $[\rho_0]$ , к моменту  $t$  в опасное состояние перейдет не более чем

$$Q = [Rec(t)] * [\rho_0] \quad (2)$$

$$[Rec(t)] = [1] - [N(t)],$$

где:

$$N_k(t) = \sum_i G_{ik}$$

$[1]$  - вектор, все компоненты которого равны 1.

Следует подчеркнуть, что при умножении вектора начального распределения на вектор  $[Rec(t)]$  как правило требуется выполнить 5÷6 (не более 10) операций арифметического умножения с последующим

сложением результатов.

Необходимые значения  $[R_{ec}(t)]$  получают заранее при изучении материала в лаборатории.

В качестве примера рассмотрим зависимость степени опасности, которую представляет токсичное загрязнение, от вида функции его распределения по глубине материала при условии, что процесс массопереноса в объеме материала подчиняется закону Фика, причем поток на поверхности стремится к бесконечности.

На рис.1 показаны три графика функций распределения одного и того же количества токсичного вещества по глубине материала. Там же приведены величины, пропорциональные максимально возможной скорости перехода вещества в опасную фазу при соответствующем начальном распределении. Хорошо видно, что степень опасности, которую представляет токсичное вещество, существенно зависит от его распределения по глубине материала. В этой связи можно несколько уточнить формулировку задачи дезактивации по отношению к квазификсированному загрязнению. А именно, вместо удаления токсичного вещества, что как правило практически невозможно, достаточно изменить функцию распределения так, чтобы уменьшить вероятность перехода загрязнения в опасную фазу.

В качестве примера рассмотрим дезактивацию загрязненного материала, процесс переноса в котором подчиняется закону Фика. На рис. 2 показаны графики функций распределения токсичного вещества по глубине материала, получающихся при непрерывной длительной обработке сильным моющим средством. Зависимости получены расчетным путем. На том же рисунке дана таблица, в которой приведены полное количество загрязняющего вещества (столбец  $Q$ ), оставшееся в материале к моменту времени  $t$ , величина, пропорциональная максимальной скорости перехода вещества в опасную фазу (столбец  $\pi \frac{Q'}{Q_0}$ ). Видно, что в период времени от  $t = 0,01$  до  $t = 0,4$  степень опасно-



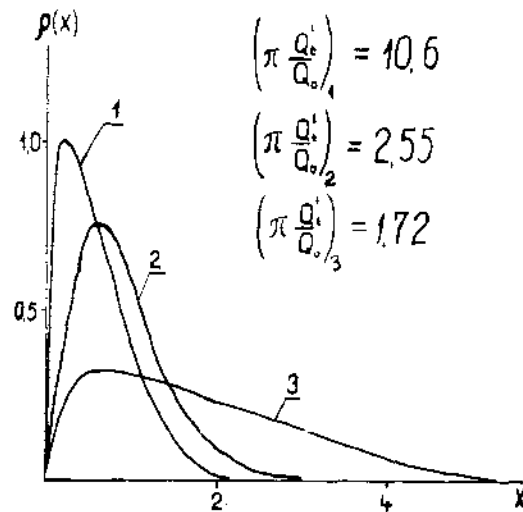


Рис. 1. Зависимость степени опасности загрязнения от функции распределения вещества по глубине материала.  
 По оси абсцисс - глубина материала; по оси ординат - плотность радиоактивного вещества.  
 Величина  $(\pi \frac{Q_t}{Q_0})$  - степень опасности для распределений вида 1, 2 и 3.

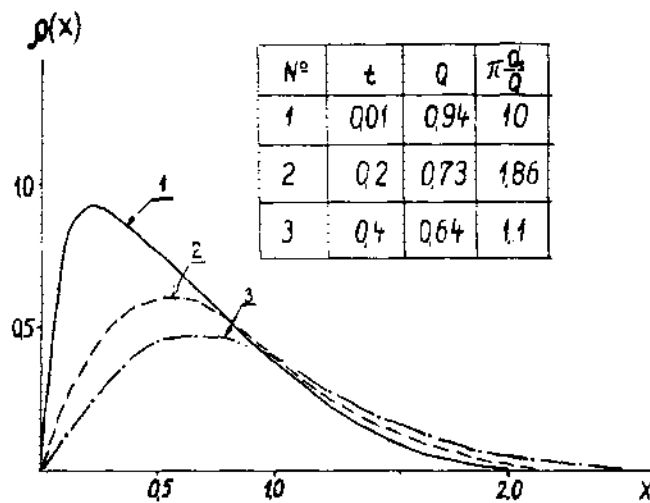


Рис. 2. Изменение степени опасности загрязнения при дезактивации.  
 По оси абсцисс - глубина материала.  
 По оси ординат - плотность радиоактивного вещества.  
 Пояснения даны в тексте.

сти убывает в 9,1 раза, и при этом удаляется только 30% загрязнения.

Применение формализма операторов Грина часто облегчает вычисление приближенных матриц  $[G]$ , а также упрощает синтез данных, если известны отдельные составляющие процесса. Например, при изучении эрозии материала в результате его истирания трудно провести эксперимент, учитывающий одновременно диффузию в объеме материала и разрушение его поверхности. Поэтому соответствующие данные удобнее получить в независимых опытах. Результирующая матрица Грина может быть найдена методом теории возмущения как оператор Грина для уравнения

$$\rho_t = H[\rho] - V \frac{\partial \rho}{\partial x},$$

где:  $H$  - оператор, учитывающий процесс массопереноса внутри материала. Как указывалось, его функция Грина предполагается известной;

$V$  - скорость разрушения поверхности.

Получение конкретных алгебраических выражений выходит за рамки данного доклада.

В результате анализа при помощи таблиц  $[Rec(t)]$  можно количественно определить степень фиксации радиоактивного загрязнения поверхности, т.е. измерить, какая часть токсичного вещества представляет реальную опасность.

#### ЛИТЕРАТУРА

1. Sittkus A., Backheer K., Staub., 23. No 9, 419 (1963).
2. Кононович А.Л., Боголапов Н.В., Ключков В.Н., Константинов И.Е. "Атомная энергия", т.33, вып.5 (1972).

ОБЕСПЕЧЕНИЕ РАДИАЦИОННОЙ БЕЗОПАСНОСТИ ПРИ ИСПОЛЬЗОВАНИИ  
ПРИБОРОВ С РАДИОИЗОТОПНЫМИ ИСТОЧНИКАМИ ИЗЛУЧЕНИЙ.

Н.В.Вершинин, Д.А.Бодрова, В.В.Волновицкая,  
П.П.Ляроцкий, П.И.Моисовицев.

Институт гигиены труда и профзаболеваний АМН СССР  
Москва

Союз Советских Социалистических Республик.

Summary.

The description of measures is given as far as prevention and current sanitary control is concerned providing radiation security using radioisotopic instruments. The demands are indicated to the sources of radiation, from instrument and their comparison with the doses of radiation.

Приборы с радиоизотопными источниками ионизирующих излучений используются во все расширяющихся масштабах в целях контроля, автоматизации и интенсификации технологических процессов, эlimинации электростатических зарядов, предупреждения несчастных случаев, пожарной безопасности и т.д. Большой контингент лиц, контактирующих с приборами, большое разнообразие и рассредоточенность приборов по объектам, широкий диапазон условий их эксплуатации возлагают повышенную ответственность на профилактический контроль качества приборов и источников, их соответствия современным требованиям радиационной безопасности.

Установившаяся в СССР система профилактического санитарного контроля новых типов радиоизотопных приборов и других изделий радиационной техники включает следующие этапы: лабораторные испытания опытных образцов, выдачу разрешений на производственные испытания опытных образцов, выдачу разрешений на изготовление и эксплуатацию установочной партии, контроль источников и приборов установочной партии на соответствие их опытным образцам, экспертизу технической документации, выдачу разрешений на серийное изготовление.

При лабораторных испытаниях опытных образцов источников проводится измерение или контроль параметров, указанных в таблице 1

Наиболее полной программе испытаний подвергаются источники альфа- и мягкого бета ( $\beta_m$ ,  $E \leq 0,25$  мэв)-излучений. Механическая, термическая и коррозионная устойчивость источников гамма, нейтронного, рентгеновского и жесткого бета ( $\beta_{ж}$ ,  $E \geq 0,25$  мэв)-излуче-

ния, имеющих одинарные или двойные ампулы из нержавеющей стали или сплавов алюминия, определяются свойствами этих материалов, которые достаточно хорошо известны. Однако, если при эксплуатации источники могут подвергаться воздействию коррозионных сред или таким механическим нагрузкам, которые способны вызвать усталостные явления, испытания производятся по полной программе.

Таблица 1.

Параметры, контролируемые при испытаниях источников.

№№	Контролируемый параметр	Источники излучения:					
		γ	n	x	β <sub>н</sub>	β <sub>м</sub>	α
1.	Выход рабочего излучения	+	+	+	+	+	+
2.	Выход сопутствующих излучений	-	-	+	+	+	+
3.	Герметичность	+	+	+	+	-	-
4.	Загрязненность поверхности	+	+	+	+	+	+
5.	Скорость выделения радиоактивного изотопа	-	-	-	-	+	+
6.	Устойчивость к ударам и вибрации	+	+	+	+	+	+
7.	Устойчивость к истиранию	-	-	-	-	+	+
8.	Термоустойчивость	-	-	-	-	+	+
9.	Коррозионная устойчивость	-	-	-	-	+	+

Что касается испытаний на устойчивость к ударам, вибрации и температуре, то принимаемые при этом испытательные нормы практически не отличаются от испытательных норм, предлагаемых в проекте 1. Испытания на устойчивость к проколам не производятся. За критерий устойчивости источников проникающих излучений принимается сохранение герметичности ампул.

У источников альфа- и мягкого бета-излучения механическая, термическая и коррозионная устойчивость обеспечивается в основном свойствами материала (керамика, эмаль, металл-матрица, окислы металлов, слоенки и т.д.), в котором заключен радиоактивный изотоп. Герметизирующие покрытия имеют вспомогательное значение, т.к. их толщина не превышает нескольких мкг/см<sup>2</sup>. Благодаря радиационно-физическим и радиационно-химическим процессам происходит ослабление связи поверхностного слоя препарата с ниже-лежащими слоями и появление на поверхности источника слабофиксированной загрязненности. К таким источникам неприменимы методы контроля герметичности, предлагаемые в проекте<sup>1</sup>, так же как и неприменимо определение закрытого источника, принятое в рекомендации ISO<sup>2</sup>.

В настоящее время в СССР принято следующее определение закрытого источника<sup>3</sup>. "Закрытый радиоизотопный источник излучения... источник, в котором радиоактивный материал заключен в оболочку (ампулу или защитное покрытие), предотвращающую контакт персонала с радиоактивным материалом и его рассеяние выше допустимых уровней в условиях, предусмотренных для использования источника".

Это определение позволяет количественно оценивать источники на соответствие их требованиям радиационной безопасности по ско-

рости выделения радиоактивного изотопа при его эксплуатации в допустимых условиях. Важной характеристикой таких источников является также количество радиоактивного изотопа, которое может выделиться из источника при аварийных нагрузках.

Исследования радиационной обстановки на производствах, использующих приборы с источниками альфа- и мягкого бета-излучения и лабораторные исследования процессов дефиксации изотопов из этих источников позволили установить предельно-допустимые скорости дефиксации (таблица 2).

Таблица 2.

Предельно-допустимые скорости дефиксации изотопов.

№№	Тип источника	Предельно-допустимая скорость дефиксации мкюри/см <sup>2</sup> месяц
1.	Из <sup>239</sup> Pu на основе эмали для нейтрализаторов электростатических зарядов.	$1 \cdot 10^{-4}$
2.	Из <sup>238</sup> Pu на основе эмали для альфа-антисцинтилляционных анализаторов.	$3 \cdot 10^{-3}$
3.	Из <sup>239</sup> Pu на основе глазури для дымоизвещателей.	$5 \cdot 10^{-4}$
4.	Из трития на титане для нейтрализаторов электростатических зарядов.	1

При таких скоростях дефиксации изотопов из источников уровни загрязнения производственной среды остаются в пределах допустимых для неконтролируемых зон.

Источники альфа- и мягкого бета-излучения в процессе эксплуатации требуют периодической очистки от пыли и загрязнений. В связи с этим они подвергаются испытаниям на устойчивость к истиранию сухой фильтровальной бумагой под давлением до 0,5 кг/см<sup>2</sup> поступательным или вращательным движением в количестве до 1500 циклов.

Коррозионные испытания таких источников проводятся в основном для выяснения их устойчивости во влажной атмосфере при повышенных температурах (98% относительная влажность при 40°C).

При лабораторных и производственных испытаниях опытных образцов приборов производится определение уровней излучения от прибора и при необходимости - измерение доз облучения персонала, занятого эксплуатацией оборудования с установленными приборами.

По максимально допустимым уровням излучения приборы разделяются на 2 группы: к 1-ой группе относятся приборы, нормальная работа с которыми может привести к облучению в дозах, превышающих предел дозы для категории "отдельные лица из населения" (гамма-дефектоскопы, гамма терапевтические аппараты, приборы для исследования буровых скважин).

Во второй группе - приборы, при работе с которыми доза облучения не выходит за предел дозы для указанной категории. В эту группу входит большинство приборов технологического контроля, нейтрализаторы электростатических зарядов с источниками из плуто-

ния-239 и др. Целесообразно также выделить третью группу - такие приборы (дымоизвещатели и др.), которые в допустимых условиях эксплуатации не могут дать дозу облучения свыше генетически значимой дозы для населения (таблица 3).

Таблица 3.  
Максимально-допустимые уровни излучений  
от приборов

Группа №	Мощность дозы, мбэр/час.	
	на поверхности	на расстоянии 1 м
1	-	3
2	10	0,3
3	0,3	0,01

Фактические дозы облучения персонала, занятого эксплуатацией приборов 1 и 2 группы (таблица 4) значительно ниже соответствующих нормативов.

Таблица 4.  
Дозы облучения персонала при работе с  
радиоизотопными приборами.

№№	Приборы (аппараты)	Группа	Доза облучения, бэр/год
1.	Нейтрализаторы электростатических зарядов с источниками из плутония-239	2	0,1 - 0,2
2.	Приборы технологического контроля с источниками бета-излучения.	2	≤ 0,1
3.	Приборы технологического контроля с источниками гамма-излучения	2	0,1 - 0,3
4.	Гамма дефектоскопы стационарные.	1	0,5 - 2
5.	Гамма дефектоскопы переносные	1	2 - 3
6.	Приборы для гамма и нейтронного каротажа скважин	1	1 - 3

Помимо профилактического контроля, органы санитарной службы СССР осуществляют текущий (периодический) надзор за радиоизотопными приборами: за соответствием условий их эксплуатации допустимым условиям, за правильностью учета и проведением дозиметрического контроля (для приборов 1 группы), а также проводит расследование каждой аварийной ситуации, т.е. любого отклонения от нормального процесса эксплуатации прибора, которое может создать повышенную радиационную опасность для работающих или населения.

Анализ аварийных ситуаций с приборами показывает, что основными причинами их являются отклонения фактических условий эксплуатации от допустимых условий для данного типа источников и приборов (главным образом по механическим и коррозионным воздействиям).

По мере накопления опыта в эксплуатации приборов данного типа происходит снижение числа аварийных ситуаций. Так при десятикратном увеличении ежегодных поставок приборов абсолютное число аварийных ситуаций практически не увеличилось.

#### Литература.

1. Draft ISO Standard for Classification of sealed, Radioactive sources ISO/TC 85/SC4/WI1 (Sec-57)77.
2. Рекомендация Международной организации по стандартизации Р 921 "Словарь по ядерной энергии", 1969 г.
3. Государственный стандарт Союза ССР "Источники излучения радиоизотопные закрытые. Термины и определения". ГОСТ № 16445-73.

HEALTH PHYSICS AND OPERATIONAL EXPERIENCES IN A TREATMENT AND PACKAGING  
FACILITY FOR SOLID AND SEMI-SOLID WASTES AT E.I.R.

J.Schwammberger, H.Brunner

Health Physics Division  
E I R, Eidg. Institut für Reaktorforschung  
CH-5303 Würenlingen, Switzerland

1. Summary and conclusions

Solid or semi-solid wastes have either to be stored for long periods or to be disposed by dumping into the ocean or enclosure into suitable geological formations. Each case requires an appropriate treatment and packaging of the wastes. Despite the relatively low specific activities there exist considerable radiation hazards for the operating personnel due to incorporation risks. At EIR Würenlingen a waste treatment facility was built that reduces such hazards to acceptable levels. The main installation is a large combination of steel boxes, well ventilated and held at reduced pressure. Operations that can be carried out include sorting of wastes, compression of loose wastes or drums, dismantling or reduction to small pieces, solidification of sludges with cement, casting of concrete mantles and covers etc. All direct contacts between personnel and radioactive material are avoided. Either the men work from outside the box with gloves or remotely operated hydraulic tools or machines, or they work inside the box with respiratory protection or in ventilated plastic suits. An incinerator plant under construction will bring further volume reduction of burnable waste, even for  $\alpha$ -activities. Our experiences are reported and show that radioactive wastes of low or medium specific activity can be treated and packed according to any requirements without hazards for the operating personnel, the surrounding population or the environments of the treatment facility or the storage sites.

2. Purpose of the laboratory and choice of the concept

Most wastes are produced and collected in forms that do not allow storage for more than a few years. For storage over extended periods of years or final disposal a treatment of the wastes is indispensable. Years of improvised waste handling experience and an extensive literature search resulted in specifications and an operations plan for each treatment method considered, from which the necessary working conditions and installations could be derived. Thus we developed a concept for an installation that should best suit our needs. The laboratory came into operation in 1970 and can be used for the following methods :

- sorting of mixed wastes, mainly into four groups: combustible, non-combustible, compressible, non-compressible;
- baling of wastes by pressing for volume reduction;
- solidification of liquid or semi-liquid wastes (sludges) with cement;
- dismantling and cutting of large waste items (filters, components, plastic sheets) into small pieces that fit into 100 or 200 liter drums;
- enclosure of wastes in cement or concrete.

The wastes must be packaged into steel drums or prefabricated concrete containers according to international transport regulations and requirements for fi-



nal disposal such as dumping into the deep ocean. Most of the wastes were low- and medium-level wastes from EIR operations (average input 100 m<sup>3</sup>/yr), but also wastes from other sources (some 40 m<sup>3</sup>/yr) such as industry, research institutes, hospitals and in smaller amounts from nuclear power stations were treated. About 10 percent were medium-level wastes. The laboratory is only in operation during the regular working hours. The crew consists of a supervisor, a health physics technician and four to five operators. In 1971/72 wastes were treated during an effective average of 30 weeks per year, the rest of the time was used for revisions, decontaminations, vacations etc. With an average yearly production of 650 drums (200 liter) of conditioned low-level waste and 80 concrete containers of medium-level wastes the capacity of the laboratory is not fully utilized. By increasing the number of operators and working during two shifts a day an input of 400 - 500 m<sup>3</sup>/yr of unconditioned wastes could be handled.

### 3. Description of the laboratory

The laboratory was installed in an already existing massive one-story reinforced concrete building, 22 x 17 x 4.5 meters. An annex contains the installations for cement mixing and the feed pump. The building (plan, Fig.1) is divided into a large storage, loading and operating hall, entrance and exit lock areas, active and inactive change rooms and office, and a personnel access and control lock. The ventilation system is housed in two rooms on top of the change rooms, together with the breathing air supply and the continuous exit air monitor.

About half of the operating hall serves as a temporary storage and transfer area for incoming wastes. The other half is the main working area with a large (8 x 6 x 3.3 meters) combination of massive airtight steel boxes, consisting of a transfer box for loading wastes either into the sorting or the operations box, a sealed operations box, a sorting box, a baling press box and a cement box with an exit lock. The boxes are connected by hydraulic mesh doors with electrical interlocks between the inner and outer doors in order to avoid operating errors and spreading of contaminations. The sorting box (Fig.2) contains a foot-pedal-controlled conveyor belt with three external working places and sealed connections for drums that can be removed into the operations box. The conveyor discharges into the press box. The sorting box is protected by two independent fire extinguishing systems with CO<sub>2</sub> and water. The baling press box (Fig. 3) accommodates a 100 ton press where loose waste or filled 100 liter drums can be compressed to 1/5 or less of the original volumes. A lucite hood is lowered on top of the baling container during pressing and the air expelled from the compressed waste is directly drawn into the exit air system. In a feed loop prefabricated cement is circulated by a pump between the cement preparation annex and the cement box where it can be filled by simple and safe means into the steel drums containing the conditioned waste and compacted by a vibrator. Sorting, press and cement box operations are done from outside from the operating hall by means of glove openings or hydraulic controls (Fig.2). All other operations such as dismantling, cutting, filling of compressed drums into larger drums are carried out in the operations box (Fig. 3). Access for working inside the sealed operations box is only possible through the personnel access lock (Fig.4) and the operators are dressed in proper protective clothing. The breathing air equipment can supply up to five operators in fully ventilated suits. In case of a power failure two reserve tanks with compressed air allow an evacuation of all five workers without any hurry or hazard by the usual procedures through the access lock. The latter serves as the control room for the supervision of operations inside the operations box with all the necessary equipment for voice-controlled intercommunication with and between individual or all operators. The intercom cables are located in the air hoses of the ventilated suits. Special quick-connect contamination-proof couplings and parallel air and intercom connections in the control lock and the box permit individual adjustment of intercom and air supplies before the operators enter the box.

All parts of the building are also connected by a loudspeaker intercom system.

The whole laboratory is very well ventilated with about 20 air changes/hr. Filtrated and if needed warm air is fed into all rooms and is then sucked through three sets of absolute filters into the operations box and from there into the other boxes. The exit air ventilators draw the air from the sorting, press and cement boxes through two filter stages with glass fiber absolute filters, of which one filter unit is located directly after each box while the second stage unit is in the exit air duct before the ventilators. Ventilation control keeps the boxes at reduced pressures of 10 - 30 mm water gauge pressure difference.

#### 4. Equipment for radiation protection

Incoming "hot" waste drums and components can be temporarily stored in the operating hall behind a 60 cm thick concrete wall. Shielded transport containers are available for transference of medium-level wastes. The boxes are made of 5 mm thick steel sheets without additional shielding, but for the treatment of medium-level waste mobile lead shields of 5 cm thickness can be installed at the respective working places. The baling press is connected to the cement box by a rail track with a hydraulically operated trolley (Fig.3) carrying a container with a cylindrical 6 cm thick lead shield which takes up the drums for pressing, concreting and unloading through the exit lock of the box.

At the three most exposed locations outside the boxes GM counter  $\gamma$ -dose rate monitors are mounted. In the storage/loading area and the exit area of the operating hall and in the control lock sets of portable instruments for  $\beta/\gamma$  dose rate and  $\beta/\gamma$  and  $\alpha$  surface contamination monitoring are available together with shielded detectors for wipe tests and air samples. Exit contamination control of the personnel is done by background-compensated  $\beta/\gamma$  hand/foot monitors in the active change room and the operating hall, a scintillator  $\alpha$  hand monitor in the active change room and by very sensitive  $\beta/\gamma$  and  $\alpha$  hand/foot monitors at the exit of the inactive change room. The air in the operating hall is continuously sampled and the filters are periodically checked for  $\alpha$  and  $\beta/\gamma$ . The exit air activity from the boxes is continuously monitored and registered for  $\alpha$  and  $\beta/\gamma$  activities. The exit of the operations box and the active change room are equipped with showers. A complete and sufficient stock of radiation protection materials is maintained.

#### 5. Operational and health physics techniques

The wastes are delivered into the operating hall in closed steel drums, in some special cases in sealed plastic bags. All treatment operations are executed inside the boxes. The interlocked sash doors permit safe transfers without leakage of activities. If the large door of the operations box has to be used, a plastic tent is connected as a temporary lock for the transfer of large equipment or waste items. All rooms are regularly controlled by wipe tests. Special care is given to the transfer of filled containers from the box to the exit lock area. Drums are washed with water in the box and transferred wet. When leaving the exit lock of the cement box they are immediately checked for contamination at the entire surface. This is done in the operating hall where final decontamination can be done if necessary, before a crane transports the containers to the exit lock area.

Working in the operating hall and at the sorting, press and cement boxes is done in ordinary coveralls without additional protective garments. For work inside the operations box the operators change completely into special underwear, coveralls, rubber boots, hoods and gloves, for wet work also disposable plastic suits. The normal respiratory protection is the army gas mask, fully ventilated protective suits of EIR designs are only worn for Tritium, Carbon-14 or Radium wastes. All protective clothes offer relatively comfortable working conditions for periods of 2 1/2 to 3 hours between half-hour or longer breaks. When leaving the operations box the operators take off clothes and respiratory protection in the access lock and undergo there a coarse contamination control. After a shower

in the active change room they check again for contamination and leave for the inactive change room where they put on their own underwear and ordinary working or street clothes. A final contamination check follows at the exit of the inactive change room. When using ventilated suits the men take a shower with the suits on at the exit of the operations box (Fig.4) before entering the access lock. There the suit is taken off with the help of another man wearing mask, hood and gloves, who makes the first contamination check. All complicated or hazardous operations are supervised and monitored by the crew's health physics technician. But each crew member completed a 4 weeks' radiation protection course at EIR and has to take care of his own radiation protection and monitoring. The supervisor of the laboratory is also a fully trained health physics technician. Independent controls of working environment and methods are performed by the working place survey group of the Health Physics Division.

For personnel monitoring direct reading pocket chambers ( 200 mR ) and film badges with quarterly evaluation are used, supplemented by TLD chips in finger-rings which we designed to stand heavy mechanical work without damage to chips or gloves. Incorporation monitoring is done by periodical urine analyses, after risky operations or incidents by special investigations and whole body counting.

#### 6. Operational experiences

Radionuclide composition, physical and chemical forms of the wastes vary a great deal and require very different treatment and packaging methods which, except the pressing, must be manually executed. A rationalized conditioning by special remotely or automatically operated equipment is out of reach for an installation of this size for space and cost reasons. In 1971 and 72 wastes with the following main activities were treated :

$\alpha$ -emitters (mostly Pu)	ca. 10 Ci	:	Tritium	ca. 260 Ci
Radium	ca. 1 Ci	:	Carbon-14	ca. 3 Ci
mixed fission products + $^{60}\text{Co}$	ca. 660 Ci	:		

The average whole body doses of the personnel due to external exposure were about 1.5 rem/yr and were similar for the entire crew. The incorporation monitoring showed no values above the investigation levels, most of them could be interpreted as representing less than 1 percent of the MPBBs. These minor incorporations resulted from Iodine-131 or Tritium work in the operations box.

No air contaminations outside the boxes in the operating hall have been found, and surface contaminations outside the boxes are extremely rare events at harmless levels less than five times the operational guides for uncontrolled zones. This proves that practically no contaminations are spread from the interior of the boxes to the outside. The reasons for this are: an excellent ventilation and relatively large negative pressure differences in the airtight boxes, rigorous and disciplined controls of personnel and material at the exits of the boxes, and frequent coarse decontamination of the boxes before the contamination levels become too high. A certain hazard exists for the hands of the operators due to relatively frequent damaging of gloves. If no wounds are inflicted, washing is in most cases sufficient for decontamination. Only in three cases the hands had to be decontaminated by a specialist from our first aid and personnel decontamination team. One operator received a cut into the hand from an Iodine-131 contaminated item. The wound was washed with saline solution and surgically cleaned by a physician. This was the only, minor incident and had no lasting consequences.

Our experiences have been excellent, the concept chosen is well suited to our needs. Summing up : good protection of the personnel depends on a reasonable combination of installations that are appropriate to the tasks and a good and disciplined working technique with reliable radiation protection and monitoring.

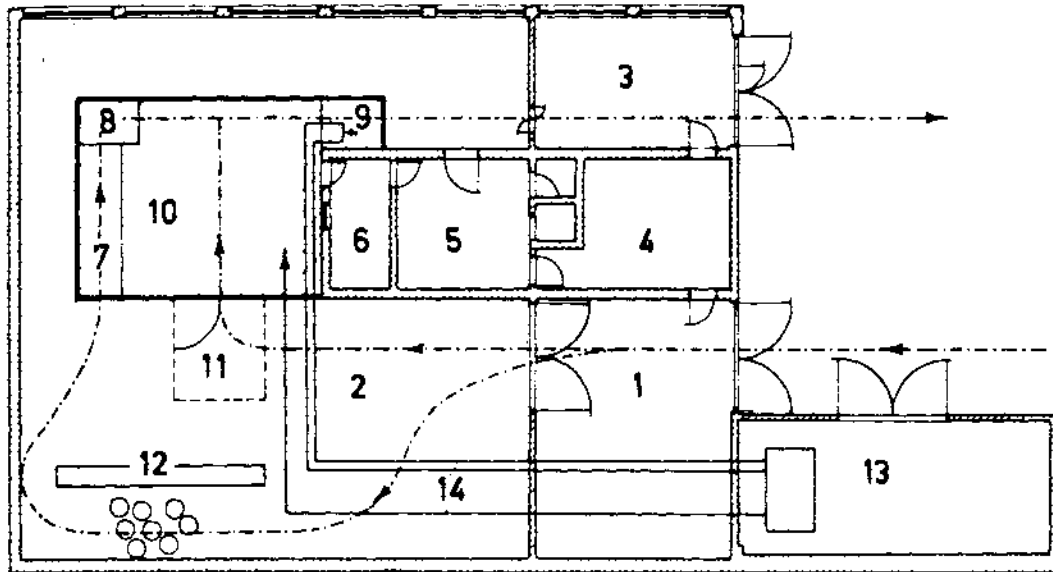
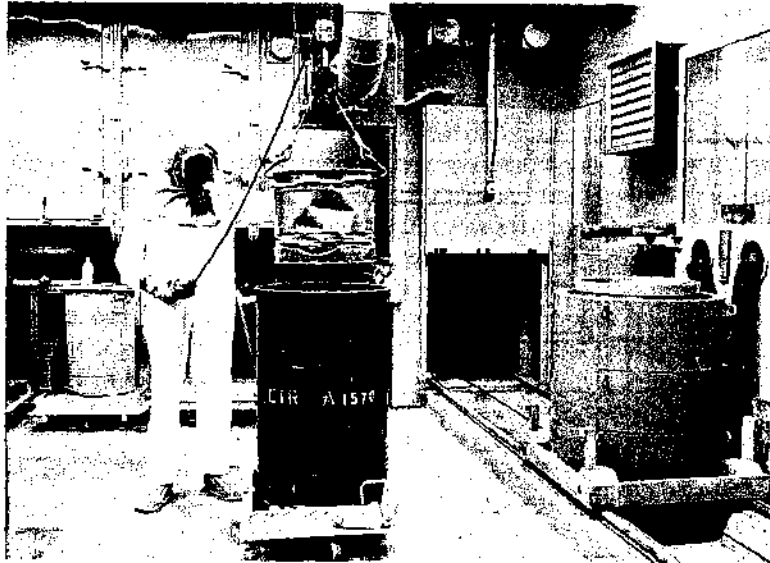


Fig. 1 : EIR waste treatment laboratory, floor plan

- |                                      |                                      |
|--------------------------------------|--------------------------------------|
| 1 vehicle + waste entrance lock area | 8 baling press box                   |
| 2 storage, loading + operating hall  | 9 cement box                         |
| 3 waste exit lock area               | 10 sealed operations box             |
| 4 inactive change room               | 11 protective tent at large box door |
| 5 active change room, shower         | 12 shielding of incoming waste store |
| 6 personnel access + control lock    | 13 cement preparation annex          |
| 7 sorting box                        | 14 cement feeding tubes              |



Fig. 2 : Exterior of the sorting box and the transfer box. An operator is loading a plastic bag into the sorting box, where another operator is sorting the waste from the conveyor belt into several drums or the press box (left background). The lower, closed sash door is used for the transfer of drums into the operations box, visible through the window at the right.  $\gamma$ -dose rate monitors are mounted at the working face of the boxes.



**Fig. 3 :** Interior of the operations box. A 100 liter drum (only partially compressed for the sake of clarity) has been removed from the baling and transfer trolley with its shielded container (at the right on the track). The sash door of the baling press box is kept open, normally it would be closed during that phase (right background). The right hand wall has a series of windows and glove ports for the external operator of the press and the transfer trolley. An entrance air filter is also visible. In the background at left the wall of the sorting box is seen with an exit air duct and the attachments and trolleys for the sorting drums below. The operator wears the usual dress for work inside the box.



**Fig. 4 :** Exit of the operations box to the personnel access and control lock. An operator in a fully ventilated PVC-suit with clear hood and intercom takes a shower before leaving the operations box through the door into the access lock. Behind the window the operator in the control lock has intercom contact with the operators in the box and regulates the air supplies. Five air and intercom hoses are connected at the right of the window. The hoses are disconnected from the suits before leaving the box. Decontaminating material and a contamination-protected fire extinguisher are below the window.

SOME PRELIMINARY INVESTIGATIONS ON THE CONTRIBUTION  
OF MUONS TO THE STRAY RADIATION LEVEL AROUND THE  
CERN 28 GeV PROTON SYNCHROTRON

M. Höfert and J. Baarli  
CERN  
Geneva, Switzerland

Abstract

Below a few GeV the predominant radiation in the environment of accelerator installations is stray neutrons. Muons may however become dominant if pions and kaons produced by the interacting primary beam are allowed to decay in the course of free flight and the resulting muons in forward direction are not sufficiently shielded.

Measurements with a counter telescope allowing for the determination of their angular distribution behind the shield around the beam direction are reported. The attenuation length for muon spectra from the decay of pions of a few GeV in several materials was determined and is compared with theoretical values. The measurements show in addition the contribution to stray radiation levels by other components penetrating the main shield of the accelerator. Their relative importance at different distances and their environmental impact are discussed.

Introduction

Radiation protection measurements around multi-GeV proton accelerators are difficult due to the largely unknown mixture of stray radiation outside the shielding. This is caused by the interactions of primary protons with target and shielding materials giving rise to a variety of secondary radiations covering a wide energy range. The predominant penetrating component of the dose measured is generally neutrons.

The contributions of muons, however, may become important if pions and kaons produced in the interaction of the primary beam are allowed to decay in the course of free flight into mu-mesons. A certain attenuation of the latter component projected in the forward direction requires more shielding than is needed for the parent pions, since the muons -- up to the energy range which is important for our problem -- only interact electromagnetically whereas the hadrons in addition undergo nuclear interactions.

The attenuation of hadrons is predominantly exponential in the shielding and may be described by a single parameter, i.e. the attenuation length  $\lambda$  which is about  $130 \text{ g cm}^{-2}$  in iron shielding for proton beams of 10-300 GeV.

Although the attenuation curves for muons (calculated and measured) turn out not to be simply exponential, they still may be characterized

over certain depths by one single parameter. Apparent attenuation lengths reported<sup>1-3</sup> for muons from primary proton energies of 25 GeV range between 430 and 900 g cm<sup>-2</sup>. These values are influenced by the choice of the production formulae of pions, their decay path (length and angle) and the resulting muon spectrum but seem less influenced by the shielding materials used in the calculations.

#### Experimental Equipment

A counter telescope was constructed to detect muons resulting from the decay of pions and kaons in the forward direction downstream of targets and after penetrating the shielding wall of the accelerator enclosure.

Three scintillation counters in triple coincidence of 5 cm width, 20 cm height and 2 cm thickness are mounted on a supporting bar at a distance of 75 cm from each other. The supporting bar pivots around a central axis in the horizontal plane and can be moved in the vertical plane as well. The pulses observed from the telescope are counted and are stored according to their height in a 256 multi-channel analyzer with a 100 MHz ADC. All measurements are made with reference to a two-fold coincidence monitor counter left in a fixed position in the radiation field during the measurements. This is done in order to take into account intensity variations of the accelerator.

#### Results

The equipment was tested first in a series of measurements performed at an angle of 6° with respect to an internal target at a distance of 45 m. The total shielding thickness between target and detector amounted to only 3000 g cm<sup>-2</sup>. A typical recorded spectrum is shown in Fig. 1. This consists of a peak at low channel numbers corresponding to a small stopping power or minimum ionizing particles and a long tail with higher stopping power.

When additional shielding material (lead, iron) is placed in front of the first counter, a second peak at higher channel numbers appears and the first peak decreases (Fig. 2). The conclusion of this effect is as follows: the radiation penetrating the shielding wall is muons "contaminated" with hadrons. Different kinds of particles in the GeV range however have roughly equal stopping power, about 2.2 MeV g<sup>-1</sup> cm<sup>-2</sup>, in a scintillator and will thus be found grouped around the same channel numbers. By introducing shielding the hadrons produce forward-peaked secondaries of lower energies that build up the second peak as these cause a higher energy loss in the detector.

By increasing the shielding thickness this second peak is found to be attenuated with a  $\lambda$  corresponding to hadrons, whereas the first peak decreases with an attenuation length which approaches a value expected for muons.

The apparent attenuation lengths for muons determined from this experiment were 379 g cm<sup>-2</sup> for lead and 457 g cm<sup>-2</sup> for iron.

A second series of experiments was performed using a test beam tuned to 19 GeV/c pions. The same build-up phenomenon was observed. Attenuation lengths for muons gave in this case values of 417 g cm<sup>-2</sup> for lead and 491 g cm<sup>-2</sup> for iron, in other words slightly higher than in the preceding experiment. From these measurements a superiority of lead of

about 20% compared to iron was found for the attenuation of muons, whereas the error in the determination of the parameter  $\lambda$  is of the order of 10%.

In a third series of measurements an attempt was made to detect muons behind a beam stop in an experimental hall. An extracted beam from the PS interacted with a target located 23 m upstream of the shielding (Fig. 3). Spectra were recorded in seven positions and all of these showed a typical Landau distribution with no "tail" towards higher channel numbers. Figure 4 is shown as an example.

When the counter telescope was turned in the horizontal plane around its vertical axis, the maximum count rate was observed when the instrument pointed in the direction of the target. The angular dependence of the muon intensity, for example for position 6 (Fig. 3), is shown in Fig. 5. The angular response of the telescope for monodirectional radiation has the shape of a triangle and is also given in the figure. It was shown by calculations that the measured angular distribution is not distorted by this response function and thus corresponds to the actual one.

A more complicated intensity distribution of the muons, which could be decomposed into three peaks, is observed at for example position 2 (Fig. 6).

Figure 3 indicates the positions in which observations with the telescope were carried out. The arrows point in the direction of measured maximum muon intensities; their lengths have been drawn proportional to the height of the measured peaks. The extensions of these arrows pass through the target -- the main source of pions and subsequently muons -- or point to some weakness in the shielding, for example in the direction of reduced thickness of iron.

Using the CERN radiation survey method<sup>4</sup> a dose rate of 1.36 mrem/h was measured in position 1. According to the results 42% of the dose rate is due to fast neutron and high energy particles, the rest is made up of charged particles and  $\gamma$  rays. The integration of the angular distribution for muons amounts to 0.33 mrem/h with the usual conversion of 10 muons  $\text{cm}^{-2} \text{s}^{-1}$  corresponding to 1 mrem  $\text{h}^{-1}$ . Their share of the total dose rate passes from one fourth to one half at greater distances, i.e. outside the experimental hall along the axis of propagation for the muons, as the fast neutron and high energy particle component falls off rather rapidly. Muons, although rather localized, will become the dominant component of stray radiation at even greater distances.

#### Acknowledgements

The authors would like to thank Mr. L. Andersson for his work concerning the electronics of the equipment, Dr. Z Khawza and Mr. M. Nielsen for their participation in the measurements and evaluation of the results, and Dr. B. Schorr for his mathematical help in the unfolding of the angular distribution.



References

1. D. Keefe and C.M. Noble, Radiation shielding for high energy muons: The case of a cylindrically symmetrical shield and no magnetic fields, UCRL-18117 (1968).
2. J. Ranft, Monte Carlo nucleon-meson cascade calculation in a block of shielding material. Description of the computer programme TRANSK, CERN MPS/Int. MU/EP 67-6 (1967).
3. R. Bruns et al., Search for intermediate bosons in proton-nucleon collisions, Columbia Univ. Rep. NEVIS-153 (1966).
4. M. Höfert, Dose equivalent and quality factor of radiation from high energy accelerators, Proc. First Symposium on Neutron Dosimetry in Biology and Medicine, p. 873, Munich 1972.

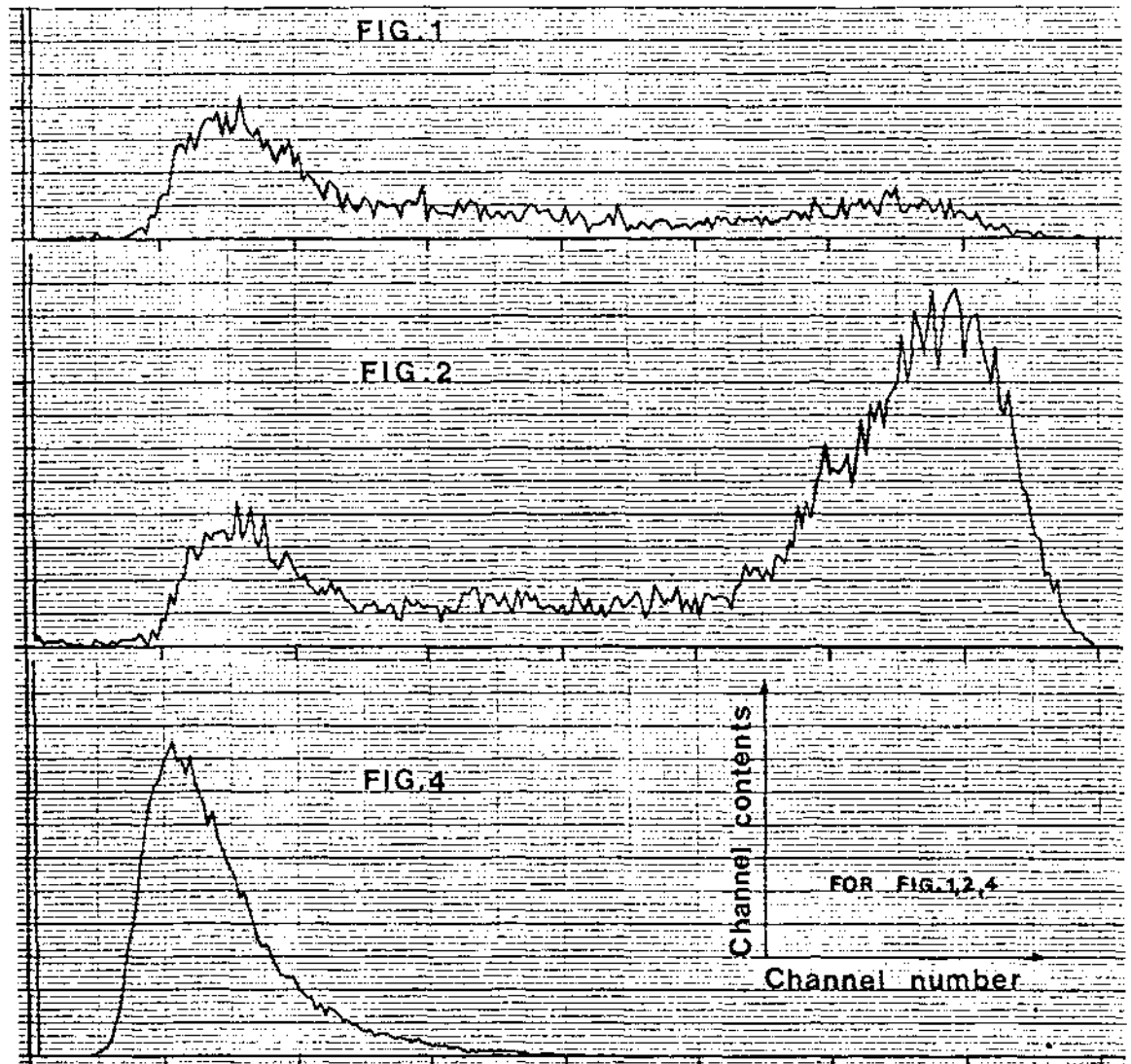


Fig. 1. Pulse height spectrum of events recorded with counter telescope behind 3000 g cm<sup>-2</sup> shielding material at 6° from an internal target.

Fig. 2. Same as Fig. 1, but with 5 cm of lead in front of the first scintillator.

Fig. 4. Pulse height spectrum of events from mu-mesons as measured in position 5 shown on Fig. 3.

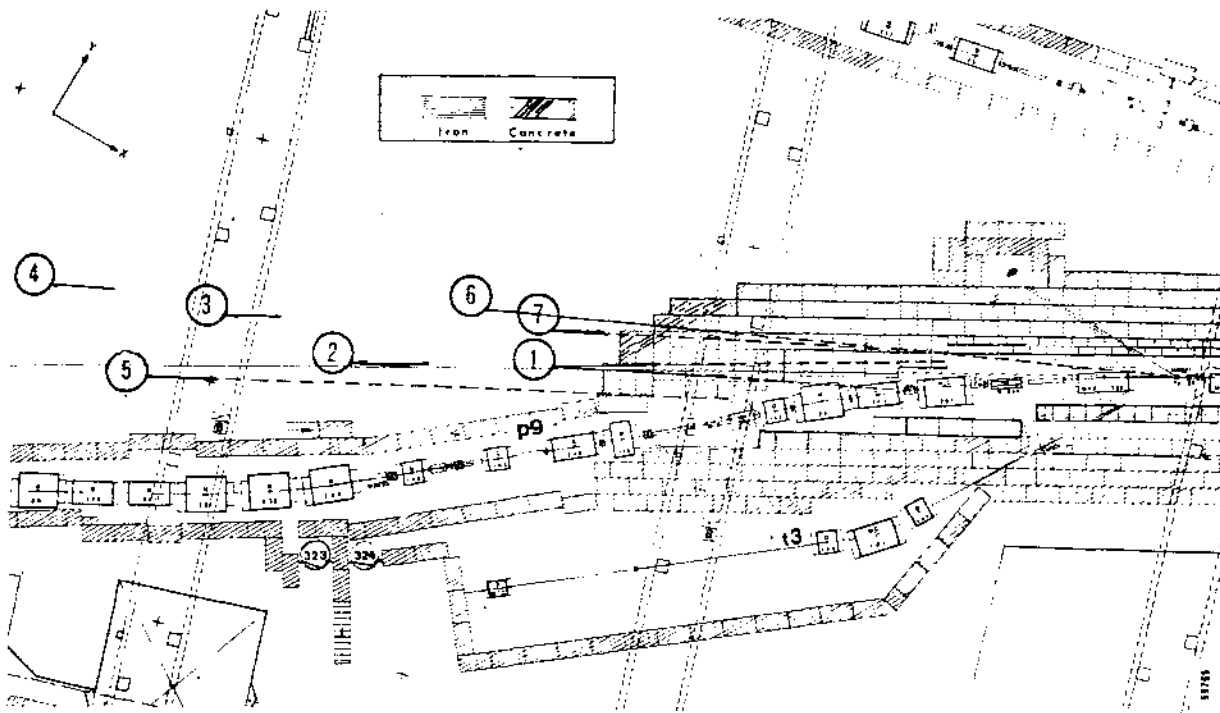


Fig. 3. Mu-meson measurements in the west experimental area downstream of an external target behind an end-stop made out of iron and concrete shielding material. The length of the arrows corresponds to the muon intensity; they are pointing in the direction of the maximum.

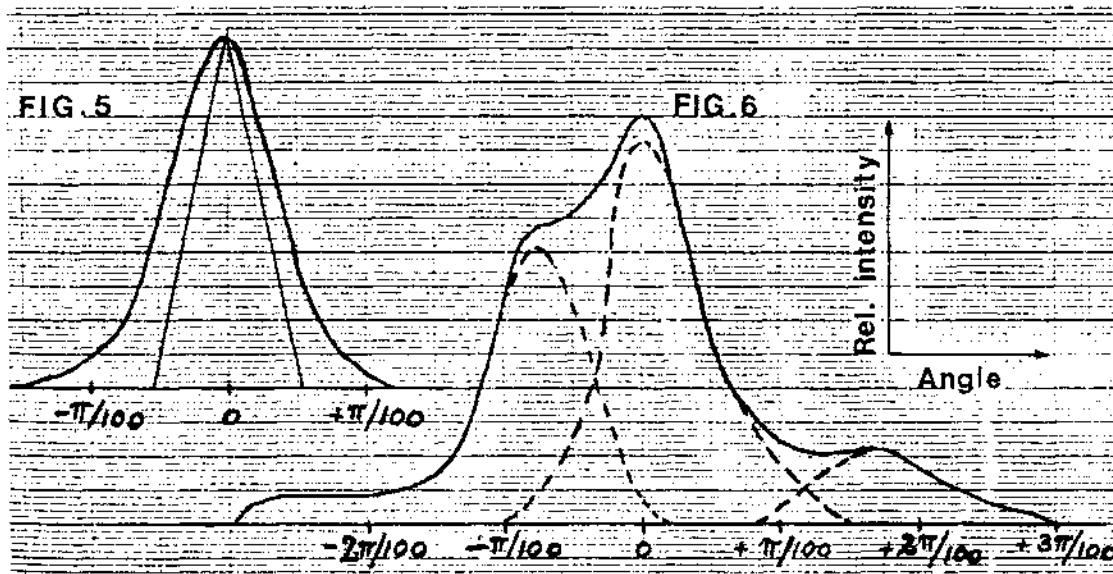


Fig. 5. Angular distribution of mu-meson intensity in position 5 of Fig. 3. The angular response function of the counter telescope having the shape of a triangle is also shown in this figure.

Fig. 6. Angular distribution of mu-meson intensity as measured in position 2 of Fig. 3.

## RADIOLOGICAL SAFETY EXPERIENCE IN HANDLING AND FABRICATION OF PLUTONIUM FUEL ELEMENTS

S. Janardhanan, T. N. Krishnamurthi and S. D. Soman  
Health Physics Division  
Bhabha Atomic Research Centre  
Bombay-400085, India

### Abstract

The problems and experience gained in radiological safety during  $\text{PuO}_2$  fuel fabrication for 'PURNIMA' reactor are discussed in this paper. Safety aspects in design, construction and commissioning of metallurgical facilities and glove boxes, measures for contamination control, air, area and criticality monitoring instrumentation and their calibration are dealt with. Radiological health data are summarised to reflect the adequacy of precautionary provisions followed in fuel fabrication work. Evolution of the methods for related safety problems like assessing neutron emission from  $\text{PuO}_2$  fuel pins and Pu in solid wastes are outlined.

### Introduction

Radiometallurgy Laboratory at the Bhabha Atomic Research Centre has been operating a plutonium fuel fabrication facility during the last two years involving handling and storage of kilogram amounts of plutonium. The facility has fabricated the complete core charge of  $\text{PuO}_2$  pins for the zero power fast reactor 'PURNIMA' at Trombay.

The fuel pin of 'PURNIMA' has stainless steel cladding and consists of a central  $\text{PuO}_2$  core of 180 mm length followed at either end, by a molybdenum plug of 80 mm length and stainless steel end plug to serve as axial reflectors in the reactor. The pin is of 11 mm diameter and of 495 mm overall length and has a plutonium loading of 123.5 g. The complete core loading called for production of 178 full fuel pins, 4 half and 4 quarter fuel pins. In addition, seven Pu-Be start-up source pins, identical in shape to the  $\text{PuO}_2$  fuel pins, were also fabricated. This paper deals with the safety aspects in the design of the handling facilities and the experience gained in the radiological safety during the fabrication campaign.

### Fuel Pin Fabrication

The feed powder  $\text{PuO}_2$ , after milling, granulation, etc. is loaded in a suitable die and pressed with a 10 ton hydraulic press, located in a glove box. The green pellets, thus obtained, were sintered in an argon and 8% hydrogen atmosphere in a molybdenum furnace inside a glove box. After sintering the length and the weights of the pellets were checked and the geometrical density compared with that obtained by an immersion method using dibromo ethane. Acceptable pellets were then inserted in the stainless steel clad tube, whose top end plug was already welded and radiographed. The tube was inserted into the glove box; a holding spring was pushed in, followed by a molybdenum plug,  $\text{PuO}_2$  pellets and another molybdenum reflector. The loaded clad tube was held in a special welding chamber in a glove box and the chamber was evacuated and filled with helium. After inserting the lower end plug, welding was carried

out remotely by argon arc welding. The fuel pins were then decontaminated, subjected to radiography and helium leak test. Approved fuel pins were loaded in birdcages and transferred to the Plutonium Store or reactor site.

#### Design Safety Features

The long biological half-life and the high energy of the emitted alpha particles together with selective localisation in bone or lung makes plutonium one of the most toxic materials when deposited inside the body. With the maximum permissible lung burden for insoluble plutonium (e.g.  $\text{PuO}_2$ ) being as low as  $0.016 \mu\text{Ci}$  ( $\approx 0.26 \mu\text{g}$ ) the glove box design called for stringent built-in safety features with regard to containment capability. Since a mass as low as 500 g of Pu could lead to a nuclear excursion under unfavourable conditions, criticality safety was to be considered through mass control, safe separation of Pu units and safe design of birdcages for storage and transport, with administrative control at each stage of handling.

#### Safety Features in Laboratory Design

The fuel fabrication facility is housed mainly in two high active area halls with entry from an active corridor. The corridor has a decontamination room at one end, and the other side, opens out to a personnel corridor, leading to a change room. A plutonium store room is located in the personnel corridor. Equipments for the various metallurgical operations are housed in glove boxes, located in the high active halls. Services of a high order of integrity, required for a class A laboratory have been provided. Laboratory area and glove boxes are provided with separate air supply and exhaust system, the equipments of which, located in a filter house, discharge the effluents through a 76 m stack, after filtration through high efficiency particulate filters. Ventilation for the active halls and corridors have been designed to give respectively 10 and 7 air changes per hour and pressure differentials have been maintained between the areas to enable air-flow from low active areas to high active ones.

#### Glove Boxes and their Safety Features

Many design safety features have been incorporated in the fabrication of glove boxes from the point of view of containment. Glove box frame and floor are made of s.s 304 for ease of decontamination. Filter boxes of aluminium are conveniently located so as to enable replacement of the inlet and outlet filters by a single hand operation through the upper port. Absolute filters of MSA Honey-comb type with an efficiency of 99.9% for  $0.3 \mu\text{m}$  particles are used. Transfer and posting-in operations are carried out through air locks with double doors and bagging-in ports. Normal atmosphere was found adequate for  $\text{PuO}_2$  fuel work; however, for Pu metal handling, the boxes could be turned to an argon system, provided with a purifier and a recirculation unit. Operations were carried out in glove boxes under -1 in.WG.

Air enters through an isolation valve, a ball valve, rotameter, a regulator and an inlet filter; and vents via an exit filter, a ball valve and a three-way solenoid valve. A mechanical pressure controller and a bellows adjust minor pressure variations but in case of accidental overpressurisation or any rupture of glove or failure of recirculation system, the three-way solenoid valve initiates emergency control by opening the box directly to the -10 in. WG. main glove-box exhaust line. The inlet regulator closes and a pressure differential switch flashes an alarm in the form of a red light on glove-box board, warning the operating staff of an emergency situation. Neoprene gloves, 0.8 mm thick, were considered adequate against the soft radiation emitted by plutonium. Apart from installed  $\text{CO}_2$  extinguishers in the halls, eutectic salt mixture in sealed PVC bag was kept handy in glove-boxes to smother any fire. The furnace coolant water is normally on main water supply. An emergency water tank has been provided to take care of failure of main water supply or loss of pressure. Filtered water from a pool was also connected to the line as an alternative for sustained supply.

#### Pre-commissioning Tests

The primary responsibility of the health physics staff at the time of commissioning of the facility was to check the adequacy of the protective features and assess operational safety.

Glove Box Containment Evaluation. The gloves and glove boxes were checked for leakage before commissioning for Pu handling. The box leakage rates were found to be less than 0.05% box volume/hour, as is prescribed for inert atmosphere boxes.

Glove Box Filter Efficiency Tests. Filter efficiency checks were carried out with uranine aerosols. Filters were approved for use only when they conformed to 99.9% efficiency for 0.3  $\mu\text{m}$  particles.

Effluent Drains Checks. High and low active drains were checked with inactive cold runs using rhodamin dye to ensure proper pump connections and valve operations.

Breathing Air Line Checks. Compressor air was checked for presence of oil mist, moisture and CO to ensure that their levels were below the tolerance limits. The minimum requirement of 3 cft/m at the breathing points was checked.

#### Operational Safety and Hazards Control

Mass of Pu, handled was initially limited to 75 g per batch to acquire experience and later, the batch size was progressively increased to 500 g Pu, after reviewing the safety aspects. In all about 85 sintering runs and about 200 in-box welding operations, covering fuel pins and start-up source pins were carried out.

Constant health physics surveillance was provided for the operations. Access control to the fuel laboratory was enforced through change rooms. In potentially active areas like filter room, decontamination room, entry was effected under health physics supervision or after obtaining special work permits. Use of protective clothing consisting of overalls, overshoes, head caps and surgical gloves for handling pellets and pins was recommended. Further, TLD's on forehead and chest, normal beta-gamma and fast neutron film badges, criticality badge and pocket dosimeters were worn while at work with  $\text{PuO}_2$ . Air line respirators connected to 15 lbf/in<sup>2</sup> airline via quick connection couplings were kept readily available for emergency use.

Equipments for sintering and weighing operations were located in a train of interconnected glove boxes to preclude the necessity of intermittent bagging out operations and consequent external exposure. Different phases of work were segregated to avoid contamination spread. The welding operations, metallography work and source pin fabrication were grouped separately in another train of glove boxes to facilitate flexibility and control of radiation exposure and contamination.

As the quantity of  $\text{PuO}_2$  handled was progressively increased, extensive radiation survey was conducted to control personnel exposure using conventional radiation monitoring instruments. The area and air monitors were strategically located in the laboratory. Provision was also made to monitor the effluent streams. The monitors along with a remote read-out on a Central Health Console give alarm at pre-set limits for initiating corrective action.

#### External Hazards and Control

$\text{PuO}_2$  powder was obtained from reprocessing nat.U fuel from Cirus reactor.

Radiation survey data of the first seven sintering runs, with 75 g Pu per batch, indicated high beta-gamma dose rates from pellets; the pellets showed a gamma dose rate of 300-450 mR/h and the beta dose rate was 1-2 R/h. The glove box panels registered a gamma dose rate of 50-75 mR/h. This also

indicated a ratio of about 6 between contact and chest level dose rates. Analysis of an aliquot sample of  $\text{PuO}_2$  by health physics staff indicated mainly  $^{95}\text{Zr}$ - $^{95}\text{Nb}$  activity and a total activity of about 5  $\mu\text{Ci/g}$  of  $\text{PuO}_2$ . Subsequently  $\text{PuO}_2$  was therefore obtained from spent fuel rods with lesser fission product content. As a result, the gamma dose rates on the sintering glove box panel came down to 12-30 mR/h even with increased quantities of 200-550 g  $\text{PuO}_2$ .

The fabrication of fuel pins as well as start-up source pins did not call for special shielding to boxes. As a measure of radiation safety, fuel pin welding and Pu-Be source pin fabrication jobs were carried out by rotation of staff. This was necessary as the beta-gamma dose rate from a full fuel pin at 1 cm was nearly 40-50 mR/h while the neutron dose rate was 130-140 mrem/h. Fuel pins were checked individually for loose contamination by an alpha probe inside the box and also with swipe counting. After radiography and helium leak test, the weld-zones were checked for fixed contamination. Before machining of the welds for most pins, the counts varied in the range of 100-1000 dpm/cm<sup>2</sup>, maximum being 64000 dpm/cm<sup>2</sup> while after machining and polishing, the levels for most pins came down to 200-400 dpm/cm<sup>2</sup>.

For start-up source pins of 0.9 Ci strength, the method of fabrication was to mix nearly 16 g of Pu as  $\text{PuO}_2$  with nearly equal quantity of Be followed by pressing and sintering in a high vacuum induction furnace. The neutron exposures incurred during fabrication of 2 source pins were of the order of 50 mrem on chest and 400 mrem on wrist per man. The contact gamma dose rate of the source pin was nearly 150 mR/h while the dose rates at 30 cm from the pin were 16 mR/h due to gammas and 25 mrem/h due to neutrons.

Cumulative dose(beta-gamma-neutron) received by a few members of the staff, directly involved in the fabrication work during the campaign period 18.6.70 to 21.3.72 are indicated below:

Person	Exposure(mrem)	Person	Exposure(mrem)	Person	Exposure(mrem)
A	177	F	658	L	763
B	482	G	867	M	820
C	199	H	541	N	379
D	827	I	1069	O	473
E	166	J	1540	P	569

From the estimated ratio of contact to chest level dose rates, maximum extremity exposure could be of the order of 9 R.

#### Air Contamination Control

Each glove box premise has a suction port with an air sampling head connected to a central air sampling pump. Filter paper samples obtained with this system as well as with annular impactors when analysed for long lived activity, did not show any air contamination in the laboratory. In addition a Pu-in-air monitor located in the laboratory detects air borne Pu by alpha spectrometry. The detector is of a silicon surface barrier type. The unit is pre-set to sound an alarm at 8 MPC hours in presence of natural radioactivity whose spill-over in 4.1 - 5.1 MeV plutonium channels is estimated to be less than 10% of the total.

During glove changing operations, respirator area was maintained. Maintenance work was carried out once on an induction furnace for which frog suit and air line respirator were prescribed. Only one instance of air contamination due to a small tear on glove arose. Due to immediate corrective action, no personnel exposure occurred.

Bioassay and whole body counting of the operating staff showed that there was no internal exposure. About 34 members of staff were monitored, after completion of the programme for Pu deposition in lung with a thin NaI(Tl) crystal with a Be window. The count rates obtained were of background levels after repeat monitoring.

### Environmental Contamination Control

Air-borne effluents were discharged after monitoring downstream through a stack. The glove box and laboratory exhaust had negligibly small long lived activity. The liquid wastes, both high active ( $>10^{-4}$   $\mu\text{Ci}/\text{cm}^3$ ) and low active ( $<10^{-4}$   $\mu\text{Ci}/\text{cm}^3$ ) ones were collected in separate tanks and sent for disposal. Maximum levels of alpha and beta-gamma activity of the liquid effluents discharged from the fuel fabrication facility for processing were nearly  $1.2 \times 10^{-6}$   $\mu\text{Ci}/\text{cm}^3$  and  $5.6 \times 10^{-6}$   $\mu\text{Ci}/\text{cm}^3$  respectively; the net activity figures over a year for alpha and beta-gamma were nearly 2 mCi and 10.5 mCi respectively. Solid wastes, suspected to contain Pu were segregated in standard containers marked 'active' while non suspect wastes were handed over to the waste management facility. Low active solid wastes generated were to the extent of 15 - 20 packets, each of 2 c.ft. volume and the packets had a maximum surface dose rate of 1 mR/h and these were also sent for disposal.

### Criticality Safety

Preliminary clearance was limited to 250 g Pu in the sintering furnace glove box, taking into account the possibility of the coolant line rupture. Presence of two batches, each of 250 g Pu, was permitted in either of the high active halls at any time. Later, on the basis of operating experience, the quantity of Pu for sintering was progressively increased to 500 g Pu and the same handling limit, was enforced for the welding box too. Administrative control ensured that water or other homogeneous materials were not brought inside the box; however small quantities, required for specific operations, were permitted after special clearance.

Birdcages have been fabricated to store and transport the fuel pins. The birdcage consists of a mild steel slotted angle frame work with an aluminium container, rigidly fixed at its centre. Inside the Al container is a square cluster of nine aluminium tubes welded together at the top and bottom to form a bundle. Each of the tubes accommodates one fuel pin in a PVC bag; thus nine pins, amounting to 1.26 kg  $\text{PuO}_2$  can be stored in the birdcage. The central Al container is provided with a tight fitting cap with a neoprene gasket to render it leak tight. A prototype birdcage was subjected to drop and water leakage tests and was approved for use.

The birdcages carrying the complete core charge of 'PUERNIMA' reactor ( $\approx 22$  kg Pu) were stored in the Pu store room in a plane array. The birdcages of size 60 x 40 x 40  $\text{cm}^3$  have been designed to maintain between the central Al containers a minimum surface-to-surface separation of 30 cm, to isolate the containers in the event of flooding. Effective neutron multiplication factor of a birdcage with 9 fuel pins, under flooding conditions (including internal flooding) has been estimated as about 0.67. Thus the nuclear safety of the individual birdcage as well as the array was ensured in the event of flooding.

### Criticality Monitor

Criticality monitors are located in the two high active halls and Pu store room. The sensing device consists of an ion chamber, connected to a period amplifier. The amplifier gives an indication of the rate of rise of the gamma field during an excursion. The criteria for alarm setting of the system were fixed as follows:

- (i) the system shall sound a positive alarm if a criticality burst of  $10^{15}$  fissions occurs at a distance of 30 ft. from the detector and delivers prompt gamma dose in 100 milliseconds,
- (ii) the system shall not give an alarm as a result of handling 10 Ci  $^{60}\text{Co}$  source at a distance of about 10 ft from the chamber.

The above criteria will be satisfied if a change in radiation level by 6 decades (i.e. 10 mR/h background to  $10^4$  R/h) triggers the alarm.

In order to study the response of the monitor, a criticality event was simulated by shooting a  $^{60}\text{Co}$  capsule of 1 Ci strength past the ion chamber. The source was ejected with compressed air over a distance of  $8\frac{1}{2}$  ft in 0.25 sec, giving a change of field from 10 mR/h to  $10^4$  R/h. The alarm limit was set at 50% of the maximum deflection obtained during calibration.

#### Estimation of Pu in Waste and Fuel Pins/Pellets

As a measure of inventory control, instruments were developed to estimate Pu content in solid wastes,  $\text{PuO}_2$  pellets and finished fuel pins.

Assessment of Pu in solid waste was carried out by counting low energy X-rays from Pu in the channels, corresponding to 11.0 to 21.5 KeV, with a 1 mm thick and 25 mm diam. NaI(Tl) crystal, having a Be window. Measurements with 1  $\mu\text{Ci}$  Pu source, in a 125 mm diam and 175 mm high standard waste container, gave twelve times the background counts in four minutes and this indicated a feasibility of estimating  $\mu\text{g}$  levels of Pu in solid wastes.

A  $\text{BF}_3$  filled annular counter was developed to measure the neutron emission from  $\text{PuO}_2$  pellets and fuel pins. Since Pu has been obtained from reprocessing of low burn-up fuel, it was possible to estimate  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  content in the pellets and pins knowing the neutron yields from spontaneous fission and (alpha, neutron) reactions in  $\text{PuO}_2$ . An approximate assessment of the neutron dose rate could also be made from the measurements.

#### Conclusion

Safe operation of the facility has been amply demonstrated by low personnel exposure and absence of unsafe incidents and this has given an incentive to fast reactor fuel development programme.

#### Acknowledgement

Thanks are due to Dr. A.K. Ganguly, Head, Health Physics Division for his directions on safety aspects during the various stages of the fuel fabrication programme.



CELLULOSE NITRATE PLASTIC FILM - ITS PREPARATION  
AND APPLICATIONS IN HEALTH PHYSICS

A.M.Bhagwat and S.D.Soman  
Health Physics Division  
Bhabha Atomic Research Centre  
Bombay-400085, India

Abstract

The paper deals with the standardisation of Cellulose nitrate(C.N.) films (thickness  $\approx 3.0$  mg/cm<sup>2</sup>) for the detection of alpha particles from a <sup>239</sup>Pu source and some of its applications. The etchant used was 6 N NaOH. The optimum etching time for the development of tracks was found to be 4.5 hours at room temperature (24-25°C). The track formation efficiency under these conditions works out to 17.9%.

Results of some of our studies are included here. It is now possible to detect low alpha activities of the order of 10<sup>-3</sup> dpm, thereby making many types of extremely low level alpha measurements possible.

Introduction

With the advent of solid state track detectors in 1958<sup>1</sup> it has been possible to detect heavy charged particulate radiation such as fission fragments, alpha particles, deuterons or protons in the presence of other radiation such as beta-gamma, neutrons etc.

Of the various plastic films used so far in such measurements, C.N. plastic film has been reported to be the most sensitive. This paper describes the preparation and standardisation of the C.N. plastic films in our laboratory and some of its applications in health physics work through the registration of alpha particle tracks.

Preparation of the C.N. plastic film

Chemicals used

- a) Cellulose nitrate (commercial grade) (N<sub>2</sub>=12.1 wt%)-17.0 g
- b) ethyl acetate (A.R.) - 61.9 g
- c) Isopyropyl alcohol (A.R.) - 5.1 g
- d) Butyl alcohol (A.R.) - 4.0 g
- e) Cellosolve acetate (commercial grade) - 8.0 g
- f) Dioctyl phthalate (DOP) (commercial grade) - 4.0 g

Using these chemicals in the proportions shown above the plastic films were prepared in the following manner by Benton's technique<sup>2</sup>:

Cellulose nitrate was first dissolved in ethyl acetate to which the other three solvents were added in the order shown above. To the solution, 4.0 g of DOP was added as a plasticiser. The mixture was then permitted to age for four days to allow it to attain chemical equilibrium before films could be prepared.

For preparation of the films, the above solution was used as such; and for obtaining thinner films - after dilution with different volumes of ethyl acetate. These solutions were poured on glass plates of size 10" x 4" which were kept in a slanting position with a slope of about 1 in 7. The glass plates were left undisturbed overnight.

The next day, the films were gradually peeled off from the glass plates by inserting water drops between the films and the glass plate. This was followed by annealing of the films at a temperature of 100°C for about 16 hours after which they were ready for experimental use.

The above films were in thickness range of 2 to 6 mg/cm<sup>2</sup>. The chemical composition of the films in wt. % was found to be: C = 37.3, H=4.8, N=9.45 and O = 47.95 (by difference).

#### Track formation by chemical etching

The final shape of the tracks formed by chemical etching is influenced by numerous factors such as the direction of entrance of the particle into the film, the physical structure of the substance, choice of the etchant and its concentration, temperature of etching, etching time etc.<sup>3,4</sup>. Under ideal conditions, i.e. when the track recording material is homogeneous and the diffusion effects are absent, the etched tracks are conical in shape with sharp terminal ends<sup>2</sup>.

The alpha particle tracks registered in our C.N. films have been found to be of two types - sharp conical and pit type (Fig.1). The sharp conical tracks indicate that ideal conditions of film preparation are not beyond reach. The etchant in our case was 6 N NaOH solution at room temperature.

#### Standardisation of our C.N.film

<sup>239</sup>Pu is one of the hazardous isotopes which is being handled in BARC laboratories. It was therefore decided to carry out the standardisation of these films with respect to this isotope. The films were exposed to alpha particles from <sup>239</sup>Pu sources on stainless steel planchets and the following parameters were studied:-

#### Etching temperature

It was found that the best track shapes could be seen when the etching was carried out at room temperature (24-25°C). At higher temperatures the track shapes tended to be pit type.

#### Useful thickness of the film

This was found to be about 3 mg/cm<sup>2</sup>. At smaller thickness the film becomes difficult to handle after etching and at larger thickness the etching times are very long (8 hours or more) at room temperature.

#### Optimum etching time

This parameter was determined for the films of thickness  $\approx$  3 mg/cm<sup>2</sup> (exact thickness of the film = 2.9 mg/cm<sup>2</sup>) and was found to be 4.5 hours at room temperature. Beyond this etching time the number of tracks was found to diminish.

#### Track formation efficiency

Under the optimum conditions as given above it was found that the films ( $\approx$  3 mg/cm<sup>2</sup>) record 17.9 tracks per 100 disintegrations in the sample, in close contact with the source, the etching time being 4.5 hours at room temperature.

### Detection of low levels of alpha contamination

Radiometallurgy section, handling Kg amounts of  $\text{PuO}_2$ , does not show any detectable floor contamination by normal monitoring techniques. To check the presence of extremely low levels of contamination, if any, five spots were chosen on the floor for the exposure of the C.N. film. These spots had not shown any loose or fixed contamination by scintillation probe monitoring. They were then covered with C.N. films followed by PVC covering and left undisturbed for about 11 days.

After exposure the films were washed with soap and water to remove the attached loose dust, if any. The films were then etched in 6 N NaOH solution for 4.5 hours at room temperature. After etching, all the films were washed, dried and examined under the microscope. A control film was also processed in the same fashion.

The experimental films showed the presence of many groups of tracks—each group representing one active speck. The minimum and maximum number of tracks in any group was found to be 6 and 238 respectively.

### Discussion

If we assume that the track formation efficiency for alpha particles here is the same as that calculated in the standardisation experiments we can calculate the activity of each speck responsible for a group of tracks. It is seen that activity of any individual speck encountered in these experiments lies in the range from  $2.14 \times 10^{-3}$  dpm to  $3.5 \times 10^{-2}$  dpm. The table below shows the levels of contamination as detected by each film.

Place of exposure for the film	Area of the film	Total No. of tracks on the film	Corresponding activity in		
			dpm	dpm/cm <sup>2</sup>	*M.P.L. of surface contamination
Below the bagging port of welding glove box	7.28	637	0.23	0.03	$1.3 \times 10^{-2}$
Below the transfer port of balance glove box	5.28	350	0.12	0.02	$8.1 \times 10^{-3}$
Below the bagging port of induction furnace glove box	6.44	1312	0.47	0.07	$2.9 \times 10^{-2}$
In front of decontamination fumehood	6.54	210	0.06	0.01	$4.7 \times 10^{-3}$
in front of decontamination fumehood	9.28	209	0.07	0.01	$3.2 \times 10^{-3}$

\* 1 M.P.L. of alpha surface contamination at BARC =  $2.5 \text{ dpm/cm}^2$

This study shows that the new technique goes a long way in lowering the detection limit for alpha contaminations. It can be adopted for routine use also as the technique is simple.

### Detection of low levels of alpha activity in bioassay samples

Encouraged by the success of C.N. films in the detection of low levels of alpha contamination, an attempt was made to study their utility for bioassay samples as well. The results of our studies with plancheted plutonium alpha activity of less than 1 dpm, from urine samples, are described below.

Two such stainless steel planchets (diameter = 2.0") of activity 0.16 and 0.393 dpm were kept in contact with C.N.film for about 11 and 7 days, thus exposing the films to 2520 and 3920 disintegrations respectively as per the counting data. Exposures were carried out in a vacuum desiccator to eliminate the interference due to natural radioactivity in air.

After exposure, the films were washed with soap and water to remove the attached loose precipitate and etched as described earlier. The number of alpha particle tracks recorded on each film were counted during scanning and found to be 3841 and 5667 respectively.

### Discussion

The number of tracks recorded on the C.N.film are higher by factors of 1.52 and 1.44 than the total number of disintegrations in the samples as calculated from their respective dpm values. The discrepancy in the two results can partly be assigned to following reasons:

- i) High statistical errors in counting of the bioassay samples which were of the order of  $\pm 39.6\%$  and  $\pm 20\%$  in our two samples respectively.
- ii) The ZnS scintillation counter is being standardised by sources having a diameter of about 20 mm or less while the area covered by the precipitate on the bioassay planchet samples may be anywhere from 40 to 50 mm in diameter. Our preliminary investigations have shown that correction for this area would enhance the dpm value of the sample by about 25%.
- iii) The precipitate on the planchets leads to degradation of energy of alpha particles. The exact degree of degradation cannot be ascertained for such low activity samples. But it has been observed that the track registration efficiency increases as the energy of the alpha particles decreases. And for ZnS scintillation counter the efficiency of counting falls off as the energy of alpha particles decreases. These factors will therefore further help to bridge the gap.
- iv) The amount of precipitate was found to vary on the two planchets under investigation.

All these factors will help in partially explaining the discrepancy. But a detailed investigation is separately necessary to explain it completely.

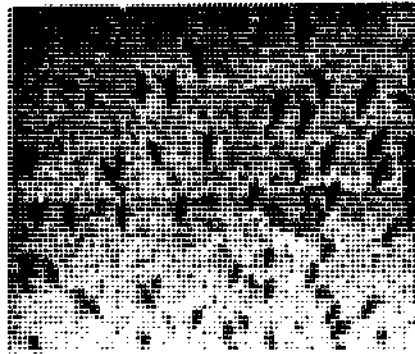
### Other applications

The C.N.film prepared in our laboratory have also been used in the following studies:

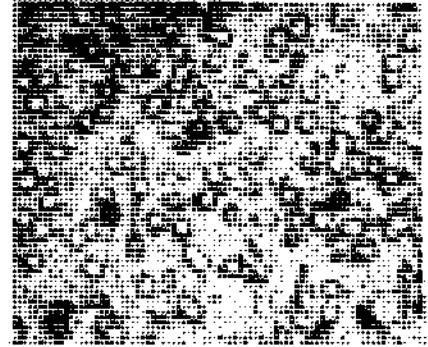
- 1) Autoradiography of electrodeposited planchet type sources. Of the few sources studied, some have revealed the presence of many clusters suggesting extreme nonuniformity of deposition (Fig.2). A detailed investigation into the causes leading to such non-uniform deposition is being carried out.
- 2) Autoradiography of single drop planchet sources. It has been observed that the activity along the edges (width of the edge  $\approx 500 \mu\text{m}$ ) is about 60% of the total for source diameters of 6 to 7 mm.
- 3) Autoradiography of  $\text{UO}_2$ - $\text{PuO}_2$  pellets (weight proportion 97.4:2.6) to study the uniformity of distribution of plutonium in Uranium (Fig.3). The information obtained should be of great help to metallurgists in evaluating
  - i) degree of non-uniformity in mixing
  - ii) sizes of individual particles of plutonium
  - iii) formation of aggregates of plutonium during mixing, if any.
- 4) Autoradiography of Uraninite mineral to locate the active grains of Uranium. This film makes the location of active grains very easy when compared with the photographic film because it is not sensitive to beta-gamma radiations

### References

1. D.A.Young, Nature, 182(1958), 375.
2. E.V.Benton, USNEDL-TR-68-14
3. D.S.Srivastava, Aligarh Muslim University, Ph.D.Thesis, 1971
4. R.L.Fleischer et al, Ann.Review of Nucl.Sc. 15(1965), 1.



(a)



(b)

Fig. 1. Alpha particle tracks (a) conical, (b) pit type.

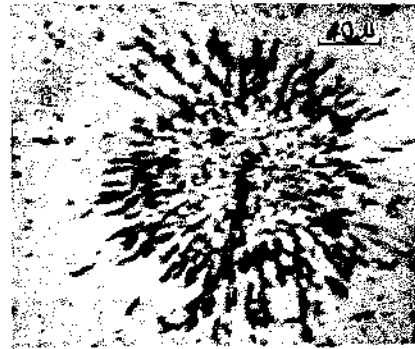
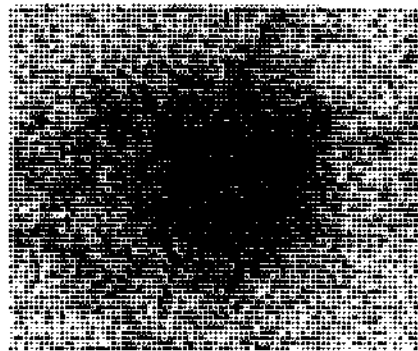
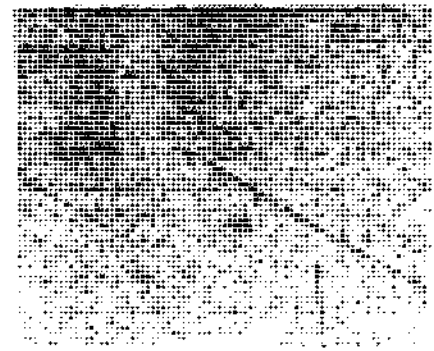


Fig. 2. A cluster from electrodeposited  $^{239}\text{Pu}$  source.



(a)



(b)

Fig. 3. Distribution of  $\text{PuO}_2$  in  $\text{UO}_2$  (a) cluster showing non-uniformity, (b) an area showing uniformity.

## RETENTION OF IODINE ON DUCT SURFACES

W. E. Grummitt

Biology and Health Physics Division, Chalk River Nuclear Labs., AECL  
Chalk River, Ontario, Canada

The behaviour of fission-product iodine in a reactor exhaust duct and filtering system is of continuing interest. At CRNL parts of the research reactors are cooled with air which is released from a stack located 770 m beyond the absolute filters. Gases are transferred to the stack through a large duct. Occasionally, small amounts of iodine are released to the system from an experimental reactor loop. These nuclides have been used in the present study to measure the hold-up of iodine in the system.

### Introduction

The behaviour of airborne radioactivity is of interest in evaluating the hazard from continuous or accidental discharge of fission products from a nuclear reactor. One of the factors affecting the amount released to the environment is the fractional deposition on containment surfaces. This report deals with the mechanism of deposition on specific surfaces and with re-emission from them.

The research reactors at CRNL are operated as test facilities in which the fuel material, cladding and the cooling conditions can be altered. A test loop, inserted in a single NRX fuel channel, is isolated from the reactor cooling system. In some cases the loop circuit may be vented to the main air duct; rare gases and halogens from a failed fuel element can then be discharged from the stack. The remaining fuel rods in NRX are water cooled and do not vent to the atmosphere.

Iodine-131 is particularly important in assessing the hazard from nuclear reactors. It has a fairly long half-life and escapes readily from ruptured fuel and the element is essential to man. The remaining fission product iodines have shorter half-lives and contribute proportionally less to the integrated dose, but they are useful for investigating the behaviour of the element in a dynamic system.

Various chemical forms of gaseous  $^{131}\text{I}$  have been recognized in addition to the elemental form<sup>1, 2</sup>. Perhaps the most troublesome of these is methyl iodide<sup>3, 4</sup>, but hypoiodous acid, HOI, is released under moist conditions at high pH<sup>5, 6</sup>. Both of these are difficult to remove from the gas phase.

Elemental iodine should deposit on duct surfaces but the more penetrating forms, which generally comprise about half the total, are unreactive and are not retained on surfaces to an appreciable extent. Assuming that the retention of  $\text{I}_2$  is reversible and that the mean life on the surface is the order of days, there should be an observable difference between the 8.065 d  $^{131}\text{I}$  and the other isotopes which are retained on the surface. At the stack one should see the sum of the following components; a) All isotopes present in the

penetrating form; b)  $^{131}\text{I}$  which has been deposited on the surface and subsequently released; c)  $^{133}\text{I}$  and possibly  $^{135}\text{I}$  in the elemental form which has undergone partial or substantial decay before escaping from the surface. Iodine-134 (52.3 m) should only be transported to the stack in the penetrating, i. e. non-elemental form.

### Methods

A flow diagram of the ventilation system is shown in Figure 1. The absolute filters are located at a short distance from the NRX reactor. The duct is constructed of mild steel 1.2 m diameter by 770 m long and is painted on the inside with one coat of Zincilate 410. This provides a sacrificial coating of zinc which protects the underlying metal.

Samples were collected by drawing a measured amount (0.1 to 2 m<sup>3</sup>) of air through a canister. Collection times varied from five minutes to two hours depending on the nature of the experiment. In experiments designed to measure the chemical form of iodine the sample was collected with a freshly prepared May pack<sup>8</sup>. In all other cases a polyethylene capsule, 12 mm x 50 mm, filled with activated charcoal, was used to collect the sample. The order and efficiency of components in the May pack is given in Table 1. Uptake of hypiodous acid by copper screens and charcoal was determined in this study. McCormack<sup>7</sup> reports that more than 70% of the HOI is retained by the carbon paper. In the present case less than 11% is found on the other components of the May pack, so more than 89% must be retained by the paper.

Table 1

		I <sub>2</sub>	Particulate	HOI	CH <sub>3</sub> I	Ref.
4 Copper screens	100 mesh	97%	nil	<3%	nil	8, 9
1 Glass fibre filter	Type GFA	3%	99%	<2%	nil	9
1 Gelman carbon paper	Type ACG/B	nil	nil	>70%	1%	7
Barnebey-Cheney #513 Activated charcoal	60g	nil	nil	<6%	99%	7

Air samples were taken at the four locations shown in Figure 1. In order to minimize loss of iodine prior to counting, the individual components were sealed in polyethylene envelopes. Sample 1 was collected through a metal tube inserted six inches into the duct so is probably not representative of the mean concentration at that point.

All the nuclides were determined simultaneously by gamma spectrometry. A 125 cc Ge(Li) detector with 2.3 keV resolution was used for most of the experiments. This was coupled to a 4096 channel analyser. Details of the method of computation were given in an earlier report<sup>10</sup>. With this system the isotopes of iodine, bromine, xenon, rubidium and cesium could be resolved easily in a single count. Repeated measurements were made to verify half-life and to check for interfering lines such as the 529.5 keV gamma transition of 2.4 h  $^{83}\text{Br}$ . In this case the maximum error in the determination of the 20.8 h  $^{133}\text{I}$  was 3% in the first hour. The energies of the lines which were selected for quantitative determinations are given in Table 2 together with the preferred gamma branching ratios.

### Results

#### Chemical Composition of Iodine

A typical result from the analyses of the May packs is given in Table 3.

TABLE 2  
Selected Gamma Ray Energies

Mass	Half - Life	Preferred $\gamma$ Line	$\nu$ per Disintegration
131	8.065 d	364.49 keV	0.790
132	2.284 h	667.8	0.999
133	20.8 h	529.9	1.00
134	52.3 m	847.04	1.00
135	6.68 h	1260.45	1.00
82	35.34 h	554.23 776.45	0.73 0.83

These samples were collected at location 2, immediately after the absolute filters. Corrections have been applied for incomplete uptake of  $I_2$  on the copper screens (97%) and for retention by the particulate filter (see Table 1). The species held by the charcoal paper is largely hypoiodous acid<sup>7</sup>, though high molecular weight alkyl iodides may be collected with this fraction.

The five iodine isotopes show a remarkably similar distribution on the May pack. This is in contrast to Keller et al<sup>11</sup> who recently observed large isotopic differences under conditions of high humidity. However, in both instances the fraction found in the elemental form was lowest for  $^{131}I$ .

TABLE 3  
Chemical Composition of Iodines and Bromine

Mass No.	131	132	133	134	135	82
Elemental	26.1%	29.7%	31.1%	31.2%	29.7%	17.7%
Particulate	1.8	2.4	2.1	2.3	2.3	2.0
HOI/HOBr	64.6	60.3	63.1	61.0	63.4	60.1
$CH_3I/CH_3Br^*$	7.5	7.6	3.8	5.4	4.2	21.2

\*This represents an upper limit; species which are not completely held by earlier components in the pack will be collected and counted as methyl iodide or bromide. Relative humidity = 35%.

#### Retention of Iodine-131

The concentration of  $^{131}I$  at various points in the duct was measured in three separate experiments. The purpose of this series was to establish the extent to which this nuclide was retained on surfaces under conditions of near constant release. The first three runs in Table 4 followed long periods with the reactor operating at constant power.

Iodine-131, corrected for air flow at the sampling point, shows little variation along the length of the duct with the reactor operating (samples 2, 3 and 4); it follows that there is negligible removal of  $^{131}I$  in the duct system. The results also suggest that  $^{131}I$  is not removed by the absolute filters. The apparent increase from the first to second sampling point is caused by poor mixing and non-representative sampling at the first location.

There tends to be a release of extra fission products at shutdown which is still evident 15 hours later (Table 4). Some of the excess  $^{131}I$  is retained on the duct surface, thus at  $t_0 + 15$  hours the concentration is lower at the stack (location 4) than at the filters (location 2).



TABLE 4

## Iodine-131 Concentrations at Various Points in the Duct

Experiment	Sampling Point				Time from Shutdown
	1	2	3	4	
1	4.35	6.85	6.64	6.59 nCi/m <sup>3</sup>	*
2	3.88	6.48	6.15	6.31	*
3	3.78	5.39	-	5.46	*
3a	5.54	8.61	-	7.28	15 h
3b	3.19	5.04	-	5.18	20 h
3c	2.03	2.45	-	2.85	40 h

\*Reactor Operating, Flow = 52,670 m<sup>3</sup>/h.

TABLE 5

## Iodine-133/Iodine-131 Activity Ratios

Experiment	Sampling Point				Time from Shutdown
	1	2	3	4	
1	5.2	5.0	4.1	4.2	*
2	5.4	5.2	4.6	4.5	*
3	10.2	9.3	-	5.0	*
3a	1.8	1.1	-	1.7	15 h
3b	1.3	1.6	-	2.0	20 h
3c	1.0	1.7	-	2.3	40 h
3d	0.8	2.6	-	2.4	60 h

\*Reactor Operating. Ratios are corrected for radioactive decay from time of shutdown as applicable.

Other Isotopes of Iodine

Table 5 shows the retention of <sup>133</sup>I, one of the four isotopes of iodine with half-lives shorter than eight days. Ratios to <sup>131</sup>I permit use of results from the first sampling point. In Table 6 the data for <sup>133</sup>I and other nuclides are normalized to facilitate intercomparison.

Under steady state conditions at constant reactor power there is moderate retention of <sup>133</sup>I on the duct. The results in Table 6 show little dependence on half-life though <sup>133</sup>I is slightly higher than <sup>134</sup>I and <sup>135</sup>I at the stack end of the duct. Tellurium-132 (78 h) is found beyond the absolute filters and this causes <sup>132</sup>I to be high and variable.

Release Following Reactor Shutdown

The foregoing were steady state experiments; the reactor loop had operated at constant flux for periods of up to three weeks. Following shutdown, it was observed that <sup>131</sup>I levels remained relatively constant (see Table 4), but the 6.68 h <sup>135</sup>I and 20.8 h <sup>133</sup>I dropped by an order of magnitude (Table 5). This rapid change provided an opportunity for observing desorption from the duct surface. Results are given in Table 7.

Bromine-82

Bromine-82 is a shielded nuclide which has a very low fission yield so would not normally be observable with the other halogens. In NRX it is probably produced by an n,  $\nu$  or n, p reaction on bromine or krypton.

TABLE 6

Concentrations of Iodine and Bromine Isotopes Relative to  $^{131}\text{I}$  (Normalized)

Isotope	Half-Life	Exp. 1 Sampling Point				Exp. 3 Sampling Point			
		1	2	3	4	1	2	3	4
133	20.8 h	1	0.95	0.78	0.80	1	0.91	-	0.58
135	6.68 h	1	0.99	0.75	0.74	1	0.92	-	0.52
132	2.28 h	1	1.1	1.0	1.0	1	1.6	-	2.0
134	0.87 h	1	0.97	0.74	0.75	1	0.93	-	0.50
82	35.34 h	1	0.67	0.69	0.68	1	0.53	-	0.51

TABLE 7

Duct Concentrations After Shutdown Relative to  $^{131}\text{I}$  (Normalized)

Isotope	Experiment	Sampling Point			*Reactor Operating
		1	2	4	Time from Shutdown
133	3	1	0.91	0.58	*
	3a	1	0.62	0.90	15 h
	3b	1	1.2	1.4	20 h
	3c	1	1.7	2.3	40 h
	3d	1	2.6	2.4	60 h

### Discussion

Morris and Nicholls<sup>1,2</sup> measured the deposition velocity on copper and galvanized steel and found that there was rapid uptake with a much slower rate of removal. In the present case the metal surface of the duct has a coating of granular zinc onto which the iodine deposits. This layer of  $^{131}\text{I}$  can be displaced with inactive iodine<sup>3</sup>, showing that the exchange is reversible.

The data indicate that only the elemental form is exchanging. If R is the fraction retained on the duct then T, the fraction which is not retained is given by  $T = 1 - R$ . Values of T can be obtained from  $^{134}\text{I}$  and  $^{135}\text{I}$  concentrations at location 4 (Table 6). The observed retention, R, is 0.26 for the first experiment and 0.49 for the third. May pack results show that 30% and 50% of the iodine present is in the form of  $\text{I}_2$  at these times.

Most of the molecules on the surface of the duct at a given time will be  $^{131}\text{I}$  because this isotope has the longest half-life. At an observed activity ratio of nine to one the numbers of atoms of mass 131 and 133 are equal in the gas phase. Normally the 52 min  $^{134}\text{I}$  is a factor of ten less abundant.

The mean life of a molecule on the surface before escape by exchange can be estimated from the data in Table 6. Most, though not all, of the 20.8 h  $^{133}\text{I}$  decays while held on the surface. Let us assume that the fraction exchanging is the same for all isotopes and that in the case of 52 min  $^{134}\text{I}$  this fraction decays completely on the surface. Now if  $R_0$  is the fraction retained on the duct which undergoes complete decay and  $R_t$  is the fraction undergoing partial decay before escape, the fraction of  $^{133}\text{I}$  which remains is

$$\frac{N_t \lambda}{N_0 \lambda} = \frac{R_0 - R_t}{R_0} \quad \text{where } \lambda = 0.693/20.8 \text{ h}^{-1}$$

but  $N_t \lambda = N_0 \lambda e^{-\lambda t}$

$N_0$  = number of atoms at zero time.  
 $N_t$  = number of atoms at t.

Solving for  $t$  gives values of 46 and 53 hours for the two experiments in Table 6. Thus the mean life of iodine on the surface is 2.1 days.

Iodine-135 has a half-life of 6.68 h and the amount remaining after 2.1 days decay is 0.6%. As practically all of the  $^{136}\text{I}$  on the surface decays in situ, one would expect the same retention as in the case of  $^{134}\text{I}$ . The data in Table 6 confirm this result.

It is evident from Table 7 that the  $^{133}\text{I}$  concentration is increasing along the duct following shutdown. The relative level at sampling point 4 reaches a value  $2\frac{1}{2}$  times the inlet concentration at  $t_0 + 60$  hours. Hence this nuclide must be coming from the metal and filter surfaces. The release probably occurs by an exchange reaction involving  $^{131}\text{I}$ . At  $t_0 + 20$  h the abundance of this isotope in the gas phase is over 95%, while the abundance of  $^{133}\text{I}$  is higher on the surface than in the gas phase. Exchange at this time will enhance the concentration of the isotope in the gas phase.

The outlet concentration of  $^{133}\text{I}$  is again the sum of two components: a penetrating fraction reduced in concentration by a factor of 10 following shutdown, and an exchanging fraction which escapes from the surface after partial decay. Note that the latter is deposited before shutdown at the higher concentration. Then the increase at the stack is:

$$Q = \frac{(1-R) F + R e^{-\lambda t}}{F} \quad \text{where } Q = \frac{\text{outlet}}{\text{inlet}}$$

$$= \frac{(0.48 \times 0.1) + 0.52 \times 0.21}{0.1} \quad F = \text{decrease in } ^{133}\text{I} \text{ after shutdown}$$

$$= 1.6 \quad \lambda = 0.693/20.8 \text{ h}^{-1}$$

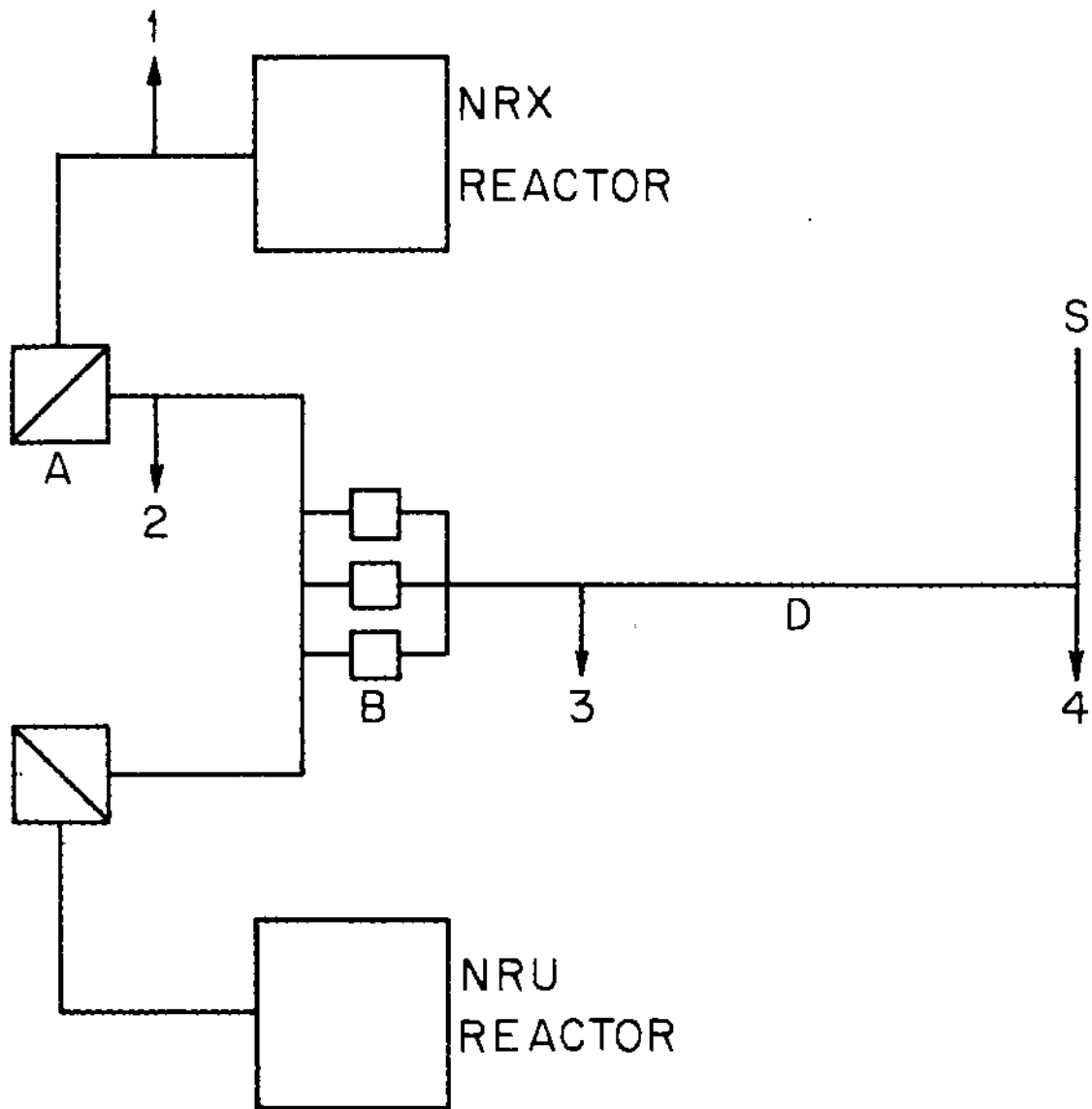
$$t = 2.1 \text{ d}$$

This value can be compared with an observed average of 1.7 from measured concentrations. The agreement supports the calculated mean residence time of 2.1 days.

#### References

1. A.E.J. Eggleton and D.H.F. Atkins, *Radiochim. Acta.* 3(3), 151 (1964).
2. W.J. Megaw and F.G. May, AERE-R-3781 (1961).
3. D.H.F. Atkins and A.E.J. Eggleton, AERE-M-1211 (1963).
4. J. Mishima, *Nuclear Safety* 9 (1), 35 (1968).
5. M.A. Styrikovich et al., *J. Soviet Atomic Energy*, 17, 735 (1964).
6. F.O. Cartan et al., Tenth Air Cleaning Conference, (1968) CONF-680821.
7. J.D. McCormack, BNWL-1145 (1969).
8. P.J. Barry, *Health Phys.* 15, 243 (1968).
9. G.R. Edwards. Personal communication.
10. W.E. Grummitt, Int. Symposium on Identification and Measurement of Environmental Pollutants, Ottawa, June (1971).
11. J.H. Keller et al., Twelfth USAEC Air Cleaning Conference, Aug. (1972).
12. J.B. Morris and B. Nicholls, International Symposium on Fission Product Release and Transport Under Accident Conditions, Oak Ridge, April (1965) CONF. 650407.

The assistance of G.R. Edwards and G. Lahaie is gratefully acknowledged.



A - ABSOLUTE FILTERS  
 B - MAIN FANS  
 D - DUCT  
 S - STACK  
 1, 2, 3, 4 SAMPLE POINTS

Figure 1. Schematic diagram of the reactor ventilation system

EXEMPLE D'ORGANISATION DE LA SECURITE NUCLEAIRE  
DANS UN ETABLISSEMENT UNIVERSITAIRE

Ph. KISSEL - C. RENARD  
Département de Protection - STEPPA  
Commissariat à l'Energie Atomique

Résumé

Le Commissariat à l'Energie Atomique a été appelé à fournir à l'Université une assistance technique en matière de Sécurité Nucléaire.

Les auteurs font la synthèse des nouveaux problèmes qui se sont posés à eux en vue de mettre en place une organisation tenant compte des moyens dont ils disposaient.

Ils précisent ensuite les modalités d'action qu'ils ont élaborées et expérimentées dans un grand établissement universitaire.

Introduction

Parmi tous les établissements dans lesquels sont utilisés des sources de rayonnements ionisants, les établissements d'enseignement et de recherches sont ceux où l'organisation de la sécurité nucléaire est certainement la plus délicate à réaliser.

Après avoir mis en relief certains des problèmes qui se posent en milieu universitaire nous définirons le cadre dans lequel un organisme tel que le Commissariat à l'Energie Atomique (C.E.A.) peut apporter son concours en vue de mettre en place une telle organisation.

Ce cadre étant dressé, nous examinerons ensuite les modalités d'action, telles qu'elles sont actuellement en cours d'expérimentation au sein d'un établissement universitaire.

I. Problèmes particuliers de sécurité nucléaire en milieu universitaire

Si les problèmes de sécurité liés au domaine de la recherche ne sont pas l'apanage exclusif de l'Université, il n'en reste pas moins que les problèmes de sécurité nucléaire liés au domaine de l'Enseignement ne se posent guère que dans ses établissements.

I.I. Problèmes de sécurité nucléaire liés au domaine de l'Enseignement

La mission de l'Enseignant étant d'éduquer ses élèves, ceux-ci se doivent d'être tenus informés des dangers attachés aux matériaux ou aux appareils qu'ils manipulent. De ce fait l'enseignant est généralement mieux attaché aux problèmes de sécurité que son collègue chercheur et, si l'individu est à la fois l'un et l'autre, nous pouvons être assurés qu'il prendra plus de précautions avec ses élèves qu'il n'en prendrait pour lui-même.

Il n'en reste pas moins que la mise en oeuvre de sources de rayonnements ionisants dans une salle de travaux pratiques astreint l'enseignant à prendre des dispositions particulières entraînant l'emploi de matériels ou d'équipements dont la maintenance et le contrôle périodique nécessitent le plus souvent la mise en oeuvre d'autres matériels tout aussi complexes et onéreux. En ce cas le soutien d'un technicien sera précieux à l'enseignant, surtout si ce technicien a une compétence particulière pour répondre à la saine curiosité des élèves et si de plus il prend le soin de les rassurer ou d'éveiller leurs craintes lorsque celles-ci ont un fondement réel.

## I.2. Problèmes de sécurité nucléaire liés au domaine de la recherche

En ce domaine les problèmes se posent différemment du fait d'une complexité plus grande des moyens utilisés.

Toute Unité de Recherche dépendant d'un organisme, fut-il public ou privé, est une source pratiquement intarissable de problèmes de sécurité. Si de plus les travaux effectués entraînent la mise en oeuvre de radioéléments ou de générateurs électriques de rayonnements, ces problèmes peuvent s'amplifier très rapidement et nécessitent, dès leur apparition, la compétence d'un spécialiste en radioprotection.

La Recherche, surtout lorsqu'elle est fondamentale, ce qui est le cas le plus fréquent dans l'Université, n'a pas une voie tracée de façon intangible et ceux qui mènent de tels travaux le savent.

La multiplicité des techniques et des moyens dont la Science dispose actuellement, ajoutée ou plus exactement multipliée, par celle des radioéléments artificiels et de leurs nombreux composés disponibles, fait que le niveau du danger doit, ou devrait être, pratiquement estimé en permanence.

Pour que cette appréciation continue des risques puisse exister, il est non seulement indispensable que le chercheur possède une réelle compétence en matière de sécurité, mais également qu'il s'astreigne à mener ses travaux avec un souci constant des dangers qu'ils peuvent présenter.

Comme une telle association n'est ni forcément courante ni forcément bénéfique aux travaux du chercheur, la solution la plus sûre, tout en étant la solution de facilité apparente, est de faire seconder le chercheur par un cadre compétent : si la jonction est bonne entre les deux parties et si les échanges de vues sont suffisamment ouverts et fréquents, les résultats doivent être satisfaisants.

## I.3. Problèmes liés à l'organisation générale de la sécurité nucléaire dans un établissement universitaire

Il paraît indéniable, au moins en France, que la réglementation actuellement en vigueur relative à la protection des travailleurs contre les rayonnements ionisants a surtout été élaborée en vue de son application dans des établissements à caractère industriel. De ce fait une adaptation devient indispensable lorsqu'il s'agit de l'appliquer en milieu universitaire.

A l'énoncé des critères pouvant être retenus en vue de définir les bases d'une organisation de la sécurité dans un établissement nous ne pouvons que mettre en évidence la nécessité de cette adaptation :

a/ responsabilité de l'employeur : si les notions de responsabilités civile et pénale restent très voisines dans les secteurs industriels et universitaires, il n'en est pas moins vrai qu'un étudiant ne peut être considéré comme un employé;

b/ structures hiérarchiques : si dans l'industrie les responsables sont désignés, dans l'université ils sont le plus souvent élus;

c/ diversité des sources de rayonnements, de leurs approvisionnements et de leurs emplois : il n'y a pas de commune mesure même entre un Centre industriel de recherches et une université, notamment en ce qui concerne la centralisation des approvisionnements;

d/ harmonisation des modalités de surveillance et de contrôle : dans l'industrie ces modalités sont imposées; dans l'université elles doivent être proposées et acceptées;

e/ choix et mise en oeuvre des moyens de prévention et d'intervention : l'industriel fait généralement le nécessaire car il est soumis à certains contrôles réglementaires et doit pouvoir offrir des garanties valables de sécurité à son personnel et à son assureur; l'universitaire n'a pratiquement

que sa seule conscience professionnelle pour prendre ou faire prendre les mesures nécessaires;

f/ discipline et diversité des personnels et visiteurs : s'il n'est nul besoin de s'étendre pour expliquer la différence de discipline dans l'un et l'autre secteur, il peut être bon de rappeler que dans l'université la diversité des personnels est très grande.

g/ information des personnels et réalisation d'exercices de sécurité : si la quasi totalité des personnels employés dans l'industrie nucléaire a suivi des conférences d'information ou des cours de formation en matière de radioprotection, seuls certains universitaires appelés à travailler sur des sources de rayonnements ont effectué un stage de formation à l'Institut des Sciences et Techniques Nucléaires; ce stage de formation sera complété d'une manière indispensable par des exercices de sécurité nucléaire dont seuls les établissements du secteur industriel ont bénéficié jusqu'alors à notre connaissance;

h/ concertation du plan général d'intervention avec les secours publics : si tout établissement détenant des substances dangereuses est classé et répertorié comme tel et si les Secours publics sont en possession d'un plan leur permettant d'intervenir dans ses installations, il est fort vraisemblable à l'heure actuelle que seuls les établissements du secteur industriel procèdent à une mise à jour rigoureuse de ces plans.

i/ sources de financement et mode de gestion des unités : si l'industrie représente un capital qui est, normalement, source de revenus, et si la gestion de ses différentes unités est coordonnée à un niveau élevé, l'unité d'enseignement et de recherches universitaires est pratiquement autonome, son financement étant presque exclusivement assuré par l'allocation de fonds publics transmis par l'intermédiaire de l'Administration : les fonds alloués étant forcément limités, il convient de reconnaître que l'affectation de crédits à l'achat de matériels ou d'équipements de sécurité ne peut être le premier investissement envisagé.

## 2. Cadre des activités C.E.A. - A.T.S.N. auprès de l'université

Les modalités d'action que nous avons élaborées et mises en place dans une grande université française l'ont été dans le cadre de l'ASSISTANCE TECHNIQUE EN SECURITE NUCLEAIRE (A.T.S.N.) apportée par le COMMISSARIAT A L'ENERGIE ATOMIQUE aux organismes publics ou privés qui lui en font la demande.

### 2.I. Assistance en matière de prévention

#### 2.I.I. Aspect technique

Des conseillers techniques sont mis à la disposition de l'Université pour étudier en collaboration avec les responsables des différentes unités :

- a/ la conception particulière des installations,
- b/ l'aménagement et l'équipement des lieux de travail,
- c/ la délimitation des zones réglementées,
- d/ la rédaction de consignes générales de sécurité nucléaire,
- e/ l'organisation de la surveillance radiologique en milieu de travail,
- f/ le problème des déchets et effluents contaminés,
- g/ l'organisation de l'intervention,

h/ les problèmes de décontamination,

#### 2.1.2. Aspects médicaux

Une assistance est également prévue mettant à la disposition du service médical universitaire des médecins-conseils du C.E.A. en vue d'organiser la surveillance médicale des différentes catégories de personnels exposés au danger des rayonnements ionisants :

a/ mise en oeuvre des examens médicaux,

b/ conduite à tenir en cas de contamination ou d'irradiation accidentelle,

#### 2.1.3. Aspects réglementaires

La détention et l'utilisation de sources radioactives (scellées ou non scellées) étant soumises à une réglementation, il importe que les responsables des différentes unités d'enseignement et de recherche soient parfaitement informés des formalités à accomplir et des engagements qu'ils sont conduits à prendre et à tenir vis-à-vis des autorités compétentes. Cette information est faite également par les conseillers techniques du C.E.A.

### 2.2. Assistance en matière d'équipement

#### 2.2.1. Aspects techniques

Le C.E.A. est amené, dans le cadre de son assistance en matière de prévention, à proposer un choix de matériels ou d'équipements individuels ou collectifs de sécurité.

Par ailleurs, il lui est souvent possible de fournir, sous forme de location, ces matériels ou équipements de sécurité.

#### 2.2.2. Aspects économiques

Un avantage indéniable du système locatif est que l'Unité désireuse d'utiliser temporairement un matériel spécifique peut non seulement échapper aux délais courants d'approvisionnement, mais encore éviter un investissement souvent important qu'un emploi momentané du matériel ne justifierait pas.

### 2.3. Assistance en matière d'intervention

L'organisation du C.E.A. est telle que les moyens d'intervention nucléaire dont il dispose pour ses propres besoins peuvent très rapidement être mis en oeuvre. De ce fait, en cas d'accident grave à caractère radioactif ou de sinistre impliquant ou menaçant une quantité notable de substances radioactives, l'Université peut demander l'intervention du C.E.A. en alertant celui-ci suivant une procédure d'alerte parfaitement définie.

Dès son arrivée dans l'établissement accidenté l'équipe d'intervention nucléaire du C.E.A. se place sous l'autorité du représentant des autorités publiques ou, en son absence, se met à la disposition du responsable de l'établissement pour l'assister de ses conseils et de ses moyens.

De plus le chef de cette équipe C.E.A. a pour mission de proposer toute assistance complémentaire qu'il juge nécessaire et possible et de provoquer la mise en oeuvre de celle-ci lorsqu'elle est demandée soit par le responsable de l'établissement, soit par le représentant des autorités publiques.

Par la suite le C.E.A. rend compte au Ministère de la Santé Publique (Service Central de Protection contre les Rayonnements Ionisants) des dispositions qu'il a été amené à prendre au cours de son intervention.



### 3. Modalités d'action du C.E.A. au sein d'un établissement universitaire

#### 3.1. Modalités d'action sur le plan technique

Il nous est avant tout apparu nécessaire de constituer un dossier suffisamment précis pour que chacun des responsables concernés de l'Université tant à l'échelon central qu'à l'échelon des unités soit systématiquement tenu informé des problèmes de sécurité nucléaire se posant dans leurs installations.

Le premier travail des conseillers techniques du C.E.A. étant de faire un bilan des dangers radioactifs et d'en déduire soit la conception particulière d'installations à l'état de projet, soit les aménagements à conseiller si ces installations sont déjà réalisées, nous avons mis au point deux documents techniques originaux :

- une fiche de sécurité nucléaire collective dite "fiche de zone"
- une fiche de sécurité nucléaire individuelle dite "fiche individuelle".

##### 3.1.1. Fiche de zone (cf. annexe I)

Cette première fiche de sécurité nucléaire permet de rassembler sur un seul document tous les renseignements utiles relatifs à une zone de travail déterminée.

Une fois établie cette fiche permet non seulement de conseiller sur le choix ou la validité des équipements et aménagements de la zone considérée, mais également de déterminer le classement de celle-ci conformément à la législation en vigueur en vue de son balisage.

Le dossier constitué par l'ensemble de ces "fiches de zone" permet de faire le recensement à un instant donné de toutes les sources de rayonnements présentes dans un établissement; donc d'ouvrir un état qu'il suffira de tenir à jour pour évaluer ultérieurement le potentiel des risques radiologiques dans les installations.

Ce dossier permet également de préparer, sur des bases valables, le plan général d'intervention, notamment en ce qui concerne l'intervention nucléaire.

##### 3.1.2. Fiche individuelle (cf. annexe II)

Cette seconde fiche de sécurité nucléaire vise à définir les risques auxquels est exposé un individu : elle est donc nominative. Ce document peut d'ailleurs être utilisé soit dans un cadre préventif, ce qui est évidemment conseillé lorsque l'on peut prévoir à l'avance l'affectation ou les affectations successives d'un individu, soit sous forme de récapitulation des travaux effectués par celui-ci durant une période antérieure déterminée.

Etablie à titre préventif, cette fiche permet au conseiller technique C.E.A. :

- de classer l'individu en regard de la législation sur la protection des travailleurs contre les rayonnements
- de fixer le choix des dosimètres individuels, de la tenue de travail et des divers équipements individuels
- de faire le point quant à la nature des risques d'exposition auxquels sera soumis l'individu en vue de sa surveillance systématique éventuelle et notamment de sa surveillance médicale.

#### 3.2. Modalités d'action sur le plan médical

L'assistance du C.E.A. en ce domaine se traduit essentiellement par un rôle de conseil.

A cet effet des médecins-conseils C.E.A. se tiennent à la disposition de leurs collègues du service médical universitaire.

Afin d'établir un lien entre les conseillers techniques et le corps médical, nous avons mis au point, en collaboration avec les médecins-conseils du C.E.A., un troisième document (cf. annexe III) intitulé "FICHE DE CONSEILS MEDICAUX" qui est une fiche de sécurité nucléaire individuelle faisant suite à la "FICHE INDIVIDUELLE". Sur la base des renseignements portés par le technicien-conseil C.E.A. sur la fiche individuelle, le médecin de l'établissement universitaire ou, sur la demande de ce dernier, le médecin conseil du C.E.A., est en mesure de définir quelles doivent être la nature des examens que doit subir l'intéressé et la périodicité de ces examens.

Comme on peut le constater ce troisième document établi et exploité par des médecins reste entièrement confidentiel et peut être versé au dossier médical de l'individu avec, en pièce jointe, l'exemplaire de la fiche individuelle adressé au corps médical.

### 3.3. Modalités d'action sur les plans administratif et financier

#### 3.3.1. Modalités administratives

Sur le plan contractuel nous avons mis au point, en collaboration avec les services administratifs centraux du C.E.A., le texte d'une "CONVENTION D'ASSISTANCE TECHNIQUE EN SECURITE NUCLEAIRE" qui rassemble tous les éléments indispensables à un tel contrat. Ce modèle de convention peut d'ailleurs être utilisé aussi bien avec un établissement industriel ou universitaire qu'avec un établissement relevant du milieu médical mais, dans ce dernier cas, un accord préalable du Ministère de la Santé Publique (S.C.P.R.I.) est nécessaire.

#### 3.3.2. Modalités financières

Depuis 1972 nos prestations sont facturées et leur règlement est effectué par un Service Central de l'administration universitaire qui prend donc à sa charge la sécurité de toutes les unités placées sous sa tutelle.

Cette centralisation confère une grande souplesse à notre action auprès des différentes unités, celles ayant le plus de besoins n'étant pas forcément les mieux financièrement dotées.

### Conclusions

Le début de notre action est encore trop récent pour que nous puissions considérer que les modalités proposées seront définitivement retenues.

Compte tenu de notre première expérience en milieu universitaire, nous espérons avoir jeté les bases d'une organisation qui permettra d'attendre avec plus de quiétude d'autres expériences de même nature.

Restant soucieux d'améliorer sans cesse tant la qualité de nos rapports avec l'université que celle de nos services, nous souhaiterions pouvoir profiter de l'expérience de ceux qui se sont, ailleurs dans le monde, penchés sur ce même problème.

EXPOSURES FROM RADIATION SOURCES  
OF NATURAL ORIGIN

ИССЛЕДОВАНИЕ И НОРМИРОВАНИЕ  
РАДИОАКТИВНОСТИ СТРОИТЕЛЬНЫХ МАТЕРИАЛОВ

Э.М.Кривош, Э.Н.Лисаченко, С.К.Тарасов,  
В.И.Камов, Н.И.Шалак

Институт радиационной гигиены  
Министерство здравоохранения РСФСР  
Ленинград, СССР

Abstract.

Ionizing radiation in dwellings is one of the main sources of radiation effect on the population. Limitation or decrease of this factor may be done by standardization of radioisotope concentration in building materials. Gamma-spectrometric analyses for Ra-226, Th-232 and K-40 in 300 samples of building materials from various regions of the USSR have been performed and predicted values of gamma radiation doses in buildings made of these materials have been calculated. To evaluate the parameters determining air concentration of Rn in dwellings Rn accumulation and mechanism of its release have been studied. For limitation of external gamma radiation levels in dwellings the following values of specific radioisotope permissible concentration have been suggested: 10, 7, 126 pCi/g for Ra-226, Th-232 and K-40 respectively. For limitation of Rn concentration in dwellings permissible level of effective Ra-226 concentration (the product of Ra-226 concentration by emanation factor) has been recommended 0.6 pCi/g.

Резюме

Ионизирующее излучение в жилых помещениях является одним из основных источников радиационного воздействия на население. Ограничение или уменьшение этого фактора возможно путем нормирования содержания радиоизотопов в строительных материалах.

Проведены гамма-спектрометрические исследования содержания радия-226, тория-232 и калия-40 в 300 образцах стройматериалов из различных районов СССР и рассчитаны ожидаемые дозы гамма-излучения в помещениях, построенных из этих материалов. Для выявления параметров, определяющих содержание радона в воздухе помещений изучен механизм накопления радона и механизм радоновыделения.

Для ограничения внешнего гамма-излучения в помещениях предложены следующие значения допустимого содержания отдельных радиоизотопов: Ra-226 - 10; Th-232 - 7; K-40 - 126 пКи/г. Для ограничения концентрации радона в жилых помещениях рекомендовано допустимое значение эффективной концентрации радия /произведенное количество радия на коэффициент эманирования/, равное 0,6 пКи/г.

## Введение

В настоящее время наибольший вклад в облучение населения в целом дают источники ионизирующей радиации естественного происхождения. Величина дозы облучения населения за счет большинства естественных источников, таких как космическое излучение, излучение земных пород, а также калия и некоторых других радиоактивных изотопов, содержащихся в организме человека, практически не может быть изменена. Особое положение в этом отношении занимает излучение строительных материалов, используемых для сооружения жилых, общественных, промышленных зданий и дорожных покрытий. Величина дозы облучения за счет излучения строительных материалов варьирует в довольно широких пределах в зависимости от содержания в них естественных радиоактивных изотопов.

Резко выраженная урбанизация населения, наблюдающаяся во всех странах, приводит к расширению масштабов жилищного строительства. При этом происходит массовое переселение людей из деревянных в кирпичные или бетонные дома, уровни излучения в которых, как правило, выше, чем в деревянных. Кроме того, развитие массового жилищного строительства приводит к необходимости изыскания новых, более дешевых материалов. В качестве таковых все чаще используются отходы горнорудной, металлургической и химической промышленности. Эти отходы могут содержать радиоактивные вещества в больших количествах, чем традиционно используемые строительные материалы<sup>2</sup>. В связи с этим возникает необходимость в ограничении использования таких материалов из-за возможности существенного повышения уровня облучения населения.

Ограничение или уменьшение уровня облучения населения за счет излучения строительных материалов возможно путем нормирования их радиоактивности, то есть путем отказа от использования материалов с высоким содержанием радиоактивных веществ. Для обоснования таких нормативов необходимо достаточно подробно изучить существующие уровни радиационного воздействия на население излучения строительных материалов, их зависимость от вида используемых материалов и т.д.

### Гамма-излучение в жилых помещениях

В различных странах /Швеция, Англия, СССР, США, Австрии, Японии, Болгарии, ФРГ, ГДР/ проведены многочисленные измерения дозы гамма-излучения в помещениях. Значения гамма-фона в помещениях из различных строительных материалов и на открытой местности согласно этим измерениям представлены в таблице I<sup>1, 3-6</sup>.

Таблица I

Дозы гамма-излучения внутри и вне помещений  
за вычетом космического фона /мрад/год/

Тип строительного материала	Внутри помещения			Вне помещения			Разность средних доз
	мин.	макс.	сред.	мин.	макс.	сред.	
Дерево	29	100	56	23	119	56	0
Кирпич	26	346	87	21	308	78	9
Бетон	48	202	115	23	120	70	45
Гранит	75	300	125	18	118	74	51

Из таблицы видно, что доза гамма-излучения в помещениях выше дозы на открытой местности /за исключением деревянных домов/.

Стены и перекрытия помещений являются источниками излучения и экранами от космического излучения и излучения земных пород. Экранирование космических лучей перекрытиями зданий невелико. После прохождения 1, 2 и 3 перекрытий космическое излучение ослабляется на 13, 18 и 21 процент соответственно. Степень экранирования излучения земных пород зависит от толщины стен помещения. Для излучения естественных радиоактивных изотопов толщина слоя насыщения составляет 100 г/см<sup>2</sup>, при этом слой 50 г/см<sup>2</sup> обеспечивает 80% насыщения. При толщинах стен и перекрытий, близких к слою насыщения и характерных для современного жилищного строительства, излучение земных пород полностью экранируется. В этом случае мощность дозы в помещении определяется концентрацией радиоактивных веществ в используемых строительных материалах. Для определения коэффициентов пропорциональности между этими величинами можно воспользоваться значениями, рассчитанными для полости в бесконечном пространстве. Эти значения вычислялись рядом авторов. И хотя ими применялись различные методы расчета, полученные коэффициенты оказались весьма близкими. Используя коэффициенты, приведенные в работе для максимально возможной мощности дозы в помещении, построено целиком из данного материала /за вычетом космического фона/ получаем:

$$D_{\text{мкр/час}} = 4,7 C_{Ra} + 6,7 C_{Th} + 0,37 C_K \quad /1/$$

где  $C_{Ra}$ ,  $C_{Th}$  и  $C_K$  - удельные активности радия-226, тория-232 и калия-40 в строительном материале в пКи/г при наличии радиоактивного равновесия в рядах урана и тория.

В реальных помещениях мощность дозы может оказаться несколько меньше значения, полученного по формуле /1/ как за счет наличия окон и дверей, так и в ряде случаев недостаточной толщины стен и перекрытий. Кроме того, для сооружения стен и перекрытий могут использоваться разные строительные материалы. В этих случаях мощность дозы в помещении будет иметь промежуточные значения между результатами расчета для отдельных материалов.

Для определения диапазона изменений концентраций радиоактивных веществ в строительных материалах нами, с помощью низкофонового гамма-спектрометра, было исследовано около 300 образцов различных строительных материалов из разных районов СССР. Результаты измерений представлены в таблице II. В ней также приведены величины мощности дозы в помещении, рассчитанные по формуле /1/. Величина годовой дозы, обусловленной излучением строительных материалов в предположении 18-часового пребывания людей в помещении и с учетом коэффициента экранирования гонад, равного 0,68<sup>9</sup> может быть вычислена по формуле:

$$D_{\text{мрад/год}} = 18,5 C_{Ra} + 26,7 C_{Th} + 1,47 C_K \quad /2/$$

При обосновании нормативов радиоактивности строительных материалов мы исходили из того, что за счет проживания в домах, построенных из материалов с повышенным содержанием радиоактивных изотопов допустимо дополнительное облучение гонад, равное 100 мрад/год, по сравнению с облучением в домах со средним содержанием радиоактивных веществ. 100 мрад/год это средняя доза облучения гонад за счет излучений естественных источников<sup>9</sup>. Поэтому такое нормирование ограничивает гонадную дозу людей, проживающих в домах, построенных из строительных материалов с повышенным содержанием радиоактивных веществ, величиной удвоенного естественного фона. Годовая доза в домах из обычных материалов может быть подсчитана по формуле /2/ для концентрации радиоактивных изотопов, равной кларковой / $C_{Ra} = 0,9$ ;  $C_{Th} = 1,41$ ;  $C_K = 21,5$  пКи/г<sup>10</sup>/. Она со-

Таблица II

Содержание естественных радиоизотопов в строительных материалах  
/пКи/г/ и мощность дозы в помещении /мкр/час/

Тип материала	Число проб	Торий-232			Радий-226			Калий-40			Мощность дозы в помещении			Суммарное содержание изотопов /в долях ПДК/		
		мин.	мак.	ср.	мин.	мак.	ср.	мин.	мак.	ср.	мин.	мак.	ср.	мин.	мак.	ср.
Красный кирпич	55	0,66	1,5	1,0	1,1	1,6	1,5	11	25	20	13	25	20	0,27	0,55	0,42
Бетон тяжелый	87	0,4	3,7	0,8	0,6	2,9	0,9	7	24	15	8	45	14	0,16	0,95	0,31
Бетон легкий	16	0,6	2,1	0,9	1,4	3,3	2,0	5	26	14	12	37	19	0,26	0,8	0,41
Гранит	2	-	-	4,5	-	-	3	-	-	40	-	-	56	-	-	1,20
Т у ф	13	1,1	2,9	2,0	2,1	2,6	2,6	26	34	18	24	41	30	0,52	0,88	0,69
Песок природный	18	0,4	0,8	0,5	0,4	1,0	0,5	2	17	8	4	15	8	0,09	0,33	0,17
Песок хвостовой	14	-	-	0,5	-	-	0,8	-	-	6	-	-	8	-	-	0,19
Цемент	7	0,4	0,5	0,4	0,4	3,2	1,2	3	11	6	5	22	10	0,12	0,46	0,22
Шлаки доменные	29	0,3	1,2	0,6	1,0	3,4	1,8	0,9	13	13	7	28	16	0,14	0,59	0,34
Шлаки фосфорные	15	-	-	0,6	-	-	0,6	-	-	4	-	-	33	-	-	0,72
Щебень	8	0,4	5,0	2,1	0,4	0,4	0,4	6	8	7	6	38	18	0,14	0,81	0,38
Облицовочные материалы	35	0,4	4,7	2,3	0,4	3,4	1,9	22	47	39	11	61	36	0,24	1,3	0,76

составляет 36 мрэд/год. Увеличение годичной дозы за счет прощивания в домах, построенных из материалов с повышенным содержанием радиоактивных веществ, определяется по формуле:

$D$  /мрэд/год/ = 18,5  $C_{Ra}$  + 26,7  $C_{Th}$  + 1,47  $C_K$  - 86 /3/  
 ПДК радиоактивных изотопов в строительных материалах могут быть рассчитаны по формуле /3/ при условии, что доза дополнительного облучения не должна превышать 100 мрэд/год. Они составляют:  
 $C_{Ra} = 10$ ;  $C_{Th} = 7,0$ ;  $C_K = 126$  пКи/г. При наличии в строительном материале этих изотопов должно выполняться условие:

$$\frac{C_{Ra}}{10} + \frac{C_{Th}}{7,0} + \frac{C_K}{126} \leq 1 \quad /4/$$

В таблице II приведены суммарные концентрации радиоактивных веществ в исследованных материалах /в долях ПДК/. Из таблицы видно, что превышение допустимой концентрации обнаружено только у гранита и некоторых отделочных материалов.

Таким образом, предлагаемые величины допустимого содержания радиоактивных веществ не приведут к ограничению применения подавляющего большинства традиционных строительных материалов. Ограничения распространяются на относительно небольшое число материалов с повышенным содержанием радиоактивных веществ. Такие материалы могут применяться при их разбавлении слабо радиоактивными компонентами, а также для сооружения дорог, плотин, времяпрепровождение людей вблизи которых ограничено.

#### Радон в воздухе жилых помещений

Наличие радиоактивных веществ в строительных материалах, помимо дополнительного внешнего облучения приводит к повышению радиоактивности воздуха в помещении по сравнению с атмосферным. Параметрами, определяющими степень радиационного воздействия радиоактивности воздуха, являются концентрация радона, торона и их продуктов распада. В литературе неоднократно отмечались случаи довольно высоких концентраций радона в помещениях, особенно в домах из материалов, изготовленных из отходов горно-рудной промышленности /вплоть до 15 пКи/л/ 11, 12. Наличие таких случаев свидетельствует о необходимости контроля за радиоактивностью воздуха в помещениях. Однако концентрация эманаций и их короткоживущих продуктов распада неудобны в качестве параметров для контроля, поскольку они связаны со степенью воздухообмена, которая варьирует в довольно широких пределах в зависимости от метеословий, длительности проветривания помещений и т.д.

Для выявления более удобного для контроля параметра достаточно точно характеризующего радиоактивность воздуха в помещении, нами был рассмотрен механизм накопления радона в помещении. Решение соответствующего дифференциального уравнения дает выражение для изменения во времени концентрации радона в воздухе помещения:

$$C = \frac{QS + V K C_{атм}}{\lambda + K} [1 - e^{-(\lambda + K)t}] + C_0 e^{-(\lambda + K)t} \quad /5/$$

где:  $C_0$  - концентрация радона в момент  $t = 0$  /Ки/м<sup>3</sup>/  
 $Q$  - удельное радоноразделение ограждений /Ки/м<sup>2</sup>сек/  
 $S$  - площадь ограждений помещения /м<sup>2</sup>/  
 $V$  - объем помещения /м<sup>3</sup>/  
 $\lambda$  - постоянная распада радона /2,1 · 10<sup>-6</sup> сек<sup>-1</sup>/  
 $K$  - кратность воздухообмена в помещении /сек<sup>-1</sup>/

\*  $x$  / предельно допустимые концентрации

$C_{атм}$  – концентрация радона в атмосферном воздухе /Кл/м<sup>3</sup>/

Из формулы /5/ следует, что установление равновесной концентрации радона в помещении определяется величиной  $\lambda + K$ . При кратности воздухообмена, равной одному обмену в час / $K = 2,8 \cdot 10^{-7}$  сек<sup>-1</sup>/, равновесная концентрация достигается уже через несколько часов. Выражение для равновесной концентрации радона с учетом того, что  $\lambda \ll K$ , записывается в виде:

$$C_{равн.} = \frac{Q}{K} \frac{S}{V} + C_{атм} \quad /6/$$

Из формулы видно, что концентрация радона в помещении всегда выше концентрации в наружном воздухе. Разность этих концентраций пропорциональна удельному радоновыделению ограждений помещения, отношению  $S/V$  и обратно пропорциональна кратности воздухообмена. Отношение  $S/V$  для обычных помещений не сильно варьирует. Его значение для типичных жилищ даны в таблице II, из которой следует, что среднее значение этого отношения составляет величину 1,5.

Таблица II

Отношение площади ограждений к объему помещения /  $S/V$  /

высота /M/ \ S /M <sup>2</sup> /	15 = 4 x 3,75	20 = 5 x 4	30 = 5x6	40=6x6,7
2,5	1,85	1,70	1,58	1,43
3,0	1,70	1,57	1,40	1,30
3,5	1,60	1,47	1,30	1,20

Формула /6/ позволяет оценить поступление радона из почвы под зданием. В работе [13] приводятся величины удельного радоновыделения почв: среднее значение  $4,5 \cdot 10^{-13}$ , диапазон изменений от  $2 \cdot 10^{-15}$  до  $1,7 \cdot 10^{-12}$  Кл/сек.м<sup>2</sup>. Расчетное значение концентрации радона в помещении размером 5 x 4 и высотой 3 м, в предположении, что почвенный радон полностью поступает в помещение, а радоновыделение ограждений отсутствует и концентрация эманации в атмосферном воздухе равна 0,1 пКи/л, приведено в таблице IV.

Таблица IV

Расчетные значения концентрации радона в помещении / пКи/л/ при поступлении радона из почвы

Q /Кл/м <sup>2</sup> сек/ \ K /час <sup>-1</sup> /	$1,7 \cdot 10^{-12}$	$4,5 \cdot 10^{-13}$	$2 \cdot 10^{-15}$
0,1	20,5	5,5	0,12
1	2,1	0,64	0,10
10	0,3	0,15	0,10

Из таблицы видно, что в помещениях с плохой изоляцией пола поступление радона из почвы может играть существенную роль. В современном многоэтажном строительстве используются бетонные перекрытия, которые практически полностью предотвращают поступление в жилые помещения почвенного радона. В таких домах основным источ-



ником радона является радонвыделение стен и перекрытий. Удельное радонвыделение ограждений является более удобным параметром, поскольку его величина, в отличие от концентрации радона, практически не зависит от воздухообмена в помещении. Это было установлено в результате рассмотрения механизма радонвыделения. Процесс радонвыделения можно разделить на два этапа: эманирование радона во внутренние поры материала /доля атомов радона, выходящих во внутренние поры, называется коэффициентом эманирования/ и диффузию радона по этим порам с выходом из материала. Такое разделение оправдано тем, что внутри зерен минерала диффузия протекает крайне медленно /коэффициент диффузии имеет порядок  $10^{-22}$  см<sup>2</sup>/сек<sup>14</sup>/. Поэтому распространяются по материалу только те атомы радона, которые вышли за счет отдачи при альфа-распаде радия во внутренние поры материала. Процесс распространения радона по порам материала описывается уравнением диффузии. При рассмотрении радонвыделения стен можно считать, что диффузный перенос радона осуществляется в направлении, перпендикулярном к поверхности стен /по координате  $x$ /. Поток радона, параллельные поверхности стены, взаимно уравновешивают друг друга, так как высота и ширина стены значительно больше ее толщины. В этом случае уравнение диффузии может быть записано в виде:

$$\frac{\partial C}{\partial t} = \lambda C_0 - \lambda C + \frac{v}{\delta} \frac{\partial^2 C}{\partial x^2} \quad /7/$$

где

$$C_0 = \frac{C_{Ra} \rho b}{\delta} \quad /8/$$

- $C_0$  - максимально возможная концентрация радона /Ки/см<sup>3</sup>/,
- $C_{Ra}$  - концентрация радия в материале /Ки/г/,
- $\rho$  - плотность материала /г/см<sup>3</sup>/,
- $b$  - коэффициент эманирования /отн.ед./,
- $\delta$  - пористость материала /отн.ед./,
- $v$  - коэффициент диффузии /см<sup>2</sup>/сек/

За начало координат принята половина глубины стены. В силу симметрии задачи поток радона при  $x=0$  равен 0. Отсюда следует первое граничное условие

$$v \frac{\partial C}{\partial x} \Big|_{x=0} = 0 \quad /9/$$

Второе граничное условие можно сформулировать, исходя из баланса активности вне стены. Будем считать, что диффузия радона из стены толщиной  $2d$  происходит в ограниченный внешний объем глубиной  $e$ . Для простоты можно принять, что на выходе из стены происходит мгновенное выравнивание концентрации радона. При этом второе граничное условие может быть записано в виде:

$$-v \frac{\partial C}{\partial x} \Big|_{x=d} - e \lambda C \Big|_{x=d} = e \frac{\partial C}{\partial t} \Big|_{x=d} /10/$$

Решение уравнения /7/ с граничными условиями /9/ и /10/ для стационарного случая /  $\partial C / \partial t = 0$  / имеет вид:

$$C_x = C_0 \left[ 1 - \frac{\operatorname{ch}\left(\frac{x}{l_0}\right)}{\operatorname{ch}\beta + \sqrt{\beta} \operatorname{sh}\beta} \right] \quad /11/$$

где

- $l_0 = \sqrt{\frac{v}{\lambda \delta}}$  - длина диффузии,
- $L = e / d \delta$  - отношение внешнего к внутреннему объему воздуха
- $\beta = d / l_0$  - отношение половины толщины стены к длине диффузии.

Концентрация радона во внешнем воздухе / $C \Big|_{x=d}$ / выражается

$$C|x=d = C_0 \left[ \frac{1 + \frac{th\beta}{\beta}}{1 + \frac{1}{d} \frac{th\beta}{\beta}} \right] \quad /12/$$

Удельное радоновыделение равняется:

$$Q = \beta \frac{\partial C}{\partial x} \Big|_{x=d} = Q_0 \frac{th\beta}{\beta} \left[ \frac{1}{1 + \frac{1}{d} \frac{th\beta}{\beta}} \right] \quad /13/$$

где  $Q_0$  - максимально возможное радоновыделение, то есть радоновыделение в случае, когда весь радон, попавший во внутренние поры, выходит наружу.

Оно равно:

$$Q_0 = C_{Ra} \beta \lambda d \rho \quad /14/$$

Для жилых помещений были рассмотрены два случая:

1. Отношение объема к объему внутривоздуха  $|d|$  велико. В этом случае

$$\frac{C(d)}{C_0} \Big|_{d \rightarrow \infty} \rightarrow 0; \quad Q \Big|_{d \rightarrow \infty} \rightarrow Q_0 \frac{th\beta}{\beta} \quad /15/$$

Это приближение справедливо для диффузии радона в сильно вентилируемое помещение, поскольку при этом эффективный внешний объем значительно превосходит объем помещения.

2. Для неветилируемого помещения внешний объем равен объему помещения и величина  $|d|$  в этом случае равна:

При  $S/V = 1,5 \text{ м}^{-1}$ ,  $d = 0,25 \text{ м}$ ,  $\delta = 0,4$ ; величина  $d = \sqrt{S d \delta} \quad /16/$   
 $d = 6,7 \frac{th\beta}{\beta}$  всегда меньше 1. Принимая  $th\beta/\beta = 1$ , получим:  $Q = 0,87 Q_0$ ,  $C|x=d = 0,13 C_0$ . Если  $th\beta/\beta < 1$ , то  $Q$  будет еще меньше отличаться от  $Q_0$ .

Таким образом, для помещений обычных размеров величина удельного радоновыделения практически не зависит от кратности воздухообмена, то есть этот параметр является удобным для контроля. Однако измерение радоновыделения возможно провести либо в уже построенных помещениях, либо на макетах ограждений. Это создает определенные трудности при радиационно-гигиенической оценке новых строительных материалов. В последнем случае желательно уметь определять его величину путем исследования небольших образцов материала.

Из выражений /14/ и /15/ следует, что удельное радоновыделение пропорционально произведению концентрации радия в строительном материале на коэффициент эманирования, которое назовем эффективной концентрацией радия:

$$C_{Ra \text{ эфф}} = C_{Ra} \beta \quad /17/$$

Кроме того величина радоновыделения зависит от  $\beta$  - отношения половины толщины стены к длине диффузии. Функция  $th\beta/\beta$  при  $\beta \leq 1$  не сильно отличается от единицы, а при  $\beta \geq 2$  имеет вид  $\sim 1/\beta$ . Таким образом, если длина диффузии радона в материале ограждения больше половины толщины ограждения, то радоновыделение можно оценивать по его максимальной величине  $Q_0$ . Следует отметить, что мы рассматривали чисто диффузное приближение. Наличие же подпора ветра, перепад температур между комнатным и наружным воздухом, могут привести к появлению конвекционных потоков, т.е. к уменьшению эффективного значения  $\beta$ . При этом радоновыделение будет возрастать, но в любом случае оно не может превысить величину  $Q_0$ . Следовательно, в реальных условиях возможны измене-

ния удельного радонвыделения в пределах от  $Q_0 \text{ th } \beta/\beta$  до  $Q_0$ . Если  $\beta$  невелико, то эти изменения будут незначительны. Результаты экспериментального определения длины диффузии радона в строительных материалах и характерные для этих материалов величины  $\beta$  приведены в таблице У.

Таблица У

Длина диффузии радона в строительных материалах

Вид материала	Длина диффузии $l_0$ /см/	Характерная толщина ограждений $2d$ /см/	$\beta = \frac{d}{l_0}$
Бетон тяжелый	15; 10; 13;	10	0,4
Бетон легкий	29; 28; 22	12 + 35	0,2 - 0,7
Красный кирпич	15	50	1,7

Принимая во внимание данные таблицы У можно заключить, что для большинства ограждений радонвыделение будет определяться максимально возможной величиной, которая является функцией только эффективной концентрации радия в строительном материале. Эффективная концентрация радия является параметром, удобным для контроля за радиоактивностью воздуха в помещении, поскольку ее возможно определять путем исследования небольших образцов материала.

Эманационным методом /при изменении до нескольких мм/ нами измерена эффективная концентрация радия в нескольких десятках образцов различных строительных материалов и рассчитаны равновесные концентрации радона в помещениях из данных материалов. Расчет концентрации радона проводился для постоянного воздухообмена, равного 1/час,  $S/V = 1,5$ . Толщина ограждений и плотность для тяжелого и легкого бетонов принимались соответственно равными  $2d = 10$  и  $20$  см;  $\rho = 2,1$  и  $1,6$  г/см<sup>3</sup>. Для всех остальных материалов принималось  $2d = 50$  см;  $\rho = 1,5$  г/см<sup>3</sup>. Радонвыделение считалось максимально возможным, концентрация радона в атмосферном воздухе не учитывалась. В таблице VI представлены также значения коэффициентов эманирования, рассчитанные с привлечением результатов гамма-спектрометрического определения концентрации радия в данных образцах.

Таблица VI

Эффективная концентрация радия в строительных материалах и расчетное значение концентрации радона в помещениях

Тип материала	Эффект. концентрация радия $C_{Ra}^{эфф} \times 10^{-2}$ /пкв/г/			Коэффициент эманирования $\epsilon$ /%/			Концентрация радона в помещении $C_{Rn}$ /пкв/г/		
	мин.	макс.	ср.	мин.	макс.	ср.	мин.	макс.	ср.
Бетон тяжелый	0,8	12	4,1	0,4	10,0	3,3	0,02	0,29	0,10
Бетон легкий	2,0	13,2	5,4	0,4	10,0	2,1	0,7	0,4	0,2
Красный кирпич	0,8	5,1	2,1	0,4	5,7	1,7	0,03	0,22	0,09
Кирпич силикатный	2,4	6,2	3,8	4,5	8,0	6,7	0,10	0,27	0,16
Тuff	3,5	10,2	6,5	1,8	6,0	3,6	0,15	0,43	0,28
Кирпич пумици-товый	-	-	6,1	-	-	5,8	-	-	0,26
Пумицит	-	-	28,0	-	-	9,0	-	-	1,2

Допустимую величину эффективной концентрации радия можно установить, исходя из условия, чтобы при постоянном воздухообмене, равном  $1 \text{ час}^{-1}$ , не было превышения СДК<sup>х</sup> короткоживущих продуктов распада радона /С<sub>т</sub>/ для населения / $1 \cdot 10^{-9} \text{ Ки/м}^3 \cdot \text{л}^3$ /. Принимая, что радоновыделение ограждений определяется максимально возможной величиной /время пребывания в помещении 18 часов в сутки/ и пренебрегая величиной С<sub>атм.</sub>, выражение для С<sub>раэфф</sub> можно записать в виде:

$$C_{Raэфф} \text{ /Ки/г/} = 10^{-6} \frac{C_{ра} \cdot K}{0,75 \alpha \rho \lambda \cdot S/V} \quad /18/$$

При воздухообмене  $1 \text{ час}^{-1}$  концентрация короткоживущих продуктов распада в помещении составляет  $0,58 C_{ра}$ . С учетом этого:

$$C_{Raэфф} \text{ /Ки/г/} = 10^{-6} \frac{C_1 \cdot K}{0,4 \alpha \rho \lambda \cdot S/V} \quad /19/$$

Значение допустимой величины С<sub>раэфф</sub> можно получить из формулы /19/ принимая  $\alpha = 0,25 \text{ м}$ ;  $\rho = 1,5 \text{ т/м}^3$ ;  $S/V = 1,5 \text{ м}^{-1}$

$$C_{Raэфф} = 0,6/\pi \text{ Ки/г/} \quad /20/$$

Как видно из таблицы VI среди исследованных материалов не оказалось ни одного, имеющего эффективную концентрацию радия, близкую к допустимой величине. Сопоставление предлагаемых нормативов концентрации радия, определяющей внешнее облучение людей, и эффективной концентрации радия, определяющей радиоактивность воздуха в помещении, показывает, что первый норматив является, как правило, более жестким. Нормирование по эффективной концентрации радия может быть лимитирующим фактором только для материалов, радиоактивность которого обусловлена, в основном, радием и обладающих повышенным эманированием / $\beta > 6\%$ /. Наблюдающиеся рядом авторов концентрации радона в помещении, превышающие  $1 \text{ пКи/л}$ , во многих случаях могут быть связаны с тем, что измерения производились при пониженном воздухообмене. Кратность воздухообмена, как правило, при этом не измерялась. Величина же допустимой эффективной концентрации радия рекомендована, исходя из среднегодовой величины кратности воздухообмена, и по этой причине ограничивает только среднегодовую концентрацию радона в помещении.

Исходя из меньшей значимости радиоактивности воздуха по сравнению с внешним облучением, контроль за эффективной концентрацией радия целесообразно проводить только для материалов с повышенным содержанием радия /более  $5 \text{ пКи/г}$ /. Следует иметь в виду, что материалы с эффективной концентрацией радия, превышающей допустимую, можно применять при условии использования противорадоновых покрытий. Наши исследования показали, что двухкратное покрытие стен масляной краской снижает радоновыделение на порядок. Кроме того концентрацию радона в помещении можно уменьшить путем увеличения воздухообмена.

С целью проверки результатов теоретического рассмотрения механизмов накопления в помещении и радоновыделения, точности методов измерения параметров, характеризующих эти процессы, а также соотношения между дозой внешнего гамма-излучения в помещении и концентрацией радиоизотопов в стройматериалах нами проведены исследования в модельном помещении. Помещение было построено из материалов, содержание радиоактивных веществ в которых, коэффициенты эманирования и другие параметры были предварительно исследованы. В помещении

х/ средняя допустимая концентрация

был обеспечен регулируемый воздухообмен. Рассчитанные и измеренные значения удельного радоновыделения различных ограждений представлены в таблице VII.

Таблица VII

Радоновыделение ограждений экспериментального помещения

Тип ограждения	Удельное радоновыделение /Ки/м <sup>2</sup> сек/10 <sup>14</sup> .	
	Расчетные значения	Измеренное значение
Внешняя стена	1,8	2,0 ± 0,1
Внутренняя стена	1,1	1,14 ± 0,06
Пол	0,7	1,20 ± 0,07
Потолок	0,9	0,80 ± 0,08

Из таблицы видно, что наблюдается хорошее согласие расчетных и экспериментальных значений удельного радоновыделения для всех ограждений, кроме пола, для которого измеренное значение в 1,5 раза выше расчетного. Это расхождение обусловлено частичным проникновением радона из подвального помещения.

Расчетные и измеренные значения концентрации короткоживущих продуктов распада радона при различных кратностях воздухообмена в экспериментальном помещении даны в таблице VIII, из которой видно хорошее согласие между этими величинами.

Таблица VIII

Концентрация продуктов распада радона в модельном помещении /Ки/л/

Воздухообмен К /час <sup>-1</sup> /	К о н ц е н т р а ц и я	
	Расчетная	Измеренная
I	0,08	0,10
1,8	0,021	0,017

Расчет максимально возможной мощности дозы в модельном помещении дал величину 19,8 мкр/час. Для сопоставления этого значения с измеренной дозой необходимо внести 2 поправки: на наличие окон и дверей и на отсутствие слоя насыщения по гамма-излучению /толщина ограждений составляла 20 г/см<sup>2</sup>/. Величины поправок составляют 0,87 и 0,65, соответственно. С учетом их расчетная мощность дозы гамма-излучения в помещении составила 11 мкр/час. Измеренная величина равна 10 мкр/час. Таким образом, наблюдается вполне удовлетворительное согласие.

### Заключение

Нормативы радиоактивности строительных материалов, разработанные на основе проведенных исследований, предназначены для ограничения радиационного воздействия на население за счет этого фактора. Предложенные нормативы не ограничивают использование в жилищном строительстве основной массы традиционных материалов. Строительные материалы, содержание радиоактивных изотопов в которых превышает нормативные величины для жилищного строительства, могут использо-

ваться для других целей. Способ ограничения радиационного воздействия на население при таком использовании указанных материалов требуют дальнейших исследований.

Широкое внедрение нормативов радиоактивности строительных материалов требует разработки оперативных и экспертных методов контроля. В качестве оперативного метода контроля целесообразно использовать измерение мощности дозы в массивах стройматериалов /карьеры, склады, отвалы и пр./ Из сопоставления формул /2/ и /4/ следует, что предложенные нормативы соответствуют мощности дозы в 4-й геометрии 47 мкр/час. Для экспертного контроля перспективно использование высокочувствительных гамма-спектрометров, а при обнаружении содержания радия-226 более 5 пКи/г - измерение эманирования образцов

#### Литература

1. КУЛЬТКВИСТ Б. Монизирующее излучение естественных источников. III, Москва, 1959
2. BERGSTROM STIG OW, WAHLBERG THOR. Radiumhaltiga byggnadsmaterial utstralskyddssynpunkt. Aktiebolaget Atomenergi, Stockholm, AES-7(67)
3. POWDER W.M., COMDON W.J. Measurement of the exposure of human population to environmental radiation. Nature (London) 206, 658, (65)
4. MAYNEORD W.V., HILL C.R. Natural and man-made background radiation. Radiation Dosimetry, 3, 401-451, Academic Press, New York (69)
5. OHLSEN H. Determination of population burden by natural external radiation on the territory of GDR. Kernenergie, 13, 3, 91-96 (70)
6. SPIERS F.W., MCBUGH M.J., APLEBY D.B. Environmental gamma-ray dose to populations; surveys made with a portable meter. Natural Radiation Environment, pp 885-905, The Univ. of Chicago Press 1964
7. CLAY F.A., van GEMERT, WIERSMA. Decrease of primaries showers and ionization of cosmic ray under layers of lead and iron. Physica, 3, 627 - 640, 1936.
8. ШАЛАЕВ И.Л. К вопросу об измерении уровня естественного гамма-фона. Тезисы доклада 4 научно-практической конф. по радиационной гигиене. Л., 1965, 71-79
- 9 Report of the UN Scientific Committee of the effects of atomic radiation; General Assembly Official Records: 17-Session supplement No. 16, (A/5216), New York, 1962.
10. ВИНЮГРАДОВ А.Г. Геохимия редких и рассеянных химических элементов в почве. АН СССР, 1957
11. GALAMBOS S. Die natürliche Radioaktivität der Baustoffe und der Wohnbauten. Wissenschaftliche Zeitschrift der Hochschule für Bauwesen, Leipzig, Heft 12, 91-94, 1966.
12. ROKA O., VAJDA Z., FREDMERSKY T. Radon concentration in the air of buildings. Proceedings of the II Symposium on Health Physics Pecs, Hungary, vol. II, pp. 81-83, 1966.
13. БЫКОВСКИЙ А.В. Гигиенические вопросы при подземной разработке урановых руд. М. Медгиз, 1963
14. GILBERT V.J., KULP J.L. Radon leakage from radioactive minerals. Amer. Mineral., 40, 481-496, 1955.
15. Нормы радиационной безопасности /НРБ-69/. Атомиздат, 1970

## EYE EXPOSURE FROM THORIATED OPTICAL GLASS

Robert C. McMillan and Steven A. Horne

U. S. Army Mobility Equipment Research & Development Center

Fort Belvoir, Virginia 22060

### Abstract

During a routine radiation survey of equipment with some optical components the Health Physicist was surprised to find a large reading on an ionization survey instrument. Significant radiation was also observed on a beta-gamma instrument. The source of the radiation was identified as thorium in high quality optical glass. Similar equipment was examined, but not all items produce readings on the survey instruments.

Thorium is added to the glass, in amounts up to 30% by weight, to provide improved optical properties. Similar results may be obtained by using other heavy elements. Thorium is carried as an impurity with some of these elements and the thorium concentration may be greater than 0.05% by weight.

Since the glass is used in an eyepiece, the amount of exposure to the eye should be investigated. The decay chain of thorium contains alpha, beta, and gamma components. Both the alpha and beta components are largely attenuated by the glass. The alpha particles that reach the eye will be absorbed in a thin surface layer, less than 100 microns, the beta components extend over a larger distance, and the gammas will produce almost constant exposure over the entire eye.

The beta-gamma exposure rate was determined by thermoluminescent dosimetry (TLD). The measured exposure rate was 1 mRem/hr averaged for 123 hours exposure at the surface of a lens which contained 18% thorium by weight.

A computer model for the emission of alphas from the glass, the absorption in air and the final absorption in surface layers of the eye provides a technique for examining the amount of exposure at different depths below the surface of the eye. The results of the model will be compared with experimental results from alpha spectroscopy.

Although the alpha particles come from only a small surface layer of the glass, the absorbed dose rate at the surface of the eye may be 50 to 1000 times greater from alpha than from beta-gamma radiation. The size of the lens, and the eye to lens distance determines the number and energy distribution of the alpha particles reaching the eye.

To reduce exposure to the eye, thin non-thoriated glass shields were inserted between the lens and the eye. The results of TLD and alpha counting with and without the shielding showed a complete removal of the alphas.

### Introduction

Our introduction to thoriated glass came as a request to survey some electro-optical equipment with high internal voltages. There was no anticipation of external radiation, but the survey was conducted and 4-5mR of radiation was detected on an ionization type of survey instrument. The eyepiece of the equipment was removed to eliminate some shielding and to bring the survey instrument closer to the expected source of the radiation. However, the radiation level decreased. This process of elimination led to the optical glass as the source of the radiation. Significant radiation was also observed with beta-gamma instrumentation. Identification of the thorium as the source was accomplished by gamma ray spectroscopy.

After the thoriated glass was identified, similar electro-optical equipment was surveyed. Some of the equipment gave positive readings and others were within the natural background. The specifications of the optics were checked, but there were no requirements for thorium.

Thorium is added to glass, up to 30% by weight, to provide improved optical properties. Specifically, glasses with an index of refraction greater than 1.65 and with the product of Abbe constant and index of refraction greater than 70 are often made with thorium. Other heavy elements may be used to obtain similar results. When some of the lanthanide compounds are used, thorium is often contained as an impurity and the thorium concentration may exceed 0.05% by weight.

The use of thorium in optical glass raises few problems unless the glass is an eyepiece. When thorium is in the eyepiece, the eye is exposed to all of the radiation generated by the thorium decay chain - Alpha, betas and gammas - and the associated bremsstrahlung radiation. Initially we considered only the beta-gamma component.

The alpha exposure presented a special problem. The high concentration of thorium gave a high flux of alpha particles. Harvey<sup>1</sup> raised questions on the external radiation hazards of alpha particles. He evaluated plane sources of alpha activity on the skin surface. Witten and Sulzberger<sup>2</sup> investigated the mode of action of thorium on human skin. They found that the thorium was carried into the epidermis. In contrast to the broad surface contamination and the penetration effects, Dean and Langham<sup>3</sup>



concentrated on the exposure of the skin to particles of high specific activities. None of these studies provided direct data for evaluating eye exposure from externally originating alpha particles.

In the present study we investigate the exposure of the eye's surface from alpha particles emitted by small quantities of thorium uniformly distributed in glass. A model of the emission and absorption processes of alpha radiation will be developed and the exposure at various layers of the eye's surface will be calculated. Comparison of the theoretical values and some experimental data will be included.

#### Thorium Concentrations

Glass manufacturers publish catalogs of glasses. Some of the concentrations approach 30% by weight. The performance characteristics of the optical elements that were in use did not require the thoriated glass. However, it was recognized that some thorium may be included in trace quantities. The uncertainties associated with thorium in glass have led us to set a preliminary level of 0.05% thorium by weight. A survey meter identifies glass with large thorium concentrations, but is ineffective in identification of trace quantities. Gamma ray analysis was not effective on highly thoriated glass to yield the correct percent of thorium. This reflects the lack of secular equilibrium. For trace concentrations, the counting for gamma analysis becomes prohibitive. X-ray fluorescence provides a technique for obtaining the thorium concentration. To be accurate, thorium standards in glass matrices similar to the unknown are required.

#### Beta-Gamma Components

It was recognized very early that the radiation levels observed with the ionization chamber contained a contribution from the alpha particles. This was most easily observed by placing a thin sheet of paper between the glass and the gauge and noting the reduced instrument readings. In an attempt to obtain a better measure of the beta-gamma exposure rates, thermoluminescent dosimeters (TLD) were placed on two lenses. Total exposure time was 24 hours. These initial measurements included some alpha excitation of the phosphors. A second set of measurements was made with a borosilicate flat glass 2.8mm thick. The exposure time was 123 hours. The results of these measurements along with the alpha count rates are shown in Table 1. The radiation that can be assigned to beta-gamma is about 1mR/hr.

Table 1. Glass shielding effects  
Lens area 6.77cm<sup>2</sup>

Lens	Th Concentration	TLD (mR/hr)		Alpha (counts/min)	
		No Shield	Shield	No Shield	Shield
82291	18.1	5.71	0.98	1700	0
86200	18.4	6.45	1.39	2100	0

These lenses have a mass of 27.2 grams. The gamma radiation will be proportional to the mass and thorium concentration. The beta component will be absorbed by the glass. Even the most energetic beta will not penetrate more than 2.5mm of the glass.

### Alpha Radiation

The major portion of the survey instrument reading may be explained by alpha radiation. However, the relationship to absorbed dose does not follow from the instrumental reading. In this section, we will derive the number of alpha particles leaving the glass surface, identify the number reaching various depths within the eye, and the associated absorbed dose rates at these levels. Lens to eye distances, and thorium concentration strongly influence these results.

#### Range of Alpha.

The range-energy relationships for glass and tissue were calculated for specific energies by a computer program using the procedures outlined by Neufeld, et al.<sup>4</sup> The program was checked by comparison with the tables for proton ranges in soft tissue given in the reference. The alpha range in soft tissue was compared with Walsh's<sup>5</sup> results and demonstrates excellent agreement.

The range of alphas in glass is dependent on the glass composition. The chemical composition of a lanthanum glass containing a trace quantity of thorium is shown in Table 2. Small changes in the thorium concentration will have negligible effects on the range of the alpha radiation in the glass.

Table 2. Glass Chemical Composition

Density = 3.64 g/cm <sup>2</sup>				
Element	Percent	Atoms/cm <sup>2</sup>	Atomic No	Atomic Wt
La	35	5.51x10 <sup>21</sup>	57	138.9
CA	35.7	1.95x10 <sup>22</sup>	20	40.08
O	23.2	3.17x10 <sup>22</sup>	8	16
As	3.8	1.11x10 <sup>21</sup>	33	74.9
Zr	2.2	5.29x10 <sup>20</sup>	40	91.2
Th	0.1	9.45x10 <sup>18</sup>	90	232.0

Even large interchanges between the lanthanum and thorium in percent by weight will have only small changes in the range provided the density remains constant. When the density changes, the range may be calculated as follows:

$$R_N = R \left( \frac{3.64}{P_N} \right)$$

where P<sub>N</sub> is the new density and R<sub>N</sub> is the new range.

The values for the range-energy for glass, air and tissue are given in Table 3. These values will be used in the subsequent calculations.

Table 3. Alpha - Range-Energy

Energy (MEV)	Glass ( $\mu\text{m}$ )	Range Air (cm)	Tissue ( $\mu\text{m}$ )
1	.25	.5	4.2
2	1.75	1.0	9.8
3	5.57	1.625	16.4
4	10.25	2.42	25.1
5	15.25	3.5	35.5
6	20.75	4.64	47.2
7	27.25	5.95	61.1
8	34.25	7.34	75.5
9	41.75	8.04	91.75

Alphas emitted from glass.

For the calculations that follow, the lens is considered as a flat circular glass disc with trace quantities of thorium uniformly distributed throughout. Thorium - 232 is taken in secular equilibrium with all of its daughter products. Although gamma analysis raised doubts that equilibrium exists, the use of this assumption will produce a maximum alpha emission.

Although the total number of alpha particles that leave the surface of the glass is important, we will calculate only those that are directed to an element of the eye's surface. Three distances then become important:  $X_a$ , the shortest distance between the eye and the lens;  $X_g$ , the distance the alpha travels in the glass; and the  $X_t$ , the distance in tissue below the surface of the eye.

Let  $N(E)$  be the number of alpha particles emitted per  $\text{cm}^3$  with an energy  $E$ . The number,  $dN_c$ , from an element of volume and directed toward the selected surface element of the eye,  $dA$ , is given by:

$$dN_c = \frac{N(E)dE(2\pi(X_t+X_a)\sin\theta/\cos\theta)((X_t+X_a)/\cos\theta d\theta)\cos\theta dA\cos\theta dX_g}{4\pi((X_t+X_a)/\cos\theta)^2}$$

$$= (N(E)/2)(\sin\theta\cos^2\theta d\theta)dAdX_gdE$$

where  $\theta$  is the angle between the eye to glass normal and the eye to volume element. This equation may be integrated over the volume of the lens and over the energies to give the total number emitted directed toward the surface element of the eye. All of the alphas emitted will not reach the eye. Absorption of energy begins in the glass, with no alphas originating at depths greater than  $40\mu\text{m}$  every reaching the surface. Additional absorption

occurs in the air and finally in the surface layers of the eye. The energy distribution of the initial alphas is known, but as absorption occurs, the energy spectrum changes.

A computer program was written to perform the integrations and calculate the rate and energies of alphas reaching various layers of the eye from different sizes of lenses. This program also calculates the absorbed dose rates at each of the layers.

Results

Table 4 summarizes the output for the model of a lens 3cm in diameter containing 0.005% thorium.

Table 4. Eye Exposures from 3.0cm  
Diameter Lens with 0.005% Thorium

Eye Penetration $\mu\text{m}$	0.1cm Eye to Lens		3.0cm Eye to Lens	
	Alpha Count $\text{cm}^{-2}\text{hr}^{-1}$	Absorbed dose rate $\mu\text{ rad hr}^{-1}$	Alpha Count $\text{cm}^{-2}\text{hr}^{-1}$	Absorbed dose rate $\mu\text{ rad hr}^{-1}$
0.	5.52	155.62	.612	16.220
5.	5.03	150.50	.443	11.632
10.	4.14	124.97	.320	8.480
15.	3.25	97.44	.223	5.888
20.	2.48	74.34	.158	4.173
25.	1.84	55.95	.114	3.006
30.	1.32	39.97	.089	2.577
35.	.91	27.30	.069	2.144
40.	.62	18.43	.043	1.473
45.	.41	12.29	.011	.376
50.	.29	8.46	0.0	0.0
55.	.20	6.05	0.0	0.0
60.	.14	4.42		

The 3.0cm eye to lens distance is typical of the operation of some of the systems we investigated. The 0.1cm data are included to obtain some indication of radiation levels at the surface of the glass.

The energy spectrum at the surface of the eye changes with the eye to lens distance. The peak of the energy occurs around 4 Mev and 0.5 Mev for the 0.5cm and 3.0cm distances respectively.

## Discussion

The 1700 alpha counts/min for the lens in Table 1 may be compared with surface count rate in Table 4 by proportions of percentages and correction for area. For the first lens the calculation is as follows:

$$\frac{(1700 \text{ counts/min}) (.005\%) (60 \text{ min/hr})}{(18.1\%) (6.77 \text{ cm}^2)} = 4.76 \text{ counts hr}^{-1} \text{ cm}^{-2}$$

In a similar way, the second lens yields 5.06 counts hr<sup>-1</sup>cm<sup>-2</sup>. Both of these numbers are lower than that given in Table 4. The alpha survey instrument used to measure the count rate has a 1.5mg/cm<sup>2</sup> window. This window will be effective in shielding low energy alphas from the detector. The agreement between the measured and computed values is excellent considering the window thickness and the differences in concentration.

About half of all incoming alpha particles are stopped and two-thirds of the energy is deposited in the first 15μm of the surface. This includes the tear layer (7μm) and part of the epithelium. The first mitotic layers occur about 45μm below the surface. Even with the lens at the surface of the eye, only one particle will reach this depth every two hours for each cm<sup>2</sup> of surface.

All of the data was reported for 0.005% thorium. The data may be multiplied by 10 to obtain the results for 0.05% or other appropriate factors to obtain values for other thorium concentrations. Smaller diameter lenses will reduce the alpha flux, but not in a simple relation to area.

In comparison with the alpha absorbed dose, using the data from Table 1, the beta-gamma component is 0.3μ rad when corrected to .005% thorium. At the surface, this represents a factor of 100-1000 less. At the first mitotic cells the two dose rates are about the same.

## Summary

Over 90% of the alpha radiation is absorbed before it reaches the first mitotic layer of the eye. From the data presented, the absorbed dose in the mitotic layer may be calculated and integrated over exposure times. No attempt has been made to relate these values to exposure criteria.

## References

1. J.R. Harvey, Health Physics, 21 (1971) pp. 866-869.
2. V. Witten, M. Sulzberger, American Journal of Roengenology, 74 (1955) pp. 90-97.
3. P.N. Dean and W.H. Langham, Health Physics, 16 (1969) pp 79-84.
4. J. Neufeld, L.C. Emerson, F.J. Davis, and J.E. Turner, Principles of Radiation Protection, Chapter 2, K.Z. Morgan and J.E. Turner. Editors, John Wiley & Sons (1968).
5. P.J. Walsh, Health Physics, 19 (Aug 70) pp. 312-316.

DOSIMETRIE DES RAYONNEMENTS COSMIQUES A BORD DU TRANSPORT SUPERSONIQUE  
CONCORDE

H. FRANÇOIS (1), R.P. DELAHAYE (2), P. SIMON (3), G. PORTAL (1)  
H. KAISER (4) et P. DURNEY (5)

Abstracts

Dosimetry of radiation on-board the French prototype supersonic transport Concorde 001 was performed during the aircraft's test flights from its Toulouse base.

The supersonic transport medical subcommittee of the General Secretariat for Civil Aviation directed these radiation measurement activities. Actual dosimetric operations were conducted by different specialized laboratories employing the following systems and techniques :

I - "Passive" integrator dosimetric systems

- a/ Nuclear emulsions in which each event is individually analyzed.
- b/ Stacks of nuclear emulsions of varying sensitivity which permit obtaining ionizing-particle distribution as a function of their TEL.
- c/ Radiothermoluminescent dosimeters.
- d/ Fast-neutron dosimeters.

2 - Dose-rate recording systems.

The authors analyze, compare, and comment upon results obtained. In addition, the dosimetric problems posed by possible solar flares and their effects upon flight plans, are discussed.

Conclusions are drawn relative to the possible dosimetry techniques to be employed in identifying radiation protection problems on-board the SST.

Introduction

Les avions qui évoluent à des altitudes supérieures à 15 000 mètres sont soumis à une irradiation naturelle différente de celle habituellement observée dans les couches basses de l'atmosphère.

La très prochaine mise en service des avions de transports supersoniques commerciaux (T.S.S.) accroîtra le nombre des personnes exposées car l'efficacité de l'écran que forme l'atmosphère terrestre aux particules du rayonnement cosmique est très diminuée à l'altitude de croisière de ces appareils.

Ce problème radiobiologique se présente sous deux aspects : d'une part il faut connaître le rayonnement cosmique permanent auquel l'avion est soumis dans les circonstances normales, d'autre part, il faut pouvoir détecter sans délai les irradiations anormales consécutives à certaines éruptions solaires. Ces dernières indications doivent pouvoir être exploitées très rapidement par les équipages.

Nous résumerons dans la première partie de cet exposé les connaissances acquises sur le rayonnement cosmique et nous exposerons ensuite quelles sont les

- 
- (1) H.FRANÇOIS, G.PORTAL, Service Technique d'Etudes de Protection et de Pollution Atmosphérique - Section de Dosimétrie Physique - Centre d'Etudes Nucléaires de Fontenay-aux-Roses - France -
  - (2) R.P.DELAHAYE, Professeur du Service de Santé des Armées, Hôpital Bégin, 94 Saint-Mandé - France -
  - (3) P.SIMON, Service des Ursigrammes - Observatoire de Meudon - 92 Meudon-France
  - (4) H.KAISER, Laboratoire de Physique Corpusculaire du Centre d'Etudes Nucléaires de Strasbourg - France -
  - (5) P.DURNEY, Service Mixte de Sécurité Radiologique - Montlhéry - France -

mesures dosimétriques que nous avons entreprises et réalisées à bord du prototype CONCORDE 001.

#### I - Composition physique du rayonnement cosmique à l'altitude de croisière de Concorde

Le rayonnement cosmique qui atteint l'atmosphère terrestre peut avoir deux origines : l'une galactique, l'autre solaire.

##### A/ Rayonnement cosmique galactique

Ce rayonnement est constitué de particules de grande énergie :

- 80 à 85 % de protons
- 15 à 19 % d'hélium

et des particules lourdes Fe, Mg, C, O.

Lorsqu'ils pénètrent dans l'atmosphère les noyaux sont désintégrés au cours de réactions nucléaires avec l'oxygène et l'azote de l'air et il apparaît des particules secondaires : neutrons, mesons, hyperons. Le nombre des particules secondaires augmente progressivement, passe par un maximum vers 18 000 - 20 000 mètres d'altitude et diminue ensuite du fait de l'absorption atmosphérique.

Les particules du rayonnement cosmique galactique sont soumises à la distribution et aux variations du champ magnétique dans le système solaire. Leur intensité varie à la surface de la terre selon le temps, la latitude et l'altitude

##### Variations dans le temps

Lorsque les zones actives du soleil émettent des nuages de plasma, on observe des réductions de l'intensité du rayonnement cosmique galactique. Ce sont les réductions de FORBUSH.

##### Variations avec la latitude

Le champ magnétique de la terre empêche les particules chargées de franchir les lignes de forces sous une inclinaison supérieure à un angle donné.

L'intensité du rayonnement cosmique galactique est maximale au pôle magnétique et minimale à l'équateur géomagnétique.

##### Variations avec l'altitude

Celles-ci sont consécutives aux épaisseurs d'atmosphère traversées.

##### B/ Rayonnement cosmique solaire

Une éruption solaire se manifeste en fait par une augmentation de la brillance d'une certaine surface de la chromosphère. La dimension et l'intensité du phénomène définissent assez bien l'importance de l'éruption solaire. On pourrait également considérer sa durée.

La fréquence des éruptions est liée à l'activité solaire qui est un phénomène cyclique. Les deux derniers maximums ont eu lieu en 1957 et en 1968 et les minimums en 1954 et 1964.

En juillet 1970 on a observé 500 éruptions, tandis qu'il n'y en eut que 3 en 1964 qui eurent toutes lieu le même jour.

Dans certains cas, les éruptions solaires provoquent des phénomènes radioactifs qui se produisent au voisinage de la chromosphère et qui s'étendent dans la couronne solaire. Ces éruptions s'accompagnent d'émission de particules, protons et électrons, qui sont accélérées et portées à des énergies considérables (de l'ordre du GeV).

Une émission de rayons X de quelques dizaines de keV se propage alors dans le milieu interplanétaire.

Pour l'habitant de la terre, les manifestations des éruptions solaires sont rares. Les émissions de rayons X se produisent lors d'une éruption sur 200 et les arrivées de particules ont lieu pour une éruption sur 1000. Ceci est dû à l'action du champ magnétique et de l'atmosphère terrestre. Le rayonnement X est absorbé par les couches les moins denses de l'atmosphère (vers 80 000 mètres) et

il ne pénètre pas au-dessous de 30 000 mètres. Ce sont les ions créés qui perturbent les ondes radioélectriques.

Quant aux particules chargées, leur pénétration augmente avec la latitude ; seules celles qui ont une énergie très élevée (quelques GeV) peuvent atteindre la surface de la terre aux latitudes moyennes et équatoriales.

Aux altitudes de croisière des avions supersoniques, la protection n'étant pas suffisante, ces événements doivent être pris en considération.

La prévision et l'identification des événements solaires est faite en permanence par des équipes travaillant en collaboration internationale.

De 1966 à 1968, six à huit événements auraient été détectables à l'altitude de vol du Concorde soit 3 à 4 pour 10 000 éruptions.

## II - Moyens dosimétriques utilisés

La complexité du problème dosimétrique posé et sa nouveauté ont conduit les responsables de la sous-commission médicale "Concorde" à faire appel aux spécialistes de différents laboratoires qui dans une large concertation scientifique et technique utilisèrent les avantages complémentaires de l'expérience acquise, du savoir-faire et des équipements disponibles.

Deux types de dosimètres ont été utilisés. Une première catégorie que nous appelons des dosimètres "passifs" qui intègrent et gardent en mémoire les événements et une autre catégorie qui s'apparente aux appareils de mesure et qui donne à chaque instant du vol les indications sur l'intensité du rayonnement.

### I/ Dosimètres passifs

#### a/ Ensemble dosimétrique pour les particules chargées et les ions lourds

Il est constitué par des émulsions nucléaires et par des matières plastiques qui permettent de déterminer les doses dues aux particules chargées.

Le système dosimétrique maintient à l'abri de la lumière un empilement constitué d'une émulsion nucléaire Ilford K5 d'une épaisseur de 100 micromètres et 5 pellicules de nitrate de cellulose de 200 micromètres d'épaisseur.

Dans le type d'émulsion nucléaire utilisé toutes les particules chargées qui composent le rayonnement cosmique, électrons, mésons, mésons chargés, protons, particules  $\alpha$ , ions lourds même s'ils sont au minimum d'ionisation, sont enregistrés et laissent une trace après développement.

Les cinq pellicules de nitrate de cellulose servent ensuite à déterminer le flux des ions lourds.

#### b/ Ensemble dosimétrique pour les neutrons

Nous avons utilisé le dosimètre photographique CEA/STEPPA contenant une émulsion nucléaire NTA pour déterminer la "dose neutron" due au rayonnement cosmique. Sept de ces dosimètres sont placés sur les sangles des parachutes des équipages.

Deux dosimètres témoins sont conservés au sol.

Le dispositif complet est renouvelé chaque mois.

#### c/ Ensemble dosimétrique pour les champs électromagnétiques et les particules ionisantes

Dans une première approche du problème qui nous était posé, nous avons essayé de mesurer la dose absorbée due à la composante électromagnétique du rayonnement cosmique.

Nous avons utilisé des dosimètres radiothermoluminescents au fluorure de lithium et au sulfate de calcium et nous avons également embarqué des empilements d'émulsions nucléaires à sensibilité variable afin de déterminer le spectre de transfert linéique d'énergie des particules ionisantes.

Nous utilisons les émulsions :

- K5 : pour l'enregistrement de toutes les particules ionisantes,
- K2 : pour l'enregistrement des particules dont le TLE est supérieur



à 1 keV par micromètre (dans l'eau)

KI : pour l'enregistrement des particules dont le TLE est supérieur à 6 keV par micromètre

KO : pour l'enregistrement des particules dont le TLE est supérieur à 8 keV par micromètre.

Ce système avait déjà été étudié et est utilisé au C.E.A. pour la dosimétrie auprès des grands accélérateurs (1). Il a fallu l'adapter au problème posé du fait de la faiblesse du niveau des doses enregistrées.

Nous avons dû étudier une méthode d'effacement du bruit de fond des plaques nucléaires. Cette méthode a été récemment mise au point et les essais effectués auprès de l'accélérateur Sturne ont donné pleine satisfaction.

La filtration totale correspondant aux divers emballages et supports des émulsions est de 450 mg/cm<sup>2</sup> environ.

## 2/ Dosimétrie par enregistrement des débits de dose

### a/ Systèmes d'enregistrement du débit de dose dû aux neutrons et aux rayonnements gamma - VAMEGA

On a utilisé un matériel appelé VAMEGA déjà employé depuis plusieurs années sur les avions subsoniques longs courriers. Ce dispositif a été modifié pour assurer la mesure des neutrons. Il comprend un détecteur gamma qui est un tube Geiger-Muller, associé à un détecteur à hélium 3 entouré d'un modérateur en polyéthylène.

### b/ Détecteur de bord AWRE installé sur tous les avions Concorde

Ce détecteur a été conçu par l'United Kingdom Atomic Energy Authority, Atomic Weapons Research Establishment (AWRE) à Aldermaston (2). Il indique le débit de dose en mrem/h sur une échelle logarithmique à 4 décades allant de 0 à 1 000 mrem/h. Il comporte également un affichage digital de la dose cumulée en mrem. Trois tubes Geiger-Muller permettent de mesurer la dose due aux particules chargées et aux gamma, un facteur de qualité égal à 1,5 étant automatiquement appliqué ; un compteur proportionnel au trifluorure de bore avec modérateur en polyéthylène permet de mesurer la dose due aux neutrons en utilisant un facteur de qualité égal à 10. Après traitement, les signaux des 2 systèmes de détection entrent dans une voie de comptage unique. L'échelle de l'indicateur est divisée en zones "normale" "alerte" et "action" ; le niveau d'action étant relié au système d'alarme central de l'avion. La définition de ces 3 zones est en cours d'étude. Sur l'avion prototype, le signal de sortie du détecteur de bord est également enregistré sur l'équipement central d'enregistrement d'essais en vol (QS system) de façon que l'on puisse faire des lectures de durée de vol et de débits de dose à des altitudes sélectionnées.

## III - Résultats et discussion

### I/ Résultats obtenus avec les ensembles dosimétriques constitués par les émulsions nucléaires K5 et les plaques de nitrate de cellulose

Dès le retour au laboratoire des dispositifs d'exposition à la suite d'une expérimentation d'un mois, les émulsions nucléaires Ilford K5 de 1 000 micromètres d'épaisseur sont exposées à la lumière blanche parallèle passant à travers le négatif d'une grille millimétrique codée. La grille impressionnée sur une des faces de la pellicule d'émulsion nucléaire détermine un système d'axes de référence qui permet de faire toujours des mesures dans les mêmes conditions et dans des volumes d'un mm<sup>3</sup>. Les traces dues aux particules chargées du rayonnement cosmique, observables dans l'émulsion nucléaire, sont soit granulaires, soit continues. Ces phénomènes sont fonction de l'énergie et de la charge des particules.

En tenant compte de l'épaisseur avant développement et de la grille, on mesure, sous microscope, la longueur en projection des traces traversant un volume d'un mm<sup>3</sup> d'émulsion nucléaire ou s'y arrêtant, les coordonnées des points d'entrée et de sortie ou d'arrêt des traces ainsi que l'épaisseur de l'émulsion

nucléaire après développement dans la zone du volume considéré. Ces mesures sont effectuées dans 10 volumes d'un  $\text{mm}^3$  présélectionnés par la grille sur chaque émulsion nucléaire contenue dans un dispositif d'exposition. Les longueurs en micromètres sont calculées à partir de ces mesures. Les traces se trouvant dans le volume d'un  $\text{mm}^3$  d'émulsion nucléaire correspondent à des particules présentant une perte spécifique d'énergie par unité de longueur variable ou non le long de leur trajectoire. En considérant des pertes spécifiques d'énergie par micromètre, la sommation de ces grandeurs sur toute la longueur de toutes les traces nous donne l'énergie totale "déposée" dans le volume d'un  $\text{mm}^3$ . La perte d'énergie par unité de longueur peut être déterminée expérimentalement par des mesures de granularité ou des mesures photométriques. Ces travaux ont déjà été faits dans le cas des émulsions nucléaires exposées à bord des cabines spatiales Apollo.

Mais le temps nécessaire à ces mesures est très grand et incompatible avec un dépouillement mensuel. De plus, dès le début de cette exploitation, on a constaté que le nombre de traces continues est de l'ordre de 10 p. cent par rapport au nombre total et que le nombre des ions lourds est nul. Cette constatation a conduit à employer une méthode plus rapide.

En partant des fins de traces de protons d'accélérateurs, d'énergie initiale de 150 MeV, enregistrées dans l'émulsion nucléaire Ilford K5, on a déterminé le parcours résiduel pour lequel les traces de protons passent de l'état granulaire à l'état continu. Il est de l'ordre de 70 micromètres, ce qui correspond à une énergie du proton de 3 MeV et une perte spécifique d'énergie de 24,2 keV par micromètre. Rappelons que la perte spécifique d'énergie des protons au minimum d'ionisation est de 0,549 keV par micromètre.

En multipliant la somme des longueurs de toutes les traces contenues dans le volume d'un  $\text{mm}^3$  par la valeur 24,2 keV par micromètre, nous obtenons la limite supérieure de l'énergie totale "déposée". Un volume d'un  $\text{mm}^3$  d'émulsion nucléaire a une masse de  $3,8 \cdot 10^{-3}$  g. Par définition, un rad correspond à une énergie de  $6,24 \cdot 10^{13}$  eV "déposée" dans un gramme de matière. La dose en millirad est donc égale à :

$$\frac{\text{Energie totale "déposée"} \times 10^3 \times 10^3}{3,8 \times 6,24 \times 10^{13}}$$

Les doses maximales dues aux particules chargées calculées selon cette méthode sont données dans le tableau I. Ces valeurs sont les moyennes obtenues à partir des 10 volumes d'un  $\text{mm}^3$  examinés dans chaque émulsion nucléaire d'un dispositif d'exposition. Ce tableau indique le nombre d'heures de vol au-dessus de 12 000 mètres d'altitude.

Il est important de noter que sur l'ensemble des émulsions exploitées de 1970 à 1973, nous n'avons jamais repéré de trace d'ion lourd.

## 2/ Résultats obtenus avec les dosimètres à émulsion NTA

Par l'intermédiaire de la même grille millimétrique, on détermine le nombre moyen des traces contenues dans 10 surfaces élémentaires d'un  $\text{mm}^2$ .

Pour ces films et pour les neutrons d'énergie comprise approximativement entre 1 et 10 MeV, on admet d'après les résultats expérimentaux que 127 000 traces par  $\text{cm}^2$  correspondent à 1 rad ; 1,27 trace/ $\text{mm}^2$  correspond donc à 1 millirad. Il n'est toutefois pas possible de déterminer la dose due aux neutrons du rayonnement cosmique à l'aide de cette équivalence. En effet, les résultats expérimentaux ont été obtenus avec des flux de neutrons purs, alors que les neutrons ne constituent qu'une partie du rayonnement cosmique. L'émulsion NTA utilisée n'a que 40 micromètres d'épaisseur. L'examen détaillé de l'émulsion nucléaire K5 de 1 000 micromètres d'épaisseur a permis de déterminer que 90 p. cent des traces sont des traces de particules chargées et non des protons de recul. Il y a donc lieu de considérer que 0,127 traces par  $\text{mm}^2$  correspond à 1 millirad. Le tableau (I) donne les doses en millirads dues aux neutrons dans le poste de pilotage.

### 3/ Résultats obtenus à l'aide de l'ensemble dosimétrique pour les champs électromagnétiques et les particules ionisantes

Trois ensembles dosimétriques sont placés à bord de l'avion :

- un à l'avant,
- un autre au centre,
- le troisième à l'arrière.

#### a/ Dosimètres radiothermoluminescents

Le tableau de la figure n° 5 donne des exemples de résultats de mesures effectuées à l'aide des dosimètres au fluorure de lithium (3).

On constatera que les doses mesurées sont très faibles et très proches de la limite inférieure de détection. La durée des vols à haute altitude a été insuffisante pendant nos essais ; il s'ensuit que la part de l'irradiation subie lors des vols est très faible par rapport à celle reçue au sol pendant la période d'intégration (7 semaines environ). Nous estimons que la marge d'erreur de ces mesures est de ce fait assez grande (coefficient de variation égal à 40 p. cent).

Pour diminuer celle-ci, nous avons décidé d'utiliser lors des vols du second semestre 1972, un produit nettement plus sensible, le sulfate de calcium activé au dysprosium. Ce dernier est en effet trente fois plus sensible que le fluorure de lithium et, bien que n'étant pas "équivalent aux tissus mous", il nous a donné d'excellents résultats lors des mesures effectuées sur des vols de ballons (4) pour le compte du Groupe Européen de Biophysique Spatiale. En effet, aux altitudes où sont effectuées ces expériences, la contribution des rayonnements électromagnétiques de faible énergie est tout à fait négligeable. Quant à la sensibilité aux particules ionisantes, elle ne diffère guère de celle du fluorure de lithium (5). Nous espérons obtenir des résultats plus précis à l'aide de ce matériau (le coefficient de variation des mesures obtenues est égal à environ 10 p. cent). Si l'on désire une précision encore meilleure, il est nécessaire soit de faire les mesures immédiatement après un vol prolongé, soit d'attendre que la fréquence et la durée des vols soient suffisantes.

#### b/ Emulsions nucléaires à sensibilité variable

Le classement des particules en fonction de leur TLE est effectué d'après les caractéristiques des émulsions (seuil de sensibilité) et l'aspect des traces. A titre d'exemple on a admis que sur une émulsion K5, les traces correspondent à un TLE inférieur ou égal à 0,5 keV par micromètre dans l'eau, les traces denses présentent un TLE supérieur. D'autre part les protons de recul provenant des interactions (n,p) engendrés dans l'émulsion, ne sont pas pris en compte.

Il est évident que ces critères sont quelque peu subjectifs et que les spectres de TLE obtenus doivent être considérés comme une simple approche du problème. Cela est d'autant plus vrai que la précision des mesures est altérée par le fait que les doses reçues au sol ne sont pas négligeables par rapport à celles reçues en vol.

Sur le tableau de la figure n° 6 nous avons reporté, à titre d'exemple, les résultats obtenus à l'aide d'un empilement d'émulsions placé à l'arrière de l'avion au mois d'avril 1972. Les valeurs mentionnées correspondent aux seules irradiations subies pendant les vols, la contribution du rayonnement cosmique et tellurique au niveau du sol a été retranchée.

Nous avons calculé la valeur de la dose absorbée à la surface du corps humain, à partir des données de ZERBY et KINNEY (6) valables dans le cas d'un faisceau isotrope de protons qui frapperait une seule face du corps humain. Il convient de préciser que dans ce cas, la dose absorbée superficielle est très voisine de la dose absorbée au point ou l'ionisation est maximale (7).

On remarquera, pour cet empilement, une nette prédominance des particules à très faible TLE (TLE 0,5 keV/micromètre). Il n'en est pas toujours ainsi ; ces proportions varient selon les conditions des vols et les emplacements dans la cellule de l'avion.

Par contre, si l'on exclut les traces de protons de recul provenant des interactions des neutrons avec les matériaux hydrogénés, on ne dénombre pas de particule de TLE supérieur à 6,2 keV par micromètre. Il est vrai qu'à l'altitude de vol du Concorde il semble que les ions lourds aient déjà été absorbés dans l'atmosphère.

Il est possible à partir de ces spectres de calculer un facteur de qualité moyen pour les rayonnements ionisants. Compte tenu des données de NEUFELD, SNYDER et TURNER (7) celui-ci est légèrement inférieur à 1,3.

Ce résultat est donné à titre d'exemple, les expérimentations ne sont pas encore assez avancées pour que l'on puisse faire une synthèse valable des résultats en fonction des conditions des vols et des emplacements dans la cellule.

#### 4/ Résultats obtenus avec les appareils de détection et d'enregistrement des débits de dose VAMEGA et AWRE

Pour le tableau n° 7 nous avons reporté les résultats maximaux moyens des mesures obtenues à l'aide de l'appareil VAMEGA lors d'un groupe de 18 vols au-dessus de l'Atlantique Nord et une série de 11 vols effectués lors de la tournée en Amérique du Sud.

On remarque que les résultats de la seconde série sont, du fait de la différence de latitude, nettement inférieurs à ceux de la première.

Les résultats obtenus à l'aide de l'appareil AWRE sont comparables. A titre d'exemple, lors de la mission du 21.7.71 effectuée au-dessus de l'Atlantique Nord, les débits de dose équivalente mesurés sont respectivement :

- Neutrons	0,20 mrem/h
- Radiations ionisantes et électromagnétiques	0,66 mrem/h
Total .....	0,86 mrem/h

En aucun cas on n'a observé une activité solaire anormale.

Si en cours d'exploitation normale le cas se présentait, l'altitude de vol de l'avion serait modifiée selon des critères qui sont actuellement étudiés par la Commission Médicale Franco-Britannique.

En fait, ce cas devrait se présenter assez rarement. De 1966 à 1968, huit événements seulement auraient été enregistrés à l'altitude de croisière.

Pendant le cycle solaire le plus actif jamais observé entre 1954 et 1964, on trouve 38 événements détectables sous forme d'irradiation à l'altitude de Concorde. Pour le cycle actuel, à peu près moitié moins actif, on arrivera sans doute à une vingtaine d'événements, probablement tous de faible intensité. En fait, dans le cycle précédent, dix événements seulement auraient provoqué une irradiation à un débit de dose supérieur à 1 millirem par heure. Pour le cycle actuel, probablement aucun n'atteindra cette valeur d'ici à 1974 ou 1975. Qu'en sera-t-il ensuite ? On a peu d'éléments pour effectuer une prévision : l'accélération de particules à des énergies aussi élevées et le fait que la terre se trouve sur leur trajectoire forment des conditions assez exceptionnelles observées un nombre de fois trop restreint pour qu'une statistique valable puisse être utilisée. On dispose cependant de deux indications :

- 1 - Pendant un cycle solaire, le nombre et l'énergie de ces événements semblent en relation avec l'activité maximale atteinte au cours de ce cycle.
- 2 - On possède des indications positives qui montrent que les prochains cycles solaires vont encore, dans les cinquante années à venir, décroître en intensité.

### Conclusions

Les doses reçues à l'altitude de croisière du T.S.S. Concorde sont faibles; nous n'avons jamais détecté d'ion lourd.

Le problème des éruptions solaires a retenu notre attention. Leur fréquence est relativement faible ; on en aurait détecté 8 au maximum de 1966 à 1968 à l'altitude considérée. Les appareils d'alarme placés à bord des prototypes n'en ont jamais décelé pendant les essais.

### Note

Ces travaux de dosimétrie ont été réalisés sous les auspices de la Commission Médicale du Transport Supersonique du Ministère des Transports Publics, présidée par Monsieur le Médecin Général Inspecteur RABOUTET et de la Sous-Commission Médicale Française du Groupe Aéromédical CONCORDE.

Les dispositifs dosimétriques ont été installés dans l'avion avec les conseils du Commandant TURCAT et de Messieurs JOATTON et DESTARAC de la S.N.I.A.S.

Figure 1

	Nbre d'heures de vol > 12000 m	Part. chargées mrad	Neutrons mrad
Janvier 1972	7h 38	2,4	0,2
Février "	8h 31	1,1	0,2
Mars "	0h 45	0,1	0,1
Avril "	0h 22	0,3	0,1
Mai "	0h 22	0,3	0,1

Résultat des doses absorbées mesurées à l'aide d'émulsions nucléaires de I000  $\mu$ - Poste de pilotage

Figure 2

Mois	Heures de vol au-dessus de 12 000 m	Doses reçues pendant les vols (millirad)		
		Avant	Milieu	Arrière
Janvier 1972	7h 38	3	3	3
Février "	8h 31	3	3	3
Avril "	0h 22	2	2	2
Mai "	3h 38	2	2	1,5
Octobre "		0,8	0,8	0,8
Novembre "		2	2	2

Dose absorbée pendant la durée des vols mesurée à l'aide de dosimètres radiothermoluminescents

( L'irradiation reçue en dehors des vols a été retranchée )

Figure 3

Intervalle de TLE keV//m	Densité de traces - traces cm <sup>-2</sup>	Dose absorbée superficielle à une fluence unitaire	Dose absorbée superficielle	Contribution relative à la dose
TLE ≤ 0,5	3950	1,6 10 <sup>-7</sup> rad	0,63 10 <sup>-3</sup> rad	73 p.cent
0,5 < TLE ≤ 1	450	3,1 10 <sup>-7</sup> rad	0,14 10 <sup>-3</sup> rad	16 p.cent
1 < TLE ≤ 2,6	80	4,3 10 <sup>-7</sup> rad	0,03 10 <sup>-3</sup> rad	4 p.cent
2,6 < TLE ≤ 6,2	6	10 10 <sup>-7</sup> rad	0,06 10 <sup>-3</sup> rad	7 p.cent
6,2 < TLE ≤ 8,1	0		0	0 p.cent
8,1 < TLE	0		0	0 p.cent
Spectre total			0,86 10 <sup>-3</sup> rad	

Résultat de l'empilement d'émulsions nucléaires du mois d'avril 1972 placé à l'arrière de la cellule

Figure 4

Série de Missions	Débit de dose équivalente dû aux neutrons	Débit de dose équivalente dû aux neutrons $\gamma^+$ + particules ionisantes mrem/h	Total mrem/h
Atlantique Nord	0,31	0,58	0,89
Amérique du Sud	0,13	0,34	0,47

Débit de dose équivalente moyenne maximum mesuré à l'aide de l'appareil VAMEGA lors de deux séries de missions

## Bibliographie

- 1 - G. PORTAL  
Premier Colloque Intern. radioprotection auprès des grands accélérateurs  
Orsay - Saclay janvier 1972 - Presses Universitaires de France, p.89
- 2 - E.W. FULLER - B. DAY  
Internat. Congress on protection against accelerators and space radiation,  
Genève April 1971
- 3 - G. PORTAL - F. BERMAN  
Proceeding of the second congress of the European Association of Radiology,  
Amsterdam juin 1971
- 4 - G. PORTAL  
Note CEA n° I586, décembre 1972
- 5 - C.L. WINGATE - E. TOCHILIN - N. GOLDSTEIN  
USNRDL-TR-909, septembre 1965
- 6 - C.D. ZERBY - W.E. KINNEY  
ORNL-TM-1D38, May 1965
- 7 - J. NEUFELD - W.S. SNYDER - J.E. TURNER - H. WRIGHT  
Health Physics 12, 227, 1966



## THE APPROACH TO RADON PROBLEMS IN NON-URANIUM MINES IN SWEDEN

Jan Olof Snihs  
National Institute of Radiation Protection  
Stockholm, Sweden

### Abstract

In Sweden there are about 60 underground mines. These are for the mining of ferrous and sulphide ores and none are for uranium. Radon measurements were made in all of these mines during 1969 and 1970. By using an assumed equilibrium ratio of 0.5 between radon daughters and radon, the radon daughter levels were calculated to exceed 30 pCi/l (0.3 WL) in 22 mines with more than 1000 underground employees and in a few mines the levels were estimated to exceed 100 pCi/l (1 WL). Hence, non-uranium miners were the largest group of Swedish workers receiving significant radiation doses. However, these workers are not legally classified as radiological workers.

The results initiated more detailed measurements and research. The sources of radon in mines are found to be radon-rich water, ventilation air from abandoned areas with caved materials and, to a lesser extent, minerals particularly rich in radium. In some mines the radon levels vary with the season with a maximum in the summer. No simple correlation to the type of minerals or their geological properties has yet been found.

Instructions on protective measures against radon in mines were issued in March 1972. The regulations are based on a maximum of 30 pCi/l (0.3 WL) of radon daughters as the average over a year. There are also regulations on the maximum delay period for preventative action, on ventilation, on respirators, on measurements and control and on medical examinations.

Epidemiological studies on the lung cancer frequency among miners have been made. The period investigated was the years 1961-1968. A significant excess of lung cancer has been found.

### Introduction

The first measurements on radon in a Swedish mine (Boliden) were made at the beginning of the 1950s. The results of these early measurements and the limited experience of radon problems in non-uranium mines at that time did not give any reason for anticipating high radon levels in Swedish mines. However, improved measuring techniques, continuous progress in radiation protection generally and internationally observed radon problems in many uranium mines resulted in new measurements being initiated at the end of the 1960s.

The first new results (from Näsleden and Långsele) indicated the possibility that radon problems might exist in Swedish mines even though they are not uranium mines. A few additional measurements (in Zinkgruvan and Danne-mora) also proved that there is no simple relation between the radon concentration in a mine and the geology of the mine; the first approach, selecting mines

of interest solely from geological considerations, had to be abandoned. In 1969 it was therefore decided to make a rough but rapid survey of the radon levels in all the mines in Sweden (about 60).

Since 1970 great efforts have been made to develop appropriate sampling and measuring techniques, and to initiate and carry out research on existing problems. As radon in mines was a new and completely unknown occupational hygiene problem for most of those concerned, much effort was put into providing appropriate information. Very shortly, it also appeared necessary to prepare special instructions for radiation protection in mines and these were issued in March 1972. Because of the relatively high radon concentrations found in many mines, it was considered necessary to consider the possibility of an excessive incidence of lung cancer among Swedish miners. An epidemiological study on lung cancer was therefore started in 1971. This report is meant to be a summary of some of the results of the work done on the problem of radon in Swedish mines.

#### Principles of sampling and measuring techniques

The measurements in Swedish mines have mainly been made on radon although they have been supplemented by measurements on radon daughters. Radon measurements are performed by taking air samples in evacuated 4.8 l commercial propane containers which are opened in the mine. The sample is transferred to an evacuated 18 l ionization-chamber via a drying agent. The lower limit for these chambers is about 0.5 pCi/l with a fresh 4.8 l sample. The samples are measured above ground, either in a field laboratory at the mine or at the National Institute of Radiation Protection (NIRP) in Stockholm. In the latter case, the sampling can be done by the mining staff and the containers sent to Stockholm by mail.

Radon daughters are sampled and measured in the conventional way, using glassfibre filters. In evaluation of the radon daughter concentration, the method of Kusnetz<sup>1</sup> is used. As there is no good Swedish expression for WL the result is expressed in equivalent pCi/l (1 WL is equivalent to 100 pCi/l). Measurements on the glassfibre filters are made above ground in general.

#### The principles of measurements

Three types of measurements have been made, namely: guiding measurements, basic measurements and checking measurements.

##### Guiding measurements

The guiding measurements were performed twice during the period 1969-1970, once during the winter and once during the summer. Three of the evacuated containers described above were sent to each mine and the mining company was asked to take one air sample in return air, one at a working place with "normal" ventilation and one sample in an unventilated drift. The samples were expected to be representative for the average radon levels in the mine, for working places and for potentially high radon levels, respectively.

In answer to questionnaires, information was given by the company about the place and time of sampling, ventilation conditions, presence of running water, adjacent minerals, number of worker etc. All results related to radon only. In estimating the corresponding radon daughter concentration, a 50 % equilibrium ratio was assumed. In accordance with the chosen Swedish terminology, this ratio is referred to as the "dose factor".

### Basic measurements

The basic measurements are made by the laboratory staff during a visit to the mine. The purpose of the measurements is to find the true radon and radon daughter concentrations, the reasons for the activity levels and the best means of decreasing the concentration if necessary. The sampling and measurement are preceded by detailed discussions between the radiological team and the representatives of the employees and the employers about ventilation systems, presence or absence of working places in particular areas, suspected high radon levels, radioactive minerals etc. The choice of appropriate places for sampling is based on these discussions.

### Checking measurements

Checking measurements are initiated by the company itself if there have been major changes in the ventilation, or when there is reason to expect high radon concentrations in new parts of the mine. Normally only radon measurements are necessary to guide possible extra preventative action. Periodic checking measurements are also needed as a consequence of the regulations on checking the exposure of the miners (see below). If the only measurements made are those on radon in air samples sent by mail to the laboratory in Stockholm, the radon daughter concentration is estimated by using the dose factor (or factors) found in the basic measurements if this dose factor is still believed to be adequate.

### Concentrations of radon and radon daughters in the mines

After the guiding radon measurements in 1969 and 1970, the radon daughter exposure level was estimated for all mines. The numbers in parenthesis in Table 1 are the first estimated results. At the time of writing (May 1973) basic measurements have been made in 26 mines, from the guiding measurements most of these were expected to have radon daughter concentrations above 30 pCi/l. As a result of these measurements there have been a few changes in the grouping of the mines and miners as indicated in Table 1 by the 1970 numbers without parentheses. This was the situation in 1970. Since then, countermeasures have been taken in many mines, if not yet in all. A few mines have shut down (not because of radon) and a few new mines have been built and the 1973 numbers in Table 1 reflect the situation as it is believed to be in May 1973. It should be noted, however, that the grouping of the mines and miners is based on the highest representative levels found in the working areas of each mine - or of the parts of one large mine. The numbers of overexposed miners have therefore probably been somewhat overestimated.

Radon daughter concentration pCi/l	Number of mines		Number of miners	
	1970	1973	1970	1973
≤10	(25) 26	26	(1100) 1400	1400
10 - 30	(13) 12	19	(1700) 2000	2700
30 -100	(17) 16	10	(1700) 700	610
100 -300	( 5) 6	0	( 140) 620	0
>300	( 0) 1	0	( 0) 21	0

Table 1. Numbers in parenthesis are those estimated from the guiding measurements 1969-1970, the ones without parentheses from basic measurements since 1969.

Great efforts have been made to reduce the radon daughter levels and this work is still in progress. The countermeasures mainly consist of changes in the ventilation system and often new ventilation shafts have to be built. However, it is expected that within about a year the numbers of minee and miners in the exposure group  $>30$  pCi/l will be reduced considerably.

The measurements in 1969 and 1970 showed that many mines had higher, some considerably higher, radon levels in the summer (May-October) than in the winter. 19 mines with radon daughter levels  $>10$  pCi/l were found to have summer values more than 50 % higher than winter values and on average for all mines the radon concentration was twice as high in the summer than in the winter. The main reason is probably reduced or changed ventilation.

Apart from the general radon and radon daughter values for the mines, it has also been of interest to find the potential high values in unventilated areas or elsewhere in the mine. Up to now 7 mines have been found to have such areas with radon levels exceeding 1000 pCi/l; radon concentrations of 1000-2000 pCi/l have been found in four mines, 6000 pCi/l in one mine, 10,000 pCi/l in one and 20,000 pCi/l in one. There are also a few mines with radon levels of 300-1000 pCi/l. In some of these areas with high radon levels, mining work has probably been carried out. The very high radon levels appeared in unventilated drifts where there was water.

#### The source of radon

There are three major factors to consider when seeking the causes of high radon concentrations; they may appear alone or in combination. These factors are ventilation, water and radioactive minerals. The first two predominate.

#### Ventilation

There are two basic ventilation principles:

- (a) The air is taken in via a shaft from the ground level down to the bottom of the mine and at each level the air is forwarded in tubes or in drifts by fans to the working places. This is illustrated in Figure 1 (left).
- (b) The air is taken in via crushed rock in abandoned upper parts of the mine or another part of the mine. This is illustrated in Figure 1 (right).

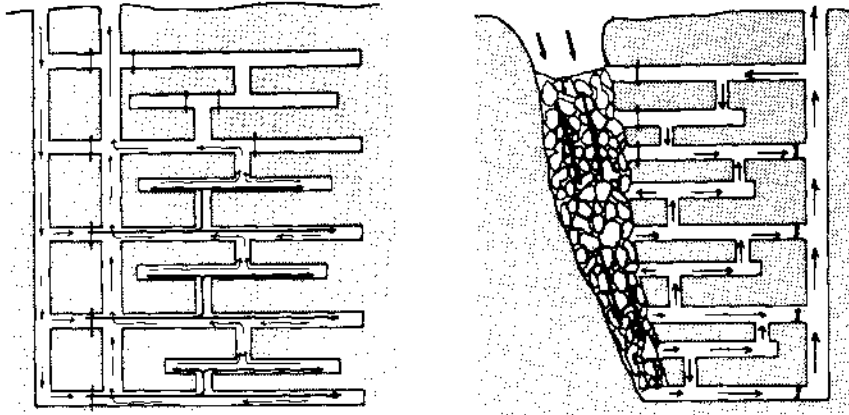


Fig. 1: Two basic ventilation principles in Swedish mines  
shaft ventilation (left) and "crushed rock" ventilation (right).

There are also combinations of these principals. The advantage of ventilation principle (b) is that during the winter the inlet air becomes comfortably warm during its passage through the crushed rock and it then contains no air impurities - except radon. During the passage through crushed rock and abandoned spaces, the air is contaminated by radon which diffuses from radium in the rock and from radon-rich water in the rock. By the time it reaches the working places the ventilation air may already have too high a radon concentration. Table 1 shows that probably 23 mines had radon daughter levels higher than 30 pCi/l in 1970. The radon source has been identified for all but one of these mines and the result is as follows:

ventilation air as the predominant source - 17 mines  
 water and rocks in the working areas as predominant source - 3 mines  
 major contributions from both the ventilation air and sources in the working areas - 2 mines

The ventilation air is the predominant radon source. Sources in the working areas are local running water and emanation from the rock wall in drifts and other spaces. Ineffective ventilation with recirculating air results in growth of the radon concentration in the air.

Even if the fact that the air is drawn through crushed rock is the main reason for the high radon levels, it is not certain that this ventilation system always leads to very high radon levels. There are examples (Mimergruvan, Blötberget, Risbergsfältet) where the intake of air is by this principle but where the radon concentrations are not especially high (15-50 pCi/l). Contributory factors are probably the venting rate, the amount of crushed rock, its structure and radium content, presence or absence of radon-rich water, etc. As it is out of the question to investigate the condition of abandoned areas and spaces with crushed rock it is very difficult to determine the real emanating source unambiguously. However, work on this problem continues.

As mentioned above, there are seasonal variations of the radon levels in many mines. This has been studied systematically in several mines by taking an air sample once a week at the same place in the mine, normally on return air. The sample has been sent by mail to the NLRP. The result from one mine is shown in Figure 2.

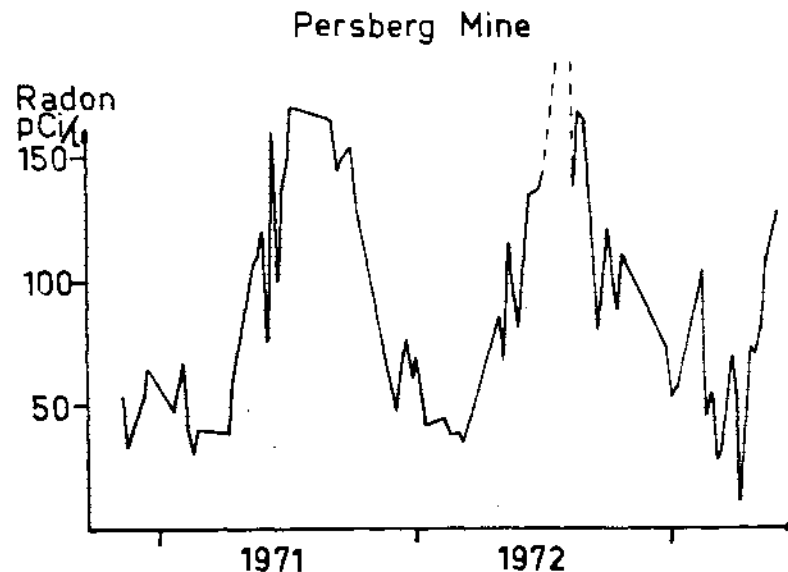


Fig. 2: Radon concentration in the return air of Persberg Mine measured once a week.

Figure 2 illustrates the seasonal variation found in some mines. The samples were taken at a depth of 150 m on return air. If the temperature outside the mine is plotted with an appropriate scale, the temperature curve and the radon curve will follow one another quite closely. It is a fact well known to mining engineers that the ventilation efficiency tends to decrease when the difference between the temperature in the mine and outside the mine decreases and the observed correlation to the temperature may reflect that phenomenon. However, one must not exclude the possibility that the variations may also be a result of different routes taken by the ventilation air or of an increase in the emanating power of the crushed rock. In a more detailed study on radon by means of a continuously running ionization chamber in a mine, diurnal variations have been observed, see Figure 3.

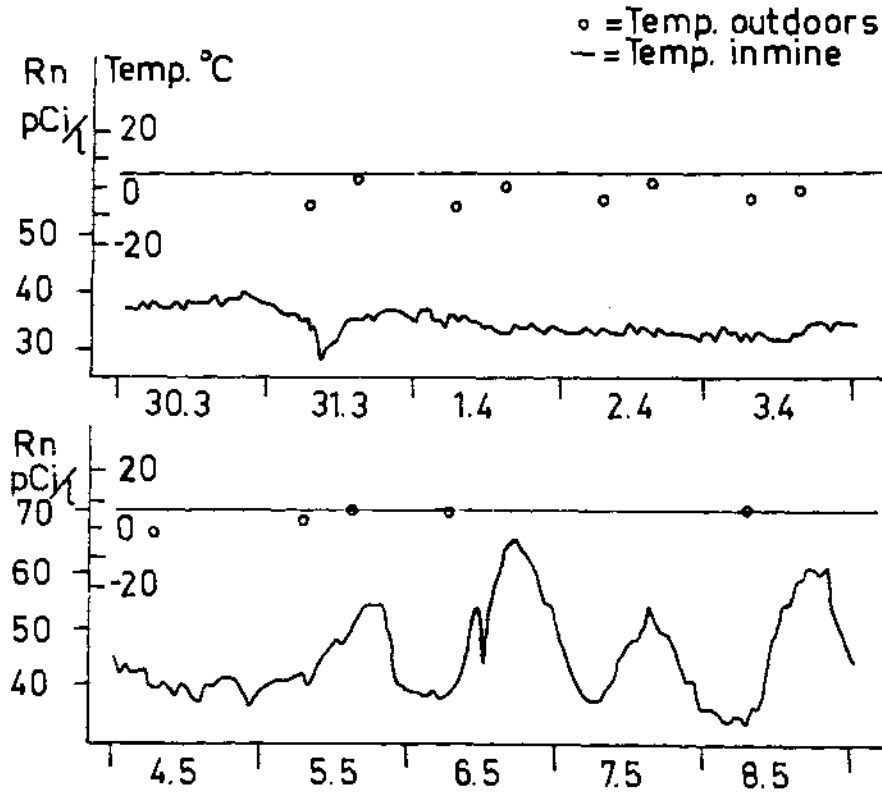


Fig. 3: Radon concentrations in the intake air at a depth of 450 m in Nygruvan Mine measured by a continuously running ionization chamber. The air has passed abandoned parts of the mine. The Figure shows March 30 through April 3 (upper curve) and May 4 through 8.

In March and April (the upper curve), the temperature outside the mine is still below the temperature in the mine at 450 m and the radon levels in the air are practically constant. In May (the lower curve) the nights are still rather cold but during the day the temperature rises above that of the mine. The radon concentration varies with a minimum during the night and a maximum during the day. This result serves to prove the influence of the temperature on the radon concentration when the temperature outside the mine is close to that inside the mine.

Water

It is difficult to prove generally the total effect of radon-rich water on the overall radon level in a mine. Nevertheless, it has been possible to show some correlations in some mines. In one mining district (Kiruna) it has been

possible to compare two adjacent mines. They have the same geology with no exceptional radium-containing minerals in the rock or in the ore. In one mine (KUJ) the radon concentration of the water is 10-20 nCi/l. The water comes from a nearby district with minerals containing radium of the order of 100-5000 pCi radium/g. The water found in that district contains up to 60 nCi/l. The radon levels in air in the mine are 20-40 pCi/l with the exception of some unventilated parts of the mine with very high radon concentrations. In the disused adjacent mine (Loussavaara) the radon concentration of the water is very low ( $<0.1$  nCi/l) and the radon concentration of the air in the unventilated mine is only 8 pCi/l. It is therefore reasonable to conclude that the water is a significant radon source in KUJ.

Radon-rich water may also cause very high radon levels in unventilated drifts. In KUJ a radon concentration in the air of 20,000 pCi/l was found in an unventilated drift. The water flow was about 50 l/s and contained 10 nCi radon per litre. Radon levels of that order of magnitude in the air are quite possible with the conditions which exist. The practical conclusion from this and other similar observations is that whenever there is a combination of bad ventilation and a large water flow there is reason to anticipate high radon levels in the air.

#### Minerals

The significance of different minerals in rock and different types of ore for the radon levels in a mine is not yet fully established.

One parameter of interest is the radium content of the rock. However, it is likely that other parameters are more relevant, as e.g. porosity, presence of cracks, humidity. Water enhances the diffusion of radon from the rock and wet mines are therefore subject to the double effect of enhancement of radon leakage and radon release directly from the water. Radioactive minerals have been found in a few mines and they have caused local radon levels of the order of 100 pCi/l. It is also possible that radioactive minerals appear in some of the crushed rock causing a high radon concentration in the ventilation air when that type of ventilation is used.

#### Protection instructions

The Swedish instructions on protection against radon in mines were issued in March 1972<sup>2</sup>. They contain a regulative part and a descriptive and explanatory part including 5 chapters on physical and biological aspect of radon and its daughters, on geological aspects, on estimation of necessary ventilation air, on sampling methods and on estimation of annual exposure. The regulative part includes regulations on permitted radon daughter exposure, on preventative action such as ventilation etc., on measurements and checks and on medical examinations. An extract from some of the most important points is presented below.

#### Permitted radon daughter exposure

1. The radon daughter exposure should be as low as practicable. If the annual exposure is more than 60,000 pCi·hrs/l, there is an overexposure and preventative action is needed.

2. The radon daughter concentration in working places should be as low as practicable. For planning of preventative action an operational limit (or "basic value" or "derived working limit") on radon daughter concentration of 30 pCi/l shall be used.

Note: 30 pCi/l is by definition equivalent to 0.3 WL.

If the radon concentration has been measured instead of the radon daughter concentration and the dose factor is not known, the dose factor shall be assumed to be at least 0.5.

Note: The dose factor is defined as the relation between the radon daughter concentration and the radon concentration.

3. If the annual radon daughter exposure is expected to exceed 60,000 pCi·hrs/l, preventative action shall be taken within the times given below. However, a total overexposure of 600,000 pCi·hrs/l during an employee's whole working time in a mine shall be avoided as counted at the latest from the time when the radon daughter concentration was determined for the first time. For estimation of total overexposure, earlier exposure shall be taken into account, if possible.

Preventative action

1. Preventative action shall be planned and performed without delay and in a reliable way.

2. The maximum time within which preventative action shall be taken in continuously used working places in the mine after a basic measurement depends on the average radon daughter concentration during the year. Corresponding radon daughter concentrations and max. times are 30-60 pCi/l, 3 years; 90 pCi/l, 1.5 years; 120 pCi/l, 1 year etc. After checking measurement the corresponding max. times are shorter by a factor of three.

Other regulations

1. Any part of the mine which has higher radon daughter concentration than 1000 pCi/l shall be sealed off by a wall impermeable to radon. Those parts of the mine which are not in use and which have radon daughter concentrations of 100-1000 pCi/l or where the concentrations are unknown shall as a rule be prohibited areas.

2. Interchange of employees shall be made if necessary to prevent over-exposure if other preventative action has not proved satisfactory.

3. Measurement to check the radon daughter concentration in the mine shall be made within the periods specified in Table 2.

Radon daughter concentration pCi/l	Time
<10	2 years
10 - 30	1 year
>30 -100	6 months
>100	3 "

Table 2. The time within which check measurements shall be made as a function of the radon daughter concentrations found in the last measurement.

4. Due to the radiological hazard in mines, special medical examinations should be made with respect to the functioning of the lungs and possible pulmonary diseases. Persons suffering from chronic disease in the bronchial system or the lungs and persons who have tuberculosis which has not with certainty become inactive or with healed tubercular changes of large extent should not be employed in work which involves particular risk of exposure to



radon daughters. Since there is a considerably enhanced risk of lung cancer for smokers and since it is feared that this risk is greater when a smoker is exposed to radon daughters, employees should be made aware of this fact and be advised against smoking, both at work and at other times.

#### Methods of calculating the exposure

As there is no good dosimeter for radon or radon daughters, the exposure is determined by indirect methods. The method proposed in the regulations is that each mine is divided into zones according to the radon daughter levels, <10, 10-30, 30-100, or 100-300 pCi/l. It is recommended that a maximum of two zones should be used. Once a year the approximate residence time in different zones is estimated for the miners and with the aid of special tables in the regulations, the exposure range is determined. The exposure ranges correspond to the same ranges as for the radon daughter level zones.

#### Epidemiological studies on the lung cancer rate

When the extent of the radon problems in Swedish mines was fully recognized, epidemiological studies on the lung cancer incidence among miners were started. There have been local studies in some mining districts, Zinkgruvan<sup>3</sup>, Boliden<sup>4</sup>, Kiruna<sup>5</sup> and Malmlberget<sup>6</sup> and an overall study including all mining districts<sup>7</sup>. This overall study started in April 1971 and the main purpose was originally to find out whether there was any excessive incidence of lung cancer at all among the miners. The study was retrospective and covered the years 1961-1968. As there were no measurements on radon in mines at that time it was not possible, in the initial study, to make any detailed exposure-effect correlations, although such analysis is in progress.

The study involved, firstly, a comparison between expected and observed numbers of lung cancers within the mining districts and, secondly, a corresponding comparison for the miners alone. The results are shown in Table 3. Appropriate corrections have been made for the age distributions in the districts in calculating the expected number of deaths as based on the death rate in the county in which the respective districts were located. In the special study on miners, the expected number of deaths is calculated as being proportional to the number of employees and to the death rate in the district.

Group	Expected numbers of lung cancers	Observed numbers of lung cancers
All men other than miners in the mining district	125	99
Underground miners only	6	26
Above ground miners only	6	7

Table 3. Expected and observed numbers of lung cancers among men aged 20-64 years during the years 1961-1968 in mining districts for non-miners and miners. The observed numbers of lung cancers are those which relate to cancer which appeared and caused death within 5 years of cessation of employment at the mine.

It can be seen from Table 3 that there is a significant excess of lung cancers among underground miners. No excess is found among miners working above ground or among other men in the mining districts. The total number of men in the mining districts is 136,600 and the number of miners is about 13,000, of which about one half are underground workers. Of the 26 observed cases of lung cancer, 21 occurred among miners who had worked more than 10 years underground. Of the total of 48 miners belonging to this group (more than 10 years under-

ground) 21 (58 %), died within 5 years of ceasing employment and 39 persons (81 %) within 15 years.

There are obvious difficulties in estimating the true radon daughter exposures in a retrospective study without earlier measurements on radon daughters in the mines. Nevertheless, attempts are in progress to reconstruct the former ventilation conditions in some mines and more reliable values for the exposure may be given than are possible today. However, if the mines and corresponding miners are divided into two groups, one experiencing less than 30 pCi/l of radon daughters and the other more than 30 pCi/l according to the measurements since 1969, the distribution will be as shown in Table 4. The numbers in Table 4 are based on very recent follow-up studies including the years 1961-1971.

Radon daughter concentration pCi/l	Number of lung cancer deaths Age at death (years)			Number of mines	Number of miners 1966
	20-64	65-79	20-79		
<30	9	7	16	9	2,760
>30	27	23	50	11	2,099

Table 4. Observed numbers of lung cancer deaths among miners who have worked more than 10 years underground. All but 2 of the miners aged 20-64 years died within 5 years of cessation of employment at the mine. 30 pCi/l is equivalent to 0.3 WL.

It is clear from Table 4 that the relative numbers of lung cancer deaths are much greater in the mines with radon daughter levels in excess of 30 pCi/l than in the other mines.

Most of the observed lung cancer deaths among miners aged 20-64 years occurred during the last half of the period of investigation (24 of 36). The year of commencement of underground work for the deceased miners is more scattered: 8 miners during 1920-30, 12 miners during 1931-40, 12 miners during 1941-50, 3 miners during 1951-50 and 1 miner in 1961.

By assuming as a first approximation that the radon daughter levels in Swedish mines found by the measurements since the end of the 1960s are representative for earlier years, it is possible to make some qualified guesses as to the exposures. This assumption is, of course, uncertain but there is some justification for it. The ventilation has been improved with time and it is reasonable to assume that this would cause the radon levels to decrease. On the other hand the mines are continually being deepened, and that will increase the amount of crushed rock and also make the ventilation more difficult, i.e. the radon levels would increase with time if no other factors were involved. The resulting effect may well have been a relatively constant radon situation over the last few decades. On this assumption it is possible to determine an exposure-effect relationship according to Table 5. The exposure ranges in Table 5 are caused by the radon daughter ranges 1-10, 10-30, 30-100 and 100-300 pCi/l and the range of employment time for the deceased workers.

Estimated cumulative exposure, WLM		Employees underground (average)	Expected number of lung cancer deaths 1961-1971	Observed number of lung cancer deaths 1961-1971	Calculated annual mortality per 10 <sup>4</sup> miners from lung cancer	
Range	Average				Expected	Observed
2- 36	15	1001	1.25	2	1.1	1.8
13- 112	48	1852	2.31	7	1.1	3.4
48- 528	218	1488	1.85	15	1.1	9.2
170-1512	696	525	0.66	12	1.1	21

Table 5. Lung cancer deaths 1961-1971 among miners aged 20-64 years. All the deceased worked more than 10 years underground and all except two died within 5 years after cessation of employment.

The last column of Table 5 is plotted in an exposure-effect diagram in Figure 4 together with the results of the lung cancer studies in the USA among uranium miners<sup>8</sup>. There appears to be good agreement. Considering the considerable statistical errors and the uncertainty of the exposures it is, however, not possible to draw any conclusion about the dose-effect relation in the region of the low exposures around 100 WLM.

The average exposure is estimated to be 163 WLM (range 90-275 WLM corresponding to the radon daughter ranges mentioned above) assuming an average employment time of 30 years. If 1 WLM corresponds to 2 rads it follows that the rate of excess lung cancer is 1.7 cases per year per rad per million miners. The studies on uranium miners in the USA gave 0.9, on Newfoundland fluorspar miners 1.1 and on British underground iron miners 3.0<sup>8</sup>.

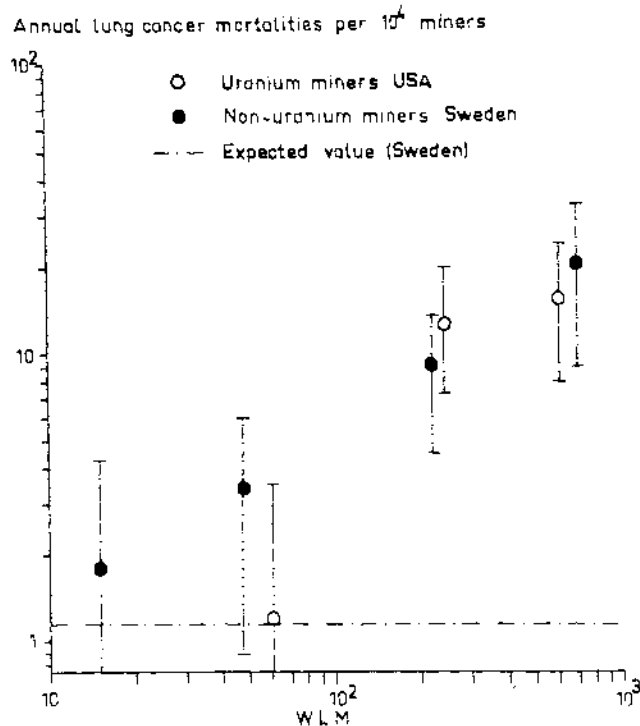


Fig. 4: Comparison between the lung cancer mortalities for American uranium miners and Swedish non-uranium miners as a function of the radon daughter exposure. 95 % confidence limits are shown for the mortality values.

### Acknowledgements

Technical assistance has been provided by Mr H Khdwall, Mr B Håkansson, Mr J Kulich, Mr C Wilson and others, assistance in the statistical analysis by Dr S Lundquist.

This NIRP investigation is supported financially and otherwise by the Swedish National Board of Industrial Safety, the Swedish Mining Association and the Swedish Atomic Research Council.

### References

1. Hearings on Radiation Exposure of Uranium Miners before the Subcommittee on Research, Development, and Radiation of the Joint Committee on Atomic Energy, Part 2, Washington, D.C.: Government Printing Office, 1967.
2. Instructions issued by the Swedish National Board of Industrial Safety, 1972, No 82, Bl 4426. Svenska Reproduktions Aktiebolag, Vällingby, Sweden (in Swedish).
3. O. Axelsson, H. Josefson, M. Rehn and L. Sundell, 1971, Lung Cancer among Non-Uranium Miners in Sweden. Läkartidningen, Vol 68, No 49, 5687-5693 and 5748-5749 (in Swedish with English Summary).
4. E. Dahlgren, 1972, Lung Cancer among Workers at the Boliden Mining Company 1950-1971. Internal Report, Boliden Aktiebolag, Boliden, Sweden, (in Swedish).
5. H. Jörgensen, 1972, Investigation on the Lung Cancer Mortality among Miners in Kiruna 1950-1970. Internal Report, LKAB, Kiruna, Sweden (in Swedish).
6. K. G. S:t Clair Renard, 1973, Lungcancer among underground miners in Malmberget 1950-1972. To be published. LKAB, Malmberget, Sweden.
7. K. G. S:t Clair Renard et al., 1972, Lungcancer among miners in Sweden 1961-1968. Gruvforskningen serie B, No 167. Svenska Gruvföreningen, Stockholm, Sweden, (in Swedish).
8. F.E. Lundin, J.K. Wagoner and V.E. Archer, 1971, Radon Daughter Exposure and Respiratory Cancer Quantitative and Temporal Aspects. National Technical Information Service, U.S. Department of Commerce. Springfield, Virginia.

QUELQUES DONNEES NOUVELLES CONCERNANT LA PROTECTION  
DES MINEURS DANS LES MINES D'URANIUM

J. PRADEL - P. ZETTWOOG - G. MADELAINE - Y. FRANCOIS  
Département de Protection - STEPPA  
Commissariat à l'Energie Atomique

Abstracts

New data concerning the safety of miners  
in uranium mines

Reports will be made, on the one hand, on the first results obtained regarding the action of radon and its elements in an experimental uranium mine at La Crouzille and, on the other hand, on the prospective use of a new portable individual dosimeter proportionate to the exposure of uranium miners to radon elements.

Introduction

Dans certaines mines d'uranium on a observé chez les ouvriers une augmentation de la mortalité par cancer du poumon. Cette augmentation semble d'autant plus marquée que les concentrations en radon et en descendants radioactifs sont plus élevées. Il importait donc d'intensifier les recherches dans ce domaine pour mieux connaître le comportement du radon et de ses descendants afin :

- de fixer des normes précises d'une sévérité suffisante mais non excessive étant donné les difficultés technologiques rencontrées pour abaisser les niveaux,

- d'améliorer l'efficacité des moyens de protection utilisés ou d'en développer de nouveaux,

- de mettre en place un système de surveillance du personnel précis, efficace et bien adapté aux conditions de la mine.

C'est pourquoi en 1969 nous avons décidé de créer dans une ancienne mine d'uranium, une mine laboratoire où il soit plus aisé de contrôler les différents paramètres expérimentaux que dans une mine en activité. Nous avons entrepris un programme de recherche dont nous présentons ici les principaux résultats actuellement acquis.

Nous décrivons également les résultats obtenus avec un nouveau détecteur individuel mesurant l'activité des descendants du radon.

I - Mine laboratoire

I/ Situation et implantation

La mine est située sur la division de La Crouzille à Fanay. Nous avons isolé du reste de la mine le quartier des Tenelles qui comprend deux niveaux séparés verticalement de 80 mètres. Le quartier est composé du montage d'entrée d'air M.560 reliant le jour au niveau 200 dans la galerie B.560; laquelle conduit en 800 m environ aux pieds des montages M. 500 et M. 501 raccordant le niveau 200 au niveau 120. La tête du montage 501 débouche dans la galerie B.301 du niveau 120. La B.301 communique avec le jour par le montage M.305. Les laboratoires sont installés, l'un au niveau 200, l'autre au niveau 120 en tête des montages M.501 et M.500; ils sont éclairés, chauffés, alimentés en courant électrique 220 V - 380 V, en air comprimé, et reliés par téléphone au central de la mine (fig. I).

La ventilation est assurée par deux ventilateurs de 18 chevaux chacun. Le ventilateur V1 souffle l'air en tête du montage M.560; il assure l'aéragé en mettant la mine en pression; V2 aspire dans la R.301, au pied du montage M.305 et assure l'aéragé en mettant la mine en dépression. Un système de trappes permet d'ajuster le débit de ces ventilateurs.

Les barrages nécessaires ont été réalisés pour isoler ce quartier des anciens travaux et limiter le circuit d'aérage à un parcours parfaitement connu.

## 2/ Aérage

Nous avons défini complètement les paramètres d'aérage de la mine laboratoire, en mesurant systématiquement les débits d'air dans chaque branche du circuit, et les différences de pression statique entre chaque noeud.

La mine est mise en pression ou dépression par l'action des ventilateurs VI ou V2. L'utilisation de diaphragmes permet de régler les débits de ventilation pour des valeurs comprises entre 2 et  $10 \text{ m}^3/\text{s}^{-1}$  environ.

La température, l'hygrométrie, la vitesse de l'air et la perte de charge entre la base et la tête des montages M.500 - 501 sont mesurées en permanence.

## 3/ Résultats

### a/ Particules ultrafines non fixées sur des noyaux de condensation

Nous rappelons uniquement ici les résultats obtenus précédemment (1) (2) au moyen de filtres précédés ou non par des tubes de Zélény ou des batteries de diffusion : l'activité sous forme de particules ultrafines, c'est-à-dire atomes non combinés ou ayant fixé quelques molécules diverses, dont le coefficient de diffusion est compris entre  $5,4 \cdot 10^{-2}$  et  $10^{-2} \text{ cm}^2/\text{s}$  (et pour lesquelles il paraît plus justifié d'adopter un coefficient de diffusion moyen de  $1,5 \cdot 10^{-2} \text{ cm}^2/\text{s}$ ), représente en moyenne 3 % de l'activité du Radium A supposé à l'équilibre avec le radon; cette activité est inférieure à 10 %, sauf en cas d'arrêt de travail dans la mine où l'on peut trouver alors des valeurs de l'ordre de 25 %.

### b/ Etude de l'influence de la ventilation sur la concentration en radon dans la mine

La concentration en radon est mesurée soit au moyen de prélèvements effectués dans des fioles recouvertes intérieurement de sulfure de zinc selon la technique en usage dans les mines françaises (4) soit au moyen de chambres de désintégration (5) de 15 litres fonctionnant à un débit de 10 l/mn permettant de mesurer des concentrations supérieures à  $0,4 \cdot 10^{-10} \text{ Ci/l}$ .

Deux chambres de désintégration ont été placées au niveau I20 (point L2) et des prélèvements ont été effectués régulièrement pour contrôler les résultats et suivre les variations rapides amorties par les chambres de désintégration. Les mesures ont été effectuées dans différentes conditions de ventilation : fonctionnement en pression en utilisant le ventilateur VI et fonctionnement en dépression en utilisant le ventilateur V2. Les différences de pression entre l'extérieur et le point L2 ont varié entre - 43 et + 55 mm d'eau. Chaque type d'aérage a été maintenu assez longtemps pour que la concentration ait atteint une valeur stable. Certains essais ont duré 4 jours (6).

Les résultats concernant les concentrations  $CR_n$ , le débit de radon  $QR_n$  en fonction du débit  $Q$  sont présentés dans les fig. 2 et 3.

On peut noter qu'à débit égal la concentration en radon dans l'air de la mine est plus faible quand la mine est en surpression que lorsqu'elle est en dépression; le facteur de réduction passe de 0,5 à 0,9 quand le débit augmente de 2 à  $10 \text{ m}^3/\text{s}$ . Ceci confirme l'observation faite dans le cas d'une mine en activité, celle de La Chapelle Largeau, où l'on a eu une réduction de 20 % des concentrations moyennes qui passaient de  $3,3 \cdot 10^{-10} \text{ Ci/l}$  en 1965 la mine étant en dépression à  $2,7 - 2,4 \cdot 10^{-10} \text{ Ci/l}$  en 1966 et 1967, la mine étant en surpression.

On observe aussi que le débit de radon dans la ventilation au niveau du point L2, pour un débit de ventilation constant de  $2 \text{ m}^3/\text{s}$  décroît régulièrement quand la pression croît en ce point (tableau I).

c/ Etude de l'influence de la ventilation sur la concentration en descendants du radon

Dans les mêmes conditions de ventilation on a effectué des mesures de l'énergie  $\alpha$  totale suivant la méthode exposée par THOMAS (2). Les résultats exprimés en "Working Levels" WL sont également précisés dans la fig. 3.

On constate aussi qu'à débit égal, le niveau exprimé en Working Levels est plus faible lorsque la mine est en surpression; le facteur de réduction est constant et égal à 0,7 lorsque le débit est supérieur à 3 m<sup>3</sup>/s.

Ces résultats ne sont probablement pas valables dans tous les cas, mais ils confirment que la mise en surpression des mines d'uranium apporte une réduction des quantités de radon dans l'atmosphère de ces mines, cette réduction pouvant être appréciable dans certains cas.

II - Détecteur individuel pour la dosimétrie des descendants du radon

La surveillance individuelle du personnel est effectuée actuellement à partir de mesures locales, le calcul des quantités inhalées par chaque agent tenant compte du temps de travail aux différents lieux.

Un détecteur individuel capable de fournir une mesure correspondant à un prélèvement continu et représentatif de l'air inhalé par le mineur permettrait d'obtenir des valeurs plus significatives. C'est pourquoi nous avons développé un tel appareil breveté permettant d'évaluer l'énergie  $\alpha$  totale des descendants du radon (7).

Pour évaluer cette énergie il est nécessaire de mesurer séparément l'activité du Radium A et celle du Radium C' qui se trouvent dans des proportions très variables. L'appareil (fig. 4) pesant environ 150 g comprend un dispositif de prélèvement alimenté par batterie permettant de collecter les descendants du radon sur une membrane millipore; le débit est de l'ordre de 4 litres par heure. Un détecteur constitué par du nitrate de cellulose coloré en rouge (LR 115 Kodak Pathé) est placé face au filtre à environ 20 mm. Les particules  $\alpha$  émises au niveau du filtre doivent traverser un collimateur à deux canons et des écrans avant d'atteindre le nitrate de cellulose.

Sur le premier canon l'épaisseur de l'écran est choisie de façon à arrêter les  $\alpha$  du Radium A et à détecter ceux du RaC' dont l'énergie résiduelle est de l'ordre de 3 MeV.

Sur le deuxième canon l'écran est tel que l'énergie résiduelle des  $\alpha$  du RaA est de l'ordre de 3 MeV alors que celle du RaC' est de l'ordre de 5 MeV.

Les caractéristiques du nitrate utilisé sont telles que les  $\alpha$  de 3 MeV sont détectés et ceux de 5 MeV ne le sont pas.

Le premier canon permet donc de mesurer le RaC' et le deuxième le RaA, ce qui permet d'évaluer aisément l'énergie  $\alpha$  totale des descendants du radon accumulés sur le filtre pendant un temps très long de l'ordre de la semaine ou même du mois.

Grâce à l'utilisation des collimateurs et des écrans les traces de particules apparaissent après attaque chimique sous forme de taches blanches toutes identiques sur fond rouge (fig.5). Il est alors aisé d'évaluer le nombre de traces par la mesure du flux lumineux de longueur d'onde convenablement choisie qui traverse le nitrate (8).

Les premiers résultats obtenus avec un prototype dans la mine expérimentale ont montré une bonne concordance pour une dizaine d'essais effectués pour des états d'équilibre variés avec les valeurs fournies par la méthode de THOMAS.

Actuellement quatre appareils sont en cours d'essai de longue durée dans les conditions d'utilisation au fond de la mine.

### Conclusions

La protection contre le radon dans les mines d'uranium ayant une importance considérable, des expérimentations de longue durée ont été entreprises dans une mine expérimentale. On a pu déjà constater que la fraction de particules ultrafines non fixées ayant un coefficient de diffusion moyen voisin de  $1,5 \cdot 10^{-2} \text{ cm}^2/\text{s}$  et non pas égal à  $5,4 \cdot 10^{-2}$  devrait être prise égale à 3 % de l'activité du Ra A à l'équilibre.

En ce qui concerne les moyens pour abaisser les niveaux de pollution, la mise en surpression des mines est d'ores et déjà à considérer comme efficace. Quant à la surveillance du personnel, l'utilisation de détecteurs individuels capables d'évaluer correctement l'énergie  $\alpha$  totale quel que soit l'état d'équilibre est à prévoir au moins pour une partie des mineurs, les premiers résultats des essais étant satisfaisants.

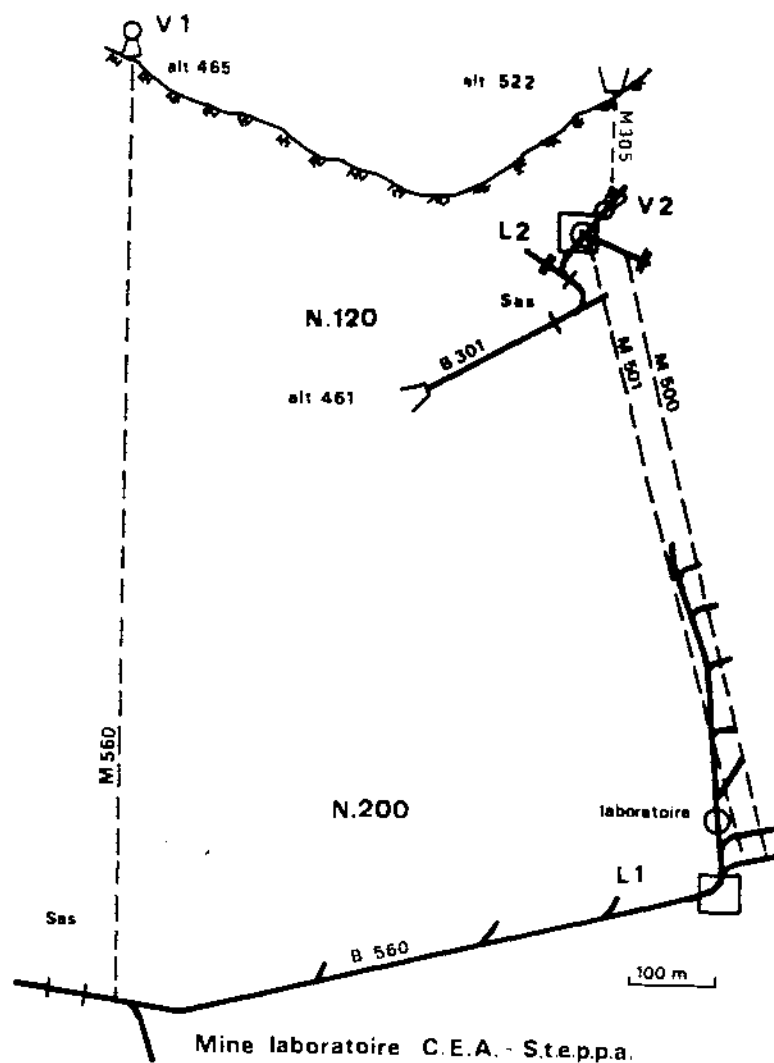


Fig.1



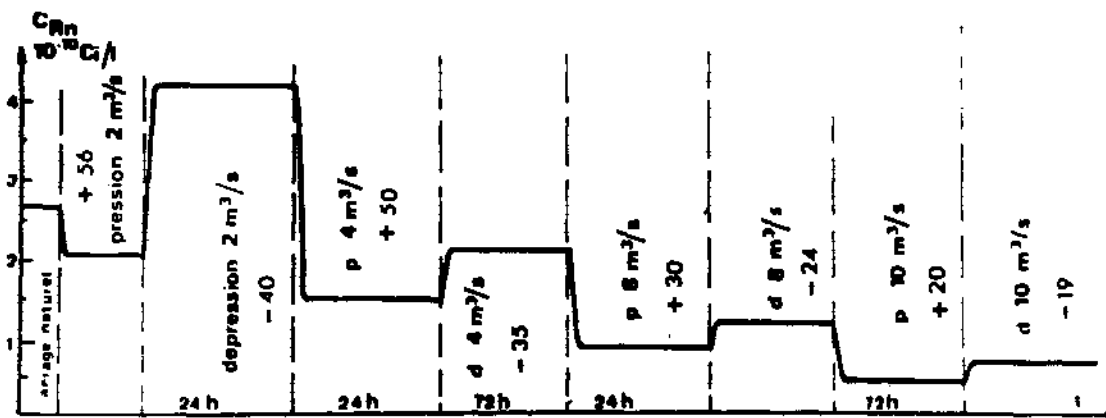
### Bibliographie

- 1 - F. BILLARD - G. MADELAINE - J. MIRIBEL - J. PRADEL  
Méthode de mesures du radon dans les mines d'uranium  
Colloque sur la protection radiologique  
Vienne 1963
- 2 - A. CHAPUIS - A. LOPEZ - J. FONTAN - G. MADELAINE  
Health Physics Vol. 25 - pp 59-65 (1973)
- 3 - THOMAS  
Modification of the Tsvoglou method for radon daughters in air  
Health Phys. Vol 19 - n° 5 (nov.1970)
- 4 - R. AVRIL - Ch. BERGER - F. DUHAMEL - J. PRADEL  
Mesures prises dans les mines d'uranium de France pour assurer la  
protection du personnel contre les dangers de la radioactivité  
Rapport Commissariat à l'Energie Atomique n° IO30 - 1958
- 5 - J. FONTAN  
Thèse de Docteur es-Sciences - Université de Toulouse (1964)
- 6 - Ph. DUPORT - G. MADELAINE  
Influence du mode d'aérage sur le dégagement du radon dans une mine  
d'uranium - juin 1973 (en cours de publication)
- 7 - A.M. CHAPUIS - D. DAJLEVIC - Ph. DUPORT - G. SOUDAIN  
Dosimétrie du radon  
Congrès de photographie corpusculaire et de détecteurs solides de traces  
Bucarest - 10-15 juillet 1973
- 8 - A.M. CHAPUIS - N. GERARD - G. SOUDAIN  
Nouveau système de lecture pour détecteur ionographique  
Radioprotection 1972, 7, 1, p.13-20

Tableau I

Débit de radon ( $10^{-7}$ Ci/s)	Pression (mm d'eau)
9,2	- 45
7,4	- 2
7,3	+ 2
5,7	+ 56

Variation du débit de radon dans la mine  
 en fonction de la pression  
 pour un débit constant de  $2 \text{ m}^3/\text{s}$



Influence du mode d'aerage sur la concentration du radon

Fig.2

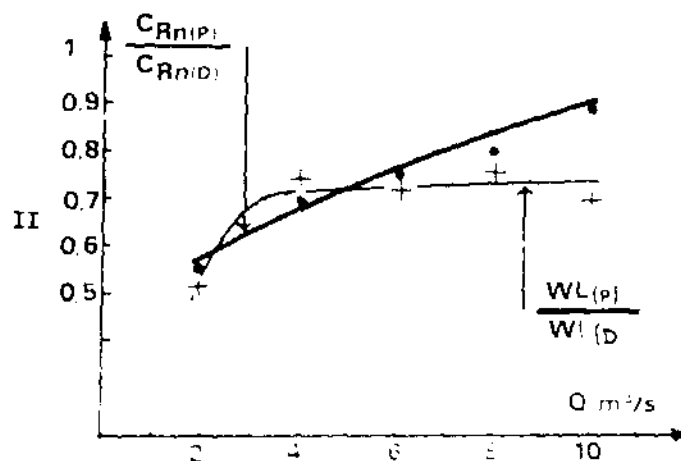
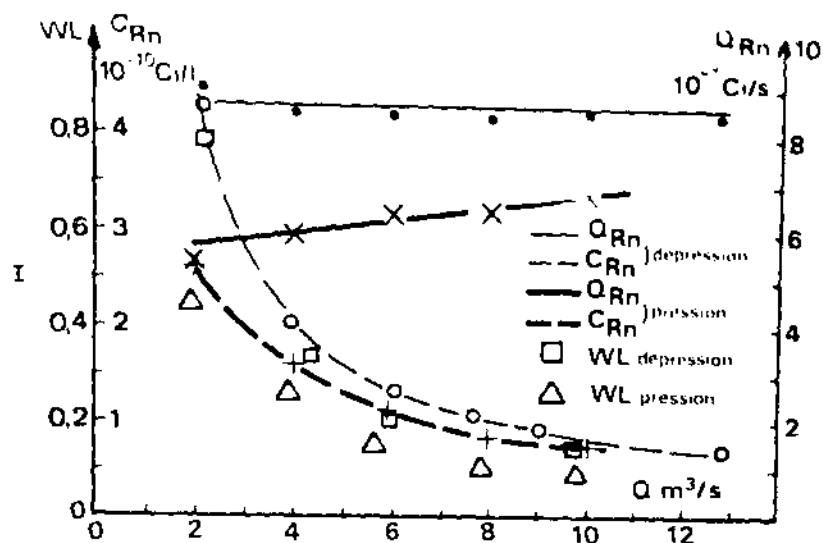
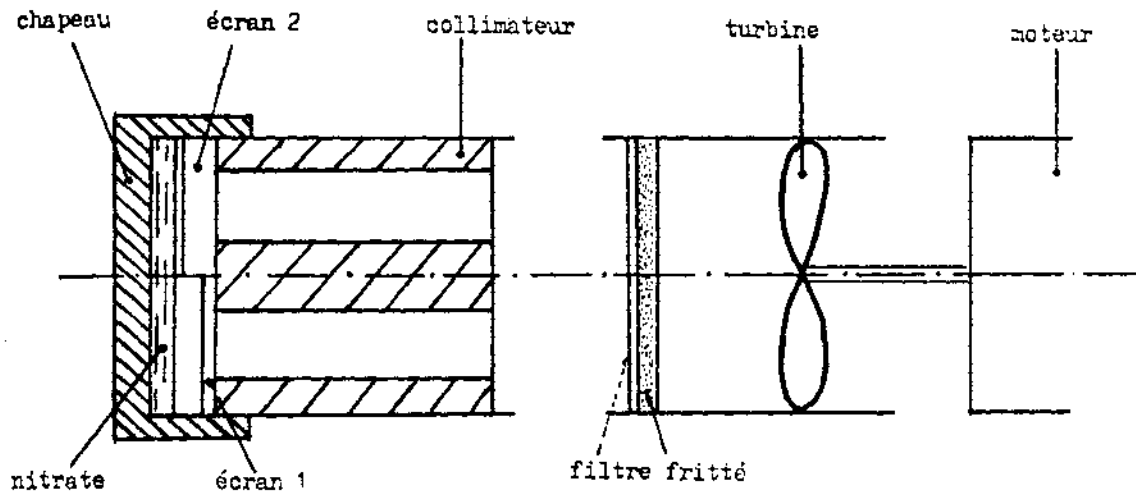


Fig. 3

- I - Concentration en radon et en descendants du radon exprimée en WL et débit de radon en fonction du débit de ventilation, la mine étant en dépression.
- II - Rapport entre la concentration en radon ou la concentration en descendants du radon pour la mine en pression et celle correspondante pour la mine en dépression, en fonction du débit de ventilation.



- Schéma de principe de la tête de prélèvement

Fig. 4

Traces de particules  $\alpha$  provenant de Radium A  
 dans le détecteur LRII5 Kodak-Pathé  
 utilisé dans l'appareil de prélèvement individuel  
 pour la dosimétrie des descendants du radon

Fig. 5



## LONG-TERM MEASUREMENTS OF RADON DAUGHTER ACTIVITY IN MINES

B. Haider, W. Jacobi

Institut für Strahlenschutz

Gesellschaft für Strahlen- und Umweltforschung mbH

München - Neuherberg, Germany

### Abstract

A rugged, portable filter monitor for the long-term measurement of Rn-daughter activity in mines with direct indication of the cumulative exposure is described. The monitor is equipped with rechargeable batteries and enables continuous or fractionated air sampling at preset time intervals over a period of 1 - 2 weeks with one battery charge. The results of continuous measurements over a one year-period in a fluorspar mine are discussed.

### Introduction

A significant excess of lung cancer mortality with increasing accumulated Radon(Rn)-exposure has been observed among uranium miners in the USA and CSSR. This fact emphasizes the suspicion that workers in mines with high Rn-content in air belong to that groups of radiation workers with the highest somatic radiation risk. Compared with this risk the surveillance of the radiation exposure of these miners is still not adequate. The main causes for this inconsistency are the complex distribution of Rn and its daughters in mine air, which varies with time and place, and the difficulties of personnel air monitoring under the abnormal working conditions in mines.

In the past air monitoring in these mines was restricted mainly on measurements of Rn or its daughters in single air probes, which were taken in more or less large intervals of time. Taking into account the varying air activity in a mine working area robust, portable and battery-operated monitors are needed which enable long-term measurements and indicate the accumulated exposure over a long time period. Several instruments of this type were proposed and tested in the last years<sup>1-6</sup>

With respect to their applicability in uranium and fluorspar mines we have studied different methods to determine the time-integral of the activity concentration of Rn and its daughters. For the continuous measurement of Rn-gas itself we used the electrostatic deposition of RaA-ions, formed by decay of Rn-atoms in a chamber with Rn-permeable walls. The results however have indicated, that the Rn-sensitivity of this method depends strongly on the air humidity.<sup>6</sup> This method seems therefore not suitable for

Rn-monitoring in mines.

For long-term measurements of the accumulated exposure to Rn-daughters in mines we have developed on the basis of the filter method portable monitors with low-power consumption. In this paper the design and properties of this monitor and the results of test measurements in a fluorspar mine are described.

Description of the Air Monitor

Two types of surveying instruments were developed: A direct-indicating monitor with a Silicon- $\alpha$ -detector and a nondirect-indicating instrument using track etch foils as integrating  $\alpha$ -detector.

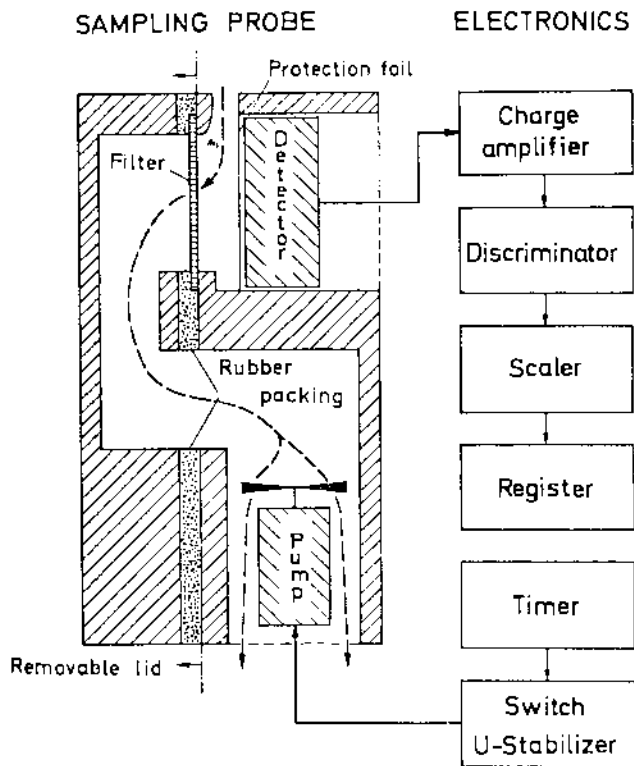


Fig. 1:  
Cross section through the sampling probe of the monitor and block diagram of the electronics

Figure 1 shows a schematic cross section through the sampling probe of the direct-indicating instrument and a block diagram of its electronics. The outside air is sucked in through slits in the sampling probe, passes the fibrous filter and is exhausted on the other side of the probe. The sampling probe can be easily opened for filter replacement. The small air blower is mounted in the sampling probe; its operation voltage of 3,5 V is stabilized to assure a constant air flow-rate. With the normally used cellulose-asbestos filters (effective filter diameter 18 mm) a flow-rate of 2.5 liter/hour was adjusted. As  $\alpha$ -detector a p-Silicium semiconductor with an active surface of 240 mm<sup>2</sup> ( $\phi=17.5$ mm) is used, whose surface is protected by a thin mylar foil of 1 mg/cm<sup>2</sup> thickness. The distance between filter and detector surface is 5 mm.

After amplification and pulse height discrimination the detector pulses are counted by a combination of an electronic scaler and a 4-digit mechanical register. The electronic part includes also a timer and switching circuit for the air blower, which enables automatic, fractionated air sampling in preset time intervals. All electronic parts are designed for low power consumption. The total power consumption of the monitor is about 300 mW, from which about 200 mW are required for the air blower. For the power supply rechargeable dryfit PC-accumulators are used, which enable with one battery charge an operation time of 8 - 9 days at continuous sampling and of about 18 days at fractionated sampling.

The electronics, the counter and the accumulators are enclosed by a stable, water and dust protected metal housing, to which the sampling probe is flanged on. All operating elements and plug sockets for external connections (rate meter, pulse height analyser) are mounted behind a lid to prevent contamination by dust and undesired changes of the adjusted operation values. Only the mechanical register can be read through a window in the metal housing. The instrument dimensions are 23 cm x 12 cm x 32 cm (height). Its total weight is 7 kg, from which about 5 kg falls to the accumulators.

In a second type of this instrument the Si- $\alpha$ -detector is substituted by a track etch foil with the same effective diameter; in this type the detector electronics and the counter are omitted. Foil etching and  $\alpha$ -track counting is done by the usual techniques and are described in detail elsewhere<sup>6</sup>

#### Instrument Calibration

For the monitoring of Rn-daughter mixtures in mines and room air the concept of potential  $\alpha$ -energy concentration and the unit 1 WL =  $1.3 \times 10^5$  (pot. $\alpha$ -) MeV/liter air have been introduced. As described earlier the described instrument was constructed with the aspect to determine the time integral over this energy concentration over long exposure periods. The total number  $Z_\alpha(T)$  of  $\alpha$ -tracks or  $\alpha$ -pulses, respectively, counted with the instrument during a time period T is connected with this integral or accumulated exposure E(T) by the equation<sup>6</sup>:

$$Z_\alpha(T) = \frac{\beta r f v}{p} \cdot E(T)$$

In this equation means  $\eta$  the counting efficiency of the used type of detector,  $f > 0.99$  the deposition efficiency of the filter,  $v = 2.5 \pm 0.3$  liter/hour the air flow rate and  $p = 7.68$  MeV the potential  $\alpha$ -energy of one  $^{218}\text{Pb}$  (RaB)- or  $^{214}\text{Bi}$  (RaC)-atom.

The correction factor  $\beta$  depends on the relative composition of the Rn-daughter mixture in the measured air, which varies with the ventilation rate  $\lambda_v$  and the rate constant  $\lambda_a$  for the attachment of free daughter atoms to particles in the considered mine area.<sup>6</sup> It was calculated on the basis of the box model for Rn-atmospheres which was developed by one of the authors<sup>7</sup> and is given in figure 2. This graph shows that the variation range of the correction factor  $\beta$  is rather small and a constant value  $\beta = 1.05$  can be applied to most mine and room atmospheres with sufficient accuracy.

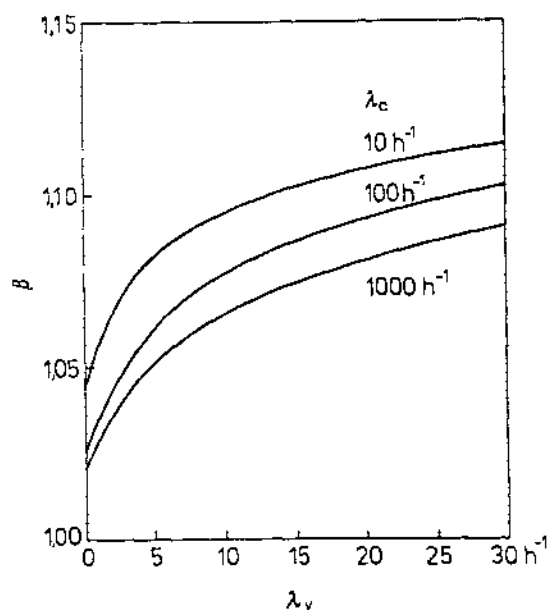


Fig. 2:  
Correction factor  $\beta$  for  
instrument sensitivity  
(see text)

The counting efficiency  $\eta$  of the direct-indicating instrument with a Si- $\alpha$ -detector was determined by comparison with calibrated  $\alpha$ -sources. At the normal operational conditions used it results  $\eta_{Si} = 0.18 \pm 0.01$ . With this value it follows an instrument sensitivity in the case of continuous sampling:

$$\begin{aligned}
 S &= Z_{\alpha}(T)/E(T) \\
 &= 0.061 \pm 0.011 \text{ counts per (pot.}\alpha\text{)MeV}\cdot\text{hour/liter air} \\
 &= 7900 \pm 1400 \text{ counts per WL}\cdot\text{hour}
 \end{aligned}$$

This corresponds to a sensitivity of  $79 \pm 14$  counts per pCi $\cdot$ hour/liter air of each daughter nuclide in the case of radioactive equilibrium in air. The built-in scaler enables a reduction of this sensitivity in steps of 1/2 to a 1/128 of this value or 62 counts per WL $\cdot$ hour. In addition the sensitivity can be reduced by the built-in timer for fractionated air sampling. The background counting rate is about 5 counts per hour. The lower detection limit of the instrument is therefore comparable with the mean concentration of Rn-daughters in atmospheric air.

The sensitivity of the instrument with track etch foils was determined by simultaneous field measurements with both types of instruments. Depending on etching conditions and the used foil material it is about a factor 0.2 - 0.6 lower than the sensitivity for the direct-indicating instrument.

#### Test Measurements

Test measurements with 2 instruments of each type were performed so far in 3 fluorspar mines in East Bavaria.<sup>6</sup> One direct-indicating monitor was continuously in use for one year at the same working area in a mine drift and was operated and controlled by the foreman of the miners in this area. He read off the counting register of the instrument normally at the beginning and the end of



each working shift. The resulting time variation of the potential energy concentration in the mine air during this one year-period is shown in figure 3. The annual mean value was about 1 WL, whereas the daily mean values are varying in the range from 0.4 - 3 WL. Figure 3 indicates that the short-time variations from day to day in this mine area are rather small. However, a rather strong long-term variation of the Rn-daughter level is observed.

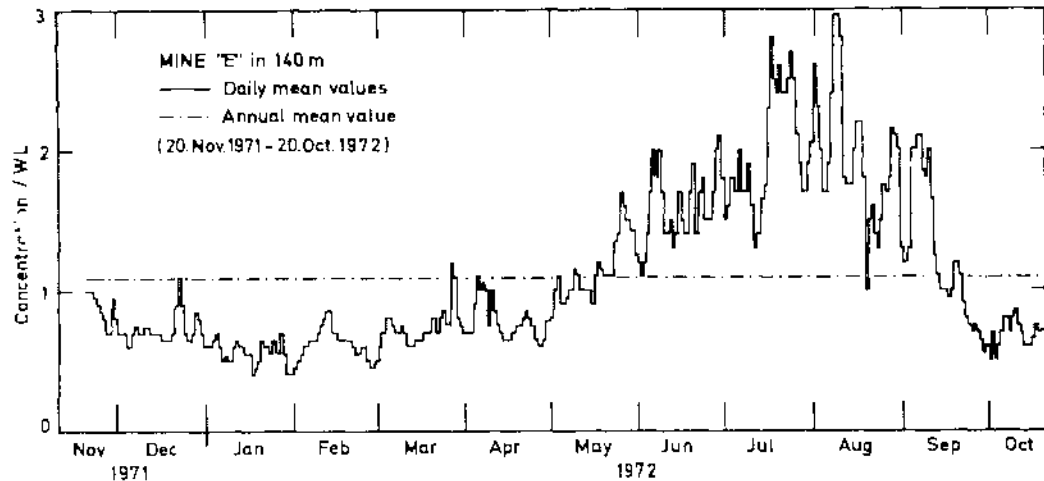


Fig. 3: Variation of the continuously measured Rn-daughter activity in a working area of a fluorspar mine in East Bavaria (Nov. 1971 - Oct. 1972).

This variation is mainly due to the change of the air ventilation during the extension of the mine gallery. During the period from November 1971 till about May 1972 a rather constant activity level of about 0.5 - 1.0 WL was observed. In the following months the fresh-air supply to the driving gallery was reduced. During this period the air-activity increased and reached rather high values of 2 - 3 WL in July - September 1972. After break-through of a new wind gate the supply with air of low Rn-content increased. Subsequently the Rn-level in the working area decreased strongly and reached a rather constant level of about 0.6 WL in October 1972.

The variation of the Rn-exhalation from the walls of the gallery due to the mining activity was probably not so significant in this fluorspar gallery, but might be certainly of more importance in other galleries, especially in uranium mines.

In either case, these test measurements indicate the necessity of long-term Rn-monitoring to get more information about the real cumulative exposure of miners.

-----

This work was financially supported by the Ministerium für Bildung und Wissenschaft of the Federal Republic of Germany.

-----

### References

- 1 BECKER, K.: Health Phys. 16, 113-123 (1969)
- 2 ROCK, R.L., D.B. LOVETT, S.C. NELSON: Health Phys. 16, 617-621 (1969)
- 3 BEDROSIAN, P.H.: Health Phys. 16, 800-802 (1969)
- 4 COSTA-RIBEIRO, C., J. THOMAS, R.T. DREW, Mc D.E. WRENN, M. EISENRUD: Health Phys. 17, 193-198 (1969)
- 5 Mc CURDY, D.E., K.J. SCHIAGER, E.D. FLACK: Health Phys. 17, 415-422 (1969)
- 6 HAIDER, B., W. JACOBI: Entwicklung von Verfahren und Geräten zur langzeitigen Radon-Überwachung im Bergbau; Research Rep. BMW-FB-K 72-14, August 1972
- 7 JACOBI, W.: Health Phys. 22, 441-450 (1972)

ОЦЕНКА РАДИОАКТИВНОГО ФАКТОРА НА ПРЕДПРИЯТИЯХ  
ПРОМЫШЛЕННОСТИ РЕДКИХ И ЦЕЛЕННЫХ МЕТАЛЛОВ.

Ляровский П.П., Елзовская Л.Т., Напитанов П.Т.,  
Лихачев В.П., Сатарина С.А., Хухлаева С.А.

Институт гигиены труда и профзаболеваний АМН СССР  
г. Москва.

Связь Советских Социалистических Республик.

Summary

When working with natural radioactive raw materials of different mineral composition and different specific activity due to the type and conditions of the particular jobs the environment in which man works can reveal different extents of activity of individual components of the factor of radiation.

Surface jobs with mineral raw materials and their processed products are extremely typical for their dusts containing long-term  $\alpha$ -irradiators. Such dusts pollute the air environment of some production premises of ore-dressing and chemical and metallurgical enterprises.

Under examination was the state of health in workers with long-term of service. No harmful effects upon the respirative organs which could be directly related to the radiation factor have been discovered so far. However the longest term up to 20 years was only in a small group of the examined.

The amount of Th incorporated into the workers' lungs evaluated by the preliminary data taken from the entire body and the Tn content in the respired air did not exceed 150 pCi.

The authors studied 3500 white ordinary rats intratracheally injected with dusts of various specific activity as well as chronically inhaled for long terms. It has been found out that with a combined dust-radiation action the lowest effective dose to affect the lungs is close to 200 rem.

In view of the increased fibrogenic properties of dusts containing radioactive elements the authors stress the necessity to consider the long-term natural radioactive component of mineral dusts when fixing the permissible limits of general pollution within the operation zone of the production premises. The authors regard as open the question on possible distant effects in workers with 30-50 years of service since with the length of service increased it is quite possible for the lungs to absorb doses equal to the maximum permissible ones.

Радиационная опасность на предприятиях промышленности редких металлов обусловлена содержанием в добываемых и обогащаемых рудах естественно-радиоактивных элементов ряда тория и урана. Несмотря на незначительное содержание в исходном минеральном сырье естественно-радиоактивных элементов (от  $1 \cdot 10^{-11}$  кюри/г до  $1 \cdot 10^{-10}$  кюри/г), в ходе изучения условий труда выявлены работы, выполнение которых может сопровождаться загрязнением воздушной среды радиоактивными аэрозолями и газами.

Для наземных работ с минеральным сырьем и продуктами его переработки особенно характерно постоянное содержание в пыли, загрязняющих воздушную среду некоторых рабочих зон и производственных помещений, долгоживущих альфа-излучателей. К таким работам относятся работы на шарошечных станках в открытых карьерах, все виды работ при сухих методах обогащения концентратов редких и цветных металлов на обогатительных фабриках, многочисленные операции по доводке концентратов, шихтовке, затариванию и т.д. Поэтому, понятен наш интерес к оценке роли естественно-радиоактивного компонента промышленных пылей предприятий промышленности редких и цветных металлов, тем более, что ряд исследователей, оценивавших последствия радиационного воздействия на рабочих урановых рудников<sup>1,2</sup>, высказывали предположение о том, что пыль находится среди таких факторов, которые могут обусловить возникновение рака легких при дозах облучения меньше тех, которые признаны необходимыми для индуцирования рака при "чистом" радиационном воздействии.

Сравнение уровней заболеваемости различных профессиональных групп из числа лиц, работающих на предприятиях промышленности редких металлов и изучение состояния здоровья стажированных рабочих не выявило повреждающих эффектов со стороны органов дыхания, которые можно было бы поставить в прямую связь с воздействием радиационного фактора. Но наибольший стаж - до 20 лет, был достигнут лишь в небольшой группе обследованных рабочих. Содержание тория в суточном количестве биосубстратов не превышало 20 мкг с калом, 10 мкг с мочой. Количество инкорпорированного тория в легких рабочих, оценивавшееся по результатам предвсмертельных измерений на счетчике всего тела и по содержанию торона в выдыхаемом воздухе, не превышало - 150 пикокюри.

На 3500 белых беспородных крыс проведены экспериментальные исследования по изучению патогенных свойств труднорастворимых торийсодержащих минеральных пылей, в связи с их радиоактивностью. При этом в экспериментах с однократным интратрахеальным введением использовались пыли различных редкоземельных торийсодержащих руд и концентратов цирконовой и лопаритовой группы с присущей им удельной активностью и ее искусственным увеличением до  $1 \cdot 10^{-6}$ ,  $1 \cdot 10^{-7}$  кюри/г. Увеличение удельной активности исследуемых пылей за счет искусственных добавок ферриторита, торанита или искусственной  $Ta_2O_5$  нами применялось для усугубления условий эксперимента, которые бы позволили, пусть даже в расчете на нереальный "худший" случай, выявить влияние радиационного фактора на фоне пылевого.

При обычной постановке исследований, с использованием пылей только природной удельной активности, как мы уже знали, и исходного уровня удельной активности и 12-ти месячного срока наблюдения было недостаточно для того, чтобы составить представление о возможных неблагоприятных последствиях воздействия такой комбинации факторов - пылевого и радиационного<sup>3</sup>.

Эксперименты с хроническим ингаляционным воздействием осуществлялись с

помощью ингаляционных установок<sup>4</sup>. Запыленность воздуха в камере с учетом дисперсности пыли и коэффициента ее задержки, была выбрана равной 300-400 мг/м<sup>3</sup> с тем, чтобы к концу затравки в легких накопилось 50-75 мг вещества - количества, достаточного для развития в легких пневмоконнотического процесса, на фоне которого выявлялось влияние радиационного компонента пыли.

Таким образом, в экспериментальных условиях создавалась модель хронических пылерадиационных воздействий на фоне различных поглощенных доз на легкие - от нескольких бар, до 750 бар и более. Величина суммарной дозы, в каждом конкретном случае диктовалась возможностью создания аналогичной дозы в процессе 50-летнего профессионального контакта лиц, работающих с естественными радиоактивными веществами разной удельной активности, не превышающей в производственных условиях  $1 \cdot 10^{-8}$  кюри/г.

Экспериментальные исследования выявили, что при комбинированном пылерадиационном воздействии минимально-эффективной дозой на легкие является доза близкая к 200 бар. Выявлению этой величины способствовали данные по изучению темпов прироста веса тела животных в динамике, функции внешнего дыхания, состояния показателей периферической крови, продолжительности жизни животных, а также результаты исследования биохимических и патоморфологических изменений тканей легкого, как критического органа.

При экспериментальном изучении сравнительной патогенности естественно-радиоактивных минеральных пылей к тем же пылям с усиленным за счет искусственных добавок радиационным компонентом многие из вышеуказанных показателей дали многочисленные свидетельства усиления фиброгенности пылей по мере увеличения их удельной активности.

Так, рис. 1 демонстрирует увеличение сухого веса легочной ткани, наблюдающееся как в сериях с рудными пылями наибольшей удельной активности, так и в аналогичных, по активности, сериях с пылью концентрата, что особенно отчетливо (достоверность >95%), проявляется к 12 месяцам от начала эксперимента.

К этому же сроку увеличивается абсолютное количество оксипролина (рис. 2) в легочной ткани животных, подвергнутых воздействию пылей наибольшей удельной активности, суммарных и нерастворимых белков, снижается интенсивность включения меченой по  $C^{14}$  аминокислоты лизина и глицина. Патоморфологические исследования выявили признаки слабо прогрессирующего диффузно-очагового пневмоконнотоза, хронического бронхита и бронхолита, плазматизации лимфоидных клеток лимфатических узлов. В эксперименте отмечено некоторое усиление патогенных свойств пылей к 15-18 месяцам от начала воздействия за счет общей дозы, близкой к 200 бар, что выражалось в более очаговом характере пневмосклероза (рис. 3) и увеличении лимфореинулосарком легких животных.

При исследовании усугубленного влияния смесей циркона с двуокисью тория и чистой двуокиси тория отмечено снижение продолжительности жизни животных (рис. 4,5). Рисунки демонстрируют сложность происходящих в организме животных процессов, так как при дозе большей 200 бар на легкие в сериях 2 и 3, в одном случае, у животных этих серий  $ET_{50}$  было существенно ниже, чем таковое у животных возрастного физиологического контроля (К) и животных 1 серии (рис. 4). В другом, несмотря на дозу превышающую 200 бар (2 серия),  $ET_{50}$  было аналогично таковому у контрольных животных (рис. 5). Основные статистические параметры, приведенные на рис. 4-5, получены при обработке данных по методу Литчфильда и Блисса.

На основании экстраполяции результатов экспериментальных исследований было установлено также, что период полувыведения тория,

входящего в состав пыли природных соединений-минералов цирксона и лопарита, попавших в легкие, для человека находится в пределах от 4,3 до 5,2 лет. Тогда при соблюдении допустимых уровней запыленности воздуха рабочей зоны расчетным путем получено, что доза, поглощенная легкими рабочих, занятых на производстве цирконового концентрата, может достигать 300 бэр, а при производстве лопаритового концентрата 1300 бэр в течение 50-летнего профессионального контакта.

Вместе с тем, ПДД для персонала за 50 лет равна 750 бэрб. В связи с этим установленная нами в эксперименте минимально эффективная доза, равная 200 бэр, может быть свидетельством необходимости учета содержания в пыли радиоактивного компонента при гигиеническом нормировании пылей в воздухе производственных помещений.

Таким образом на основании проведенных исследований можно сделать следующие выводы:

1. Минимальная эффективная доза на легкие белых беспородных крыс при комбинированном пыларadiационном воздействии равна 200 бэр.

2. Некоторые виды работ с естественно-радиоактивными веществами, такими как лопаритовый концентрат, должны относиться к радиационно-опасным, так как в производственных условиях возможно получение легкими рабочих за 50 лет профессионального контакта доз, больших 200 бэр.

3. При работах с веществами малой удельной активности (не более  $1 \cdot 10^{-9}$  юри/г) радиационная безопасность может быть достигнута соблюдением общегигиенических требований, касающихся допустимых уровней общей запыленности воздуха рабочей зоны, которые должны устанавливаться с учетом наличия в пыли долгоживущего альфа-активного компонента.

#### Библиография.

1. Staff Report of the Federal Radiation Council No 8 revised guidance for the Control of radiation Hazards in uranium mining, USA, 1967.

2. Radiation exposure of uranium mines. Federal Radiation Council, Washington, 1968.

3. Л.Т.Еловская, Т.А.Кочеткова "О гигиеническом значении радиоактивных примесей полиметаллических руд на горнообогатительных комбинатах. В сб. "Борьба с пылеобразованием на производстве, Изд-во "Медицина", Москва, 1964, стр. 106-116.

4. И.П.Валезнев, Л.Т.Еловская, П.И.Моисейцев. Авторское свидетельство № 265307, 1969.

5. Блисс и Литчфилд цитир. по М.Л.Беленькому "Элементы количественной оценки фармакологического эффекта", Ленинград, 1963.

6. Нормы радиационной безопасности НРБ-69, атомиздат, 1972.

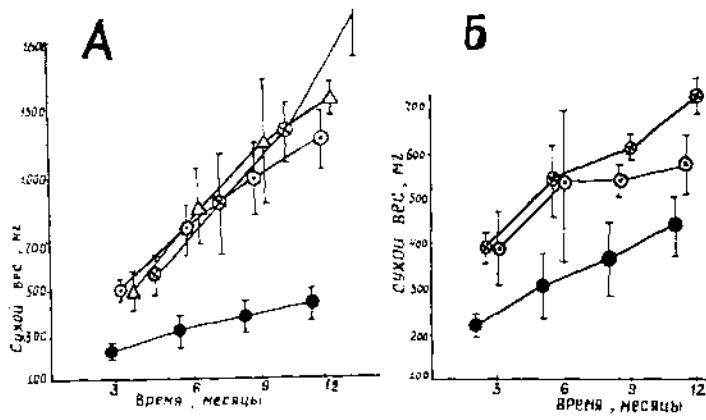


Рис. 1. Сухой вес легочной ткани при однократном интотрахеальном введении пылей циркониевых руд - А и циркониевых концентратов - Б

Обозначения

- А  
 -  $10^{-11}$  нри/г  
 Δ -  $10^{-9}$  - " -  
 -  $10^{-7}$  - " -
- Б  
 -  $10^{-10}$  нри/г  
 -  $10^{-8}$  - " -  
 - контроль

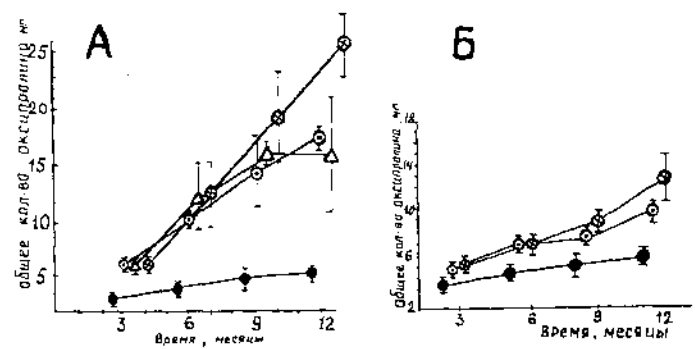


Рис. 2. Оксипролинлегочной ткани.

Обозначения те же.

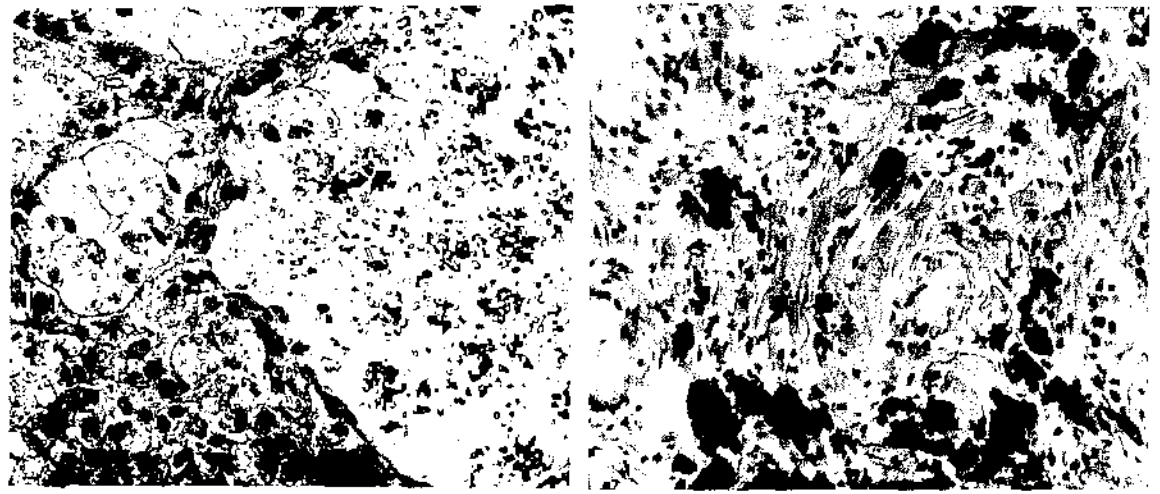


Рис. 3. Легкие крысы при ингаляции пыли: а)  $10^{-11}$  нри/г через 6 мес. Макрофаги с большим количеством пылинок не гибнут, пролиферация гистиоцитов альвеолярных перегородок; б) чистой двуокиси циркония через 15 мес., резко выраженный фиброз в зоне отложения пыли. Гематоксилин-эозин,  $\times 350$ .

обознач. серии	ET, сутки, при	с	К	О	1	2	3	4
	16%	50%	84%	ET50	TR	fTR	TR	fTR
К	400	500(48-57)	625	1.14				
1	353	415(375-457)	483	1.06	1.2	1.175		
2	460	530(485-577)	650	1.09	1.06	1.167	1.27	1.14
3	220	304(268-342)	425	1.145	1.65	1.205	1.36	1.18
4							1.74	1.17
обознач. серии	N	S	f	s	SR	fSR	SR	fSR
К	12	1.24	1.12					
1	9	1.16	1.077	1.07	1.14			
2	19	1.18	1.08	1.05	1.142	1.02	1.115	
3	21	1.375	1.145	1.06	1.19	1.05	1.165	1.164

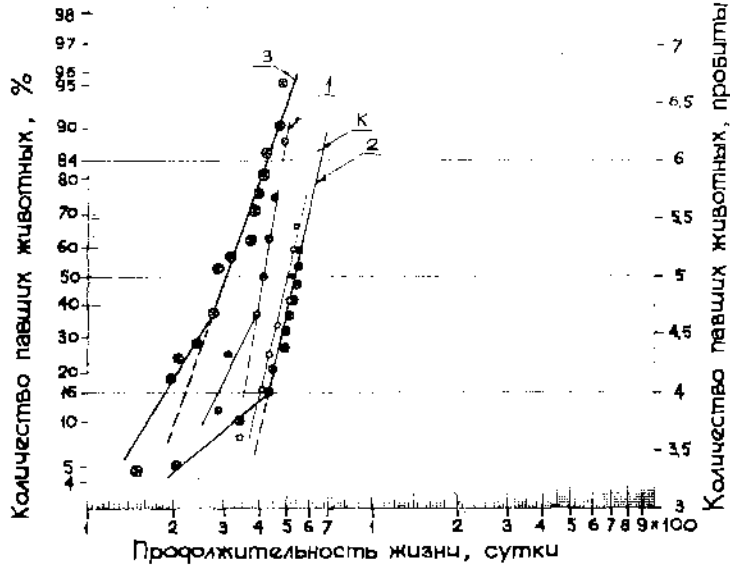


Рис. 5 Выживаемость животных при хроническом ингаляционном воздействии аэрозолей лопаритового концентрата /1 серия - 0,025 % Th; 2 - 10,16 % Th; 3 - 46,2 % Th/; К - контроль.

обознач. серии	ET, сутки, при	с	К	О	1	2	3	4
	16%	50%	84%	ET50	TR	fTR	TR	fTR
К	396	515(470-560)	750	1.135				
1	352	460(415-505)	600	1.17	1.19	1.22		
2	330	395(350-440)	480	1.185	1.24	1.16	1.27	
3	264	370(320-420)	520	1.33	1.47	1.36	1.24	1.38
4	284	370(325-415)	490	1.19	1.19	1.22	1.07	1.24
							1.0	1.36
обознач. серии	N	S	f	s	SR	fSR	SR	fSR
К	17	1.87	1.12					
1	9	1.295	1.18	1.0	1.07			
2	9	1.295	1.14	1.10	1.0	1.19		
3	5	1.39	1.24	1.27	1.07	1.27		
4	16	1.3	1.095	1.18	1.0	1.19	1.17	1.26

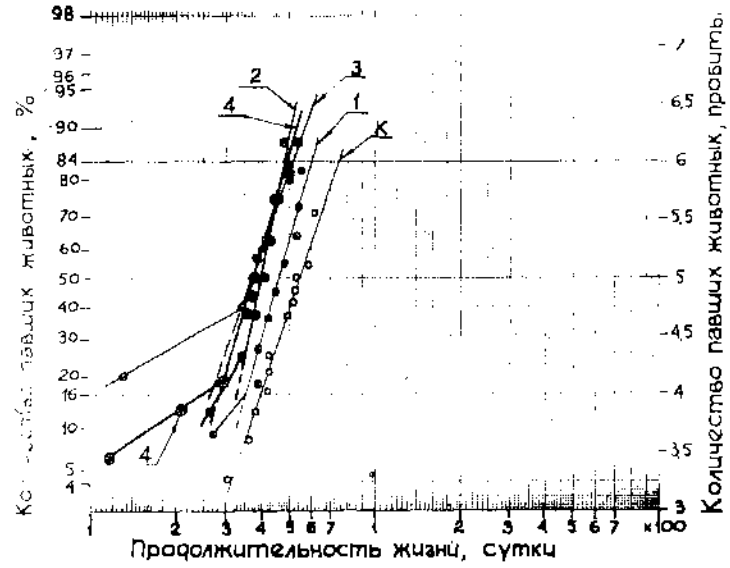


Рис. 4 Выживаемость крыс при ингаляционном воздействии аэрозолей цирконового концентрата разной удельной активности /1 - 0,025 % Th, 2 - 4 % Th/ чистой двуокиси тория /4/ и цирконового концентрата в сочетании с торием /3/. ET - эффективное время /продолжительность жизни для 16, 50 и 84 % павших животных/.



## A RADON DAUGHTER MONITOR FOR USE IN MINES

A. C. JAMES and J. C. STRONG

National Radiological Protection Board,  
Harwell, Didcot, Berks, U.K.

### Abstract

A self-contained monitor for the measurement of airborne RaA, RaC and the working level, WL, in the shortest practicable time and with a minimum of calculation is described. The ratio of two gross alpha-counts gives, directly, the ratio of airborne alpha-emitter concentrations, RaC':RaA, after collection on a filter. This is achieved because the contribution from airborne RaB to the measured alpha-activity is minimised by choosing short sampling and counting times. The RaA concentration is derived from the first alpha-count, during air sampling. WL is derived from the second alpha-count. No background correction is required. Systematic errors in estimated quantities are small. With a total measuring time of eleven minutes, the limits of detection are approximately 1 pCi/litre for RaA and  $10^{-4}$  for WL.

### Introduction

A radiation hazard from the short-lived radon daughters in uranium and other metal mine atmospheres has been convincingly shown.<sup>1</sup> Most of the epidemiological studies made in this context have used the working level (WL) as the unit of exposure. 1 WL represents the concentration of radon daughter activity in an atmosphere, and is defined as any combination of the daughters (RaA, RaB, RaC and RaC') in one litre of air that results in the emission of  $1.3 \times 10^5$  MeV of potential alpha energy in decaying to RaD. The unit is numerically equal to the total alpha-energy arising from the decay of 100 pCi/litre each of RaA, RaB and RaC, but does not depend on the state of daughter disequilibrium. Nevertheless, it is often useful to know the RaA concentration and state of daughter disequilibrium as well as the WL. The concentration of the first daughter, RaA, because of its short half-life, responds rapidly to the radon gas concentration. This information can be valuable, for example, in locating a point of injection of fresh radon and/or daughters into an airstream. With the location of such a point, corrective action can be considered. In this case, a quick, on-the-spot assessment is very helpful, as additional measurements may be indicated. For this purpose, the RaA and RaC concentrations adequately describe the state of daughter equilibrium, whilst a measurement of RaB adds only little useful information.

A prototype radon daughter monitor (RDM) has been developed with the above points in mind. We have chosen to measure only the RaA and RaC concentrations, hence the counting procedure and the calculation of results have been greatly simplified. Both sampling and measurement normally take only 11 minutes and results can be calculated using a slide rule. An important feature of the method is that electronic requirements are simple, amounting only to the scaling of gross alpha-activity.

## The Radon Daughter Monitor

Fig. 1 is a photograph of the prototype monitor. The instrument is portable and weighs 5 kg.

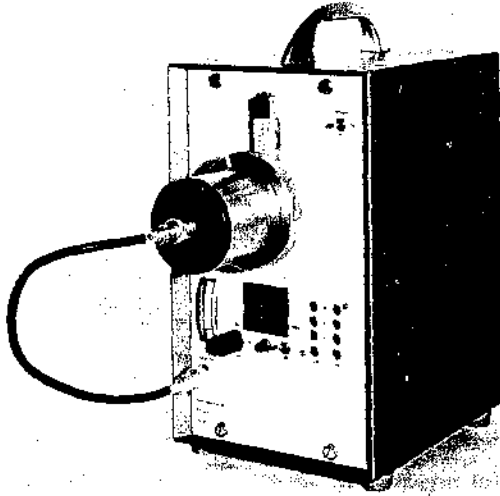


Figure 1: The Radon Daughter Monitor

### Mechanical details

Air is drawn through a glass fibre filter (GF/A, Whatman UK) at 10 litres/min. The carbon vane pump (L10, Rotheroe & Mitchell UK) is battery powered. Sampling rate is monitored and all components are mounted in a sealed case.

### Alpha-counting

Alpha activity on the filter is detected by a 450 mm<sup>2</sup> diameter surface barrier diode (ORTEC) mounted in the filter holder. The front electrode of the detector has been specially thickened to 0.5 mg cm<sup>-2</sup> of gold. This reduces sensitivity to chemical contaminants that might be picked up from mine air.

### Scaling

A charge sensitive pre-amplifier and an amplifier with an adjustable threshold are used. The discrimination level is set for a particle energy of 800 keV in Silicon. This effectively rejects beta-pulses from RaB and noise picked up from the pump motor. Integrated circuits and a binary display are used for two decades of scaling logic. Four higher decades are counted and displayed by a mechanical register (Landis & Gyr UK). A maximum count rate of 1000/sec is attained by this arrangement. Power for the circuitry is supplied by a battery of mercury cells, with a separate dry battery for the display bulbs.

### Operation of the monitor

Operation is controlled by a single switch, the sampling and counting times being measured by a watch.

### The Sampling and Counting Scheme

We have chosen a procedure that is simple to use underground. Thus, sampling times are restricted to 2, 5 or 10 minutes. Equal sampling and counting times are used. A fixed, one minute, delay between the 2 alpha-counts reduces the likelihood of timing error.

### Conversion factors

Mercer's general formulation of radon daughter decay<sup>2</sup> was used for calculations on a digital computer. Computations were checked against published data<sup>3,4</sup>.

Fig. 2 shows the build up of alpha-activity for equal sampling and counting times when equal airborne concentrations of radon daughters are collected on a

filter. RaA contributes almost half of the total alpha-activity during sampling. At short time intervals, both during and after sampling, the contribution from RaB is small.

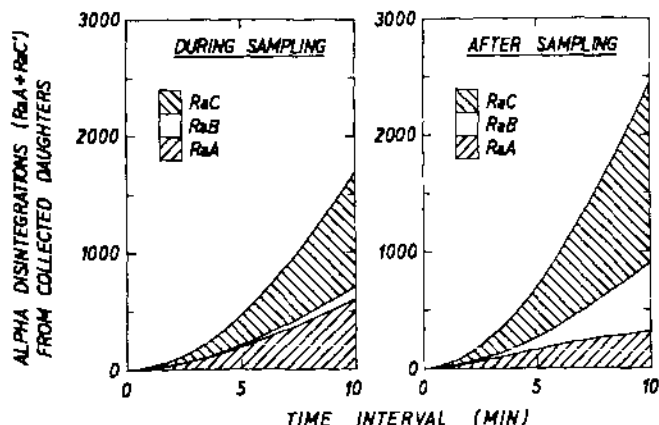


Figure 2: Alpha disintegrations from 1pO1/l. each of RaA, RaB and RaC, sampled at 10 l./min and collected on a filter. Disintegrations after sampling refer to equal sampling and counting times with a 1-minute delay in between. The envelopes of the curves give total alpha disintegrations.

In order to estimate the desired unknown concentrations, RaA and RaC, from only 2 gross alpha-counts, some assumption must be made about the RaB concentration. We have assumed that the ratio RaB/RaA is a uniquely defined function of the ratio RaC/RaA.

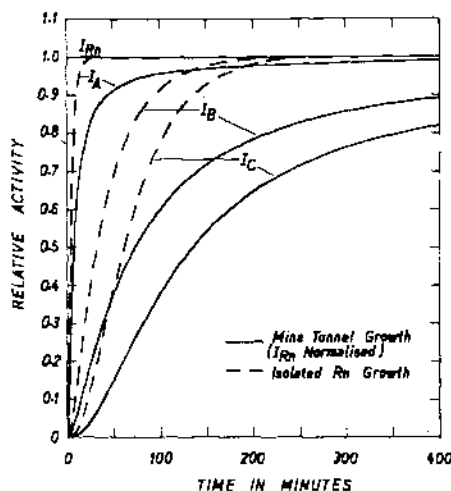


Figure 3: Relative activities of daughters and parent radon as a function of growth time, according to two theoretical models of growth.

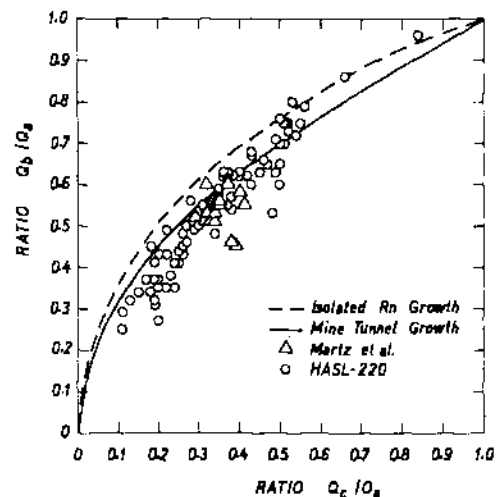


Figure 4: Ratios of radon daughter activities for two theoretical growth models. Plotted points are ratios measured in U.S. uranium mines.

The RaB approximation. Fig. 3 shows the growth of radon daughters from parent radon under 2 different conditions. The 'Mine Tunnel' model<sup>5</sup> describes growth during de-emanation of radon at a uniform rate from the walls of a mine tunnel. Air is moving through the tunnel, thus growth time is equated to transit time. The 'Isolated Radon' model<sup>3</sup> describes growth in still air. Fig. 4 shows that a different function relates RaB/RaA to RaC/RaA in the 2 models. Measured values of these ratios, from U.S. uranium mines<sup>6,7</sup>, are also plotted in the figure. The measured values are better represented by the 'Mine Tunnel' model, but they do tend to fall below this (solid) curve. Rolie<sup>8</sup> found that

similar, relatively small, departures from the simple model are predicted when 'young' and older air mix in a mine, e.g. downstream of a junction. Plate-out of unattached daughters on tunnel walls can also disturb the daughter equilibrium. However, the measurements plotted in Fig. 4 indicate that daughter equilibrium is adequately described by the solid curve. We have assumed this to be generally true. The curve is a good fit of the relationship<sup>8</sup>

$$RaC/RaA = (RaB/RaA)^2.$$

Calculated factors. Figs. 5-7 show the factors calculated to relate equilibrium ratio,  $Q_C/Q_A$ , RaA concentration,  $Q_A$ , and WL to alpha-counts recorded by the RDM. Curves are given for both growth models. Full computer tabulations are available from the authors. These allow for the slightly lower counting efficiency observed for RaA than for RaC with GF/A filters<sup>9</sup>. The calculated factors have been verified by comparing laboratory measurements with the RDM and simultaneous measurements by the modified Tsivoglou method<sup>10</sup>.

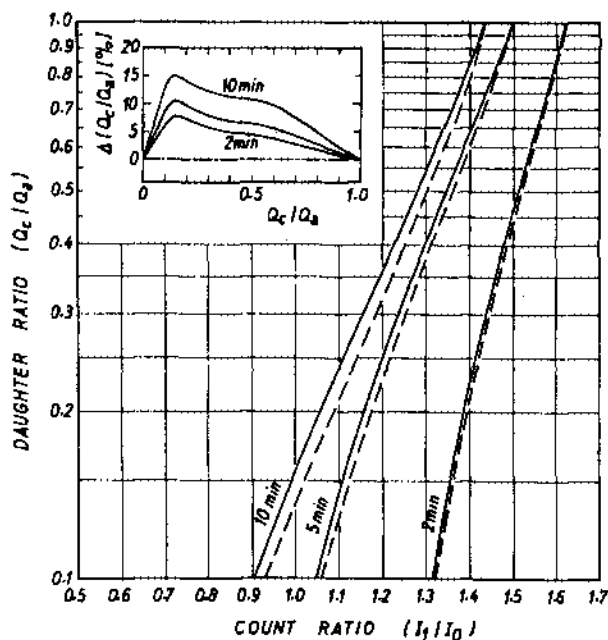


Figure 5: Curves relating the measured ratio of two alpha-counts to radon daughter ratio,  $Q_C/Q_A$ , for different sampling times. Equal counting efficiencies for RaA and RaC are assumed. Solid curves assume 'Mine Tunnel' model, dashed curves 'Isolated Rn'. Inset shows percentage systematic error in estimate of  $Q_C/Q_A$  if the wrong model chosen.

Systematic errors. The possible errors caused by departures from an assumed model are smallest for short sampling times. In Figs. 5-7, the inset figures show the magnitude of systematic errors that would arise when sampling in still air, if the 'Mine Tunnel' model were assumed to hold. These are a function of the true  $Q_C/Q_A$ . Reference back to Fig. 4 shows that similarly small errors would have been recorded for the values measured in uranium mines.

Statistical precision. Fig. 8 (a, b & c) shows the calculated coefficients of variation in estimated quantities for a range of airborne daughter concentrations. Fig. 8(a) also shows that a realistic background alpha-count of 1/min has a negligible effect on the precision of estimating  $Q_A$ . Therefore, neglecting background, the precision in an expected value, say  $Q_A$ , can be calculated as  $S(Q_A = 1)/\sqrt{Q_A}$ . Similarly, the minimum detectable concentrations, corresponding to a coefficient of variation of 0.5, can be calculated from these curves. For high radon daughter concentrations, the maximum counting rate of 1000/sec limits the sampling time. A two-minute sample is advised for concentrations in excess of 3 WL.

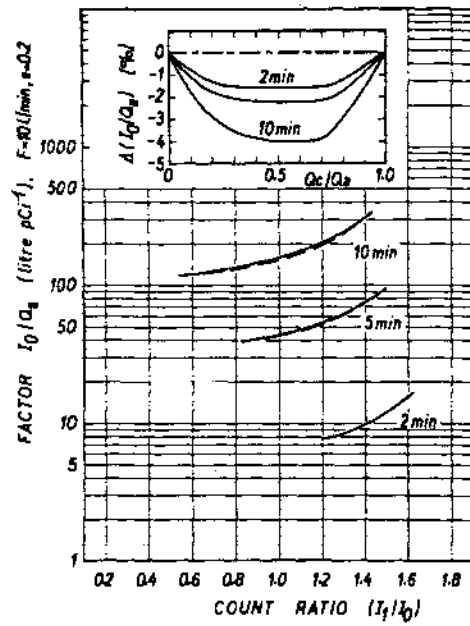


Figure 6: Factors relating count during sampling,  $I_0$ , to RaA concentration,  $Q_A$ , for different sampling times (counting efficiency 0.2). Solid and dashed curves refer to different models as in Fig. 5. Inset shows percentage systematic error arising from choice of the wrong model.

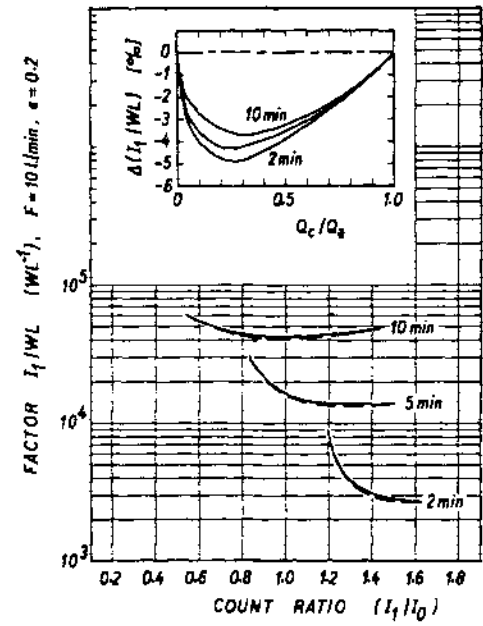


Figure 7: As Fig. 6, but showing factors relating count after sampling,  $I_1$ , to working level, WL. With a 5-minute sample and  $Q_C/Q_A \geq 0.3$ , the factor is almost constant.

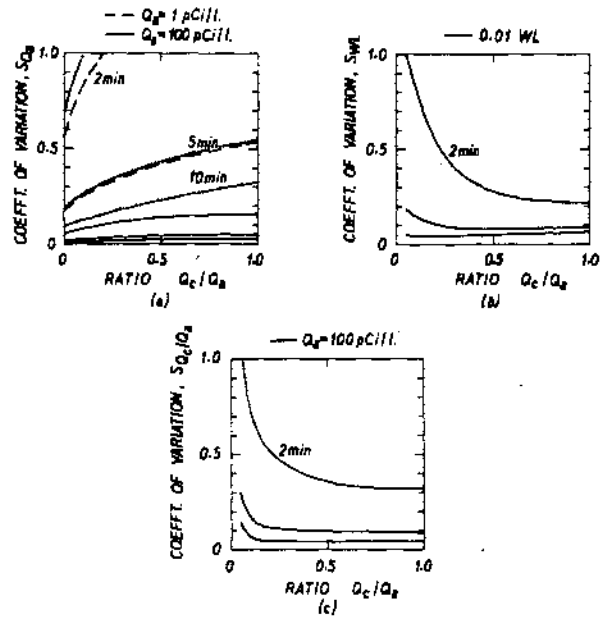


Figure 8: Statistical precision in estimates of (a)  $Q_A$ , (b) WL and (c)  $Q_C/Q_A$  as a function of daughter equilibrium for 2, 5 and 10 minute sampling times. In (a) two sets of data are given; for  $Q_A = 1$  and 100 pCi/l. Dotted curves show the effect of 1 count/min background when  $Q_A = 1$  pCi/l.

### Comparison with Other Methods

The table displays for comparison statistical precision, number of counts required for a measurement, total time and the subsequent computation for our method (RDM) and 4 established methods. Two of these methods<sup>6,10</sup> give individual daughter concentrations. The other two are presented as rapid methods, giving WL only: The Single Gross Alpha Measurement Procedure<sup>11</sup> (SGAMP) and the Instant Working Level Meter<sup>12</sup> (IWLM). Statistical precision for the RDM compares favourably with other methods, whilst the monitor offers significant advantages in field use. Under conditions of disequilibrium, both the RDM and SGAMP are subject to systematic errors. These are of the order of 10% for the SGAMP<sup>11</sup>.

TABLE 1  
COMPARISON OF METHODS FOR MEASURING RADON DAUGHTER CONCENTRATIONS\*

	Spectrometry	Modified Tsivoglou	SGAMP (Kusnetz)	IWLM	RDM
RaA	± 5%	± 12%	-	-	± 6%
RaC/RaA	± 6%	± 12%	-	-	± 9%
WL	± 3%	± 3%	± 1%	± 11%	± 1%
No. of counts	2	3	1	2	2
Time	35 min	35 min	16 min	4 min	11 min
Computation	Simultaneous equations		Slide rule	Direct Readout	Slide rule

\*Calculated coefficients of variation for 100 pCi/l. each of RaA, RaB, RaC. Sampling flow rate 10 l./min, except IWLM (3 l./min). Counter efficiency 0.2.

#### Field experience with the RDM

The monitor has undergone continuous development based on underground and laboratory comparisons with established methods. Development has reached the stage where the pump and counting system function reliably underground, even in very humid conditions. Good correlation has been obtained between routine measurements of WL with the RDM and the standard Kusnetz method<sup>13</sup>.

#### Conclusion

The radon daughter monitor described gives a rapid, comprehensive and sensitive measurement of radon daughter activity. Ease of measurement is achieved with only small and acceptable systematic errors. The complexity of the instrument has been reduced to a minimum.

#### Acknowledgements

We thank Mr. M. J. Duggan for criticism and advice and Dr. G. W. Dolphin for encouragement.

### References

1. F. E. Lurdin, J. K. Wagoner and V. E. Archer, NIOSH-NIEHS Joint Monograph No. 1, June 1971.
2. T. T. Mercer, AMA Arch. Industr. Hyg. and occup. Med., 10, 372-380 (1954).
3. R. D. Evans, Health Phys, 17, 229-252 (1969).
4. N. H. Harley and B. S. Pasternack, Health Phys, 17, 109-114 (1969).
5. J. W. Thomas and R. Epps, Personal communication (1970).
6. D. E. Martz, D. F. Holleman, D. E. McCurdy and K. J. Schiager, Health Phys, 17, 131-138 (1969).
7. A. J. Breslin, A. C. George and M. S. Weinstein, HASL-220 (1969).
8. R. Rolle, Health Phys, 23, 118-120 (1972).
9. A. C. James and G. F. Bradford, in preparation.
10. J. W. Thomas, Health Phys, 23, 783-789 (1972).
11. R. Rolle, Health Phys, 22, 233-238 (1972).
12. F. G. Croer, R. D. Evans and D. A. Gordon, Health Phys, 24, 387-395 (1973).
13. H. L. Kusnetz, Am. Ind. Hyg. Ass. Q., 17, 85 (1956).

RADON EMANATION STUDIES IN JADUGUDA  
URANIUM MINE

A.H.Khan and M. Raghavayya  
Health Physics Division  
Bhabha Atomic Research Centre  
Bombay-400085, India

Abstract

Radon gas emanating from the rock surfaces is the major source of radon in uranium mines and ventilation is the only effective means of keeping the radon levels within the acceptable limits. Quantitative estimation of the radon emanation rates is therefore essential for calculating the fresh air requirements of a working face.

Measurements of radon emanation rates have been carried out in laboratory and in the underground mine. In laboratory, the uranium ore sample is enclosed in a large air-tight glass container. Radon from the sample diffuses out and accumulates in the air volume of the container. Samples are periodically drawn from the container and radon activities are determined. The radon emanation rate is then computed from the activities obtained at different sampling intervals.

This paper describes in detail the studies conducted in the laboratory and in the uranium mines. Results obtained are presented and compared with the emanation rates published elsewhere in literature.

INTRODUCTION

The major radiation hazard in a uranium mine originates from the short-lived radon - progeny, concentrations of which mainly depend on that of the parent, radon-222. Formed within the ore body, radon enters mine atmosphere by diffusion through the rock surface. The rate of emanation is characterised by the ore grade and porosity of the rock<sup>1,2</sup>. Suction effect caused by lowering of the atmospheric pressure is another additive factor<sup>3</sup>. Although underground water<sup>4</sup> and broken ore piles also contribute substantially to the radon content of a mine drift, the main supply comes from the continuous diffusion through the ore body. Ventilation plays the most effective role in reducing the airborne radiation levels underground. Knowledge of radon emanation rate is therefore essential for an efficient and economic design of a uranium mine ventilation system.

This paper describes the efforts being made at Jaduguda to estimate the radon emanation rate in the uranium mine. The present investigations were confined to study the dependence of emanation rate on the grade of ore. Simple laboratory and field methods have been described. Emanation rates obtained in Jaduguda Uranium Mine have been compared with those reported in literature from elsewhere.



## METHOD OF EMANATION RATE MEASUREMENT

### Laboratory Experiments

Small pieces of uranium ore were placed in glass jars of approximately one litre capacity, and were covered with air tight lids provided with a stop cock. The gas emanating from the ore samples was allowed to accumulate in the free-air-space of the emanation jars. Radon levels within the jars were usually found to reach measurable concentrations in 5 to 6 hours after sealing. The radon build-up in the jar follows an exponential pattern, and reaches a constant concentration value after a period of about a month. In order to establish that our measurements were done during the near linear region of the build-up curve, the build-up was followed and was found to be fairly linear within the first 5 days. All our sampling for emanation studies were done within the first two days. Samples of air were drawn from the jar at desired intervals directly into evacuated scintillation flasks. Air drawn from the emanation jar during sampling was replaced by introducing radon free air. Sampling from each jar was repeated at suitable intervals and the concentration of radon at each sampling instant was estimated by measuring the activity in the scintillation flask after a lapse of about 200 minutes.

The rate of diffusion of radon through unit surface area of the ore piece is termed as the Emanation Rate, 'J'. It can be calculated using the following formula suggested by Thompkins et al<sup>1</sup>.

$$J = \frac{K(C_2 - C_1 e^{-\lambda t})}{3600} \times \frac{V}{A} \quad \dots\dots(1)$$

Where  $K = \frac{\lambda}{(1 - e^{-\lambda t})}$  = depletion factor,

$C_1$  = radon concentration in the accumulation volume at instant  $t_1$ , Ci/l,

$C_2$  = radon concentration in the accumulation volume at instant  $t_2$ , Ci/l,

$\lambda$  = decay constant of  $^{222}\text{Rn}$ ,  $\text{h}^{-1}$

$A$  = emanating area of rock surface,  $\text{cm}^2$

$V$  = Radon accumulation volume, l.

### Field Experiments

For field measurements underground, holes of 34 mm dia and 1 to 3 metre length were drilled in the ore body. Holes were thoroughly washed to remove loose particles of ore dust. After flushing with compressed air the holes were sealed with rubber stoppers provided with a stop cock. Radon emanating from the inner surface of the rock was allowed to accumulate in the drill hole. Samples of radon were directly collected in pre-evacuated scintillation flasks at known time intervals. Volume and surface area of the drill hole were measured. The concentrations of radon obtained within the holes at different sampling instants were used to estimate the radon emanation rate using the relation shown in equation (1).

### EMANATION RATE DATA

Radon emanation rates of twenty one ore samples from different areas of the mine were measured in the laboratory. Grade of ore in respect of each sample was estimated radiometrically. The ore grades thus obtained were grouped into discrete classes for simplicity. For instance, all values from 0.070 to 0.090 were considered as 0.08 and those from 0.090 to 0.110 as 0.10 and so on.

The mean of emanation rates of the samples corresponding to the different groups of ore grade are presented in Table-1.

Table-1

Radon Emanation Rates of Uranium Ore Samples

Grade of ore (% $U_3O_8$ )	Radon Emanation Rates(J) $\times 10^{-16}$ Ci/cm <sup>2</sup> .sec.
0.02	0.10
0.04	0.41
0.06	0.14
0.08	0.25
0.10	0.67
0.12	0.35
0.14	0.32
0.16	0.85
0.20	0.12
0.24	0.61

Range of J:  $0.10 \times 10^{-16}$  to  $0.85 \times 10^{-16}$  Ci/cm<sup>2</sup> .sec.

For field measurements, drill holes at 15 locations in the mine were chosen and the emanation rates were estimated. Rock samples were chipped off from around the individual holes and the ore grades were estimated. The values of J obtained for different groups of ore grades around the drill holes are given in Table-2.

Table-2

Radon Emanation Rate in the Mine

Grade of ore (% $U_3O_8$ )	Radon Emanation Rate(J) $\times 10^{-16}$ Ci/cm <sup>2</sup> .sec.
0.02	0.13
0.06	0.37
0.08	0.36
0.10	0.19
0.12	1.20
0.16	1.18
0.18	1.35
0.20	1.69

Range of J:  $0.13 \times 10^{-16}$  to  $1.69 \times 10^{-16}$  Ci/cm<sup>2</sup> .sec.

DISCUSSION

The radon emanation rates obtained in laboratory experiments are generally lower than those obtained under actual mining conditions. The deviations in the two ranges vary from a factor of 1.3 to 2. This variation may be attributed to a variety of reasons. One is that the ore pieces used in the laboratory studies were very small as compared to the massive ore body inside the mine. Secondly, since the mine employs exhaust type of ventilation, the barometric pressure under ground is depressed in comparison to that outside. The difference in the pressure is of the order of 32 mm of water gauge. It has been reported that reduction of pressure increases the emanation of radon.

Contrary to expectations, the attempts made to establish a precise relation between the ore grade and emanation rate did not yield any conclusive result in respect of laboratory experiments. Although no definite explanation could be given for this anomaly, it is likely that the violent forces at work during blasting might have altered the rock characteristics to varying degrees and hence the inconsistency. In case of the underground experiments, however, the emanation rate did appear to follow the ore grade, as may be seen from Figure-1.

As the atmospheric concentrations of radon in our mines are generally found to be well within the permissible limits, a comparison of emanation rate in Jaduguda mines with those elsewhere may be of interest. For this reason, emanation rates prevalent in American mines and in some soils have been compiled in Table-3.

Table-3  
Comparative Values of Emanation Rates

Area/Countries	Radon Emanation Rate J, $\times 10^{-16}$ Ci/cm <sup>2</sup> .sec.
Jaduguda Mines, India	0.13 to 1.69
New Mexico, USA, (Mines) <sup>2</sup>	500.00
Southern Utah, USA, (Mines) <sup>5</sup>	150.00
Socorro, New Mexico (Soil) <sup>6</sup>	0.90
Pelindaba, South Africa (Soil) <sup>7</sup>	0.14
Tailings used as back fill in Jaduguda	14.20

This comparison shows that radon emanation rate in Jaduguda mine is much lower than those obtained in American mines. Though the ore grade in American mines are presumably higher than that in our mine (0.07% U<sub>3</sub>O<sub>8</sub>), their emanation rates are too high to be accounted for by the ore grade alone. It is known that in New Mexico uranium occurs in sand stone and in Utah in shales. These rocks are highly porous as compared to our densely packed archaean and metamorphic rocks. The soils of socorro and Pelindaba, though having lower uranium and radium concentrations, have radon emanation rates comparable with that of Jaduguda mine. Thus, the porosity appears to affect the emanation rates of radon substantially.

Rate of radon emanation was determined by the authors from coarse uranium mill tailings, used as backfill in the mine. The rate was  $14.2 \times 10^{-16}$  Ci/cm<sup>2</sup>.sec. The radium content of these sands was of the order of 60 pCi/g while that of the ore of grade 0.07% U<sub>3</sub>O<sub>8</sub> is about 200 pCi/g. The emanation rate of this ore as seen from Figure-1 is  $0.41 \times 10^{-16}$  Ci/cm<sup>2</sup>.sec. The emanation rate from the sand is thus about 35 times that of the ore despite the radium content being only about one third. The emanation rate from the tailings would therefore be about a hundred time greater than that from the ore when normalised to equal radium content. The porosity of the sand was about 50% while the porosity of the ore was about 0.5%. The porosity ratio of the sands to the ore is therefore the same as the emanation ratios, indicating that porosity plays a far more important role in radon emanation than the ore grade alone.

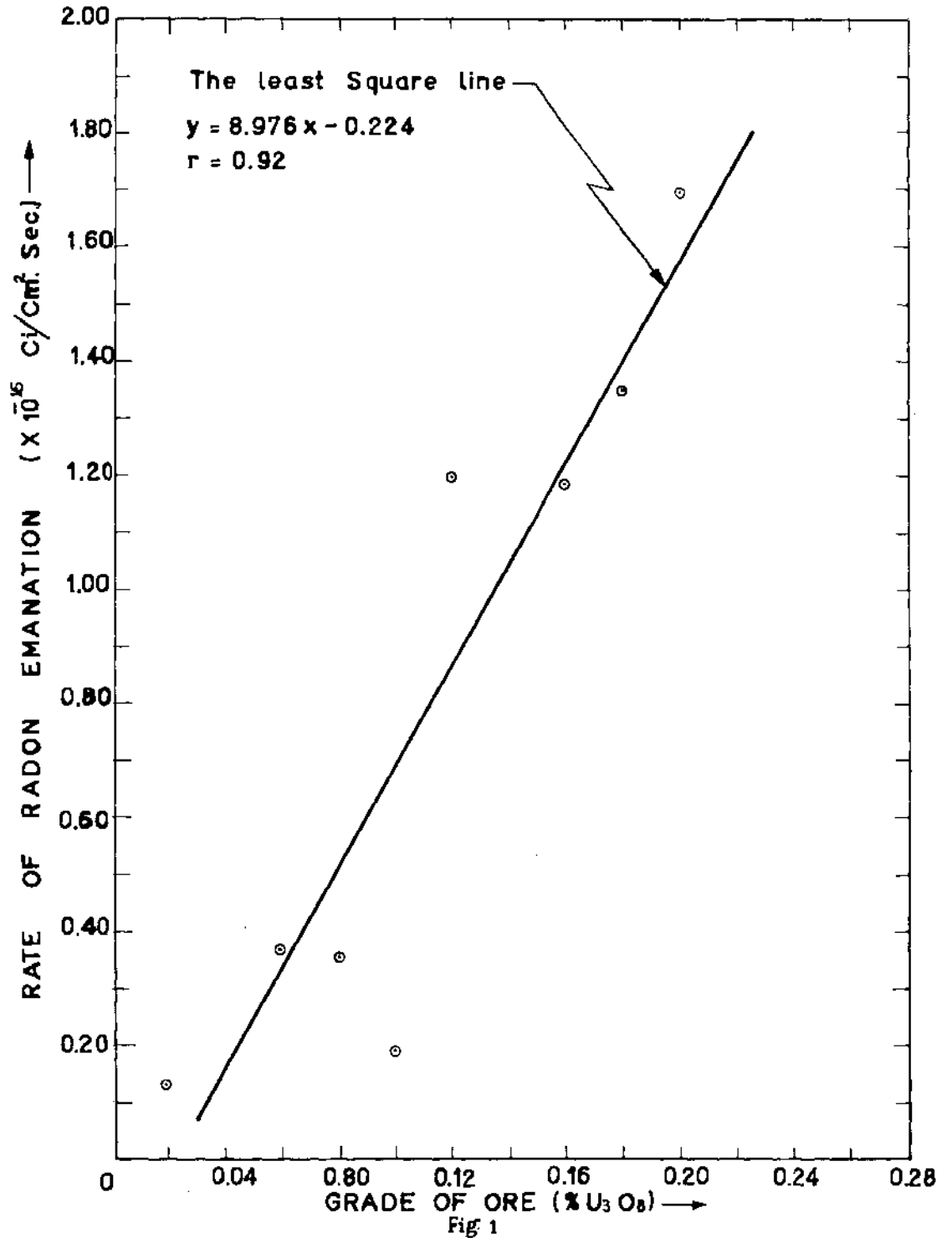
#### ACKNOWLEDGEMENT

Authors are grateful to Dr.A.K.Ganguly, Director, Chemical Group, and Mr.S.D.Soman,Head, Radiation Hazards Control Section for their interest in the work. Assistance received from Mr.K.P.Eappen is gratefully acknowledged.

#### REFERENCES

1. R.W.Thompkins and K.C.Cheng, "The Measurement of Radon Emanation Rates in a Canadian Uranium Mine", The Canadian Mining and Metallurgical Bulletin, December,1964.
2. R.W.Thompkins, "Radiation in Uranium Mines", Canadian Mining Journal, October,1970.
3. Johanna Pohl - Ruling and Egon Pohl, "The Radon-222 concentration in the Atmosphere of Mines as a Function of the Barometric Pressure", Health Physics, Vol.16, 1969(pp 579-584).
4. M.Raghavayya, "Estimation of the Concentration of Radon Dissolved in Uranium Mine Waters", Proceedings of an IAEA Regional Seminar on Radiation Protection Monitoring, held in Bombay during December 9-12,1968.
5. E.C.Tsivoglou and H.E. Ayer, "Ventilation of Uranium Mines", AMA Archives of Industrial Hygiene and Occupational Medicine, Nov.1954, Vol.10.
6. M.H.Wilkening and J.E.Hard, "Radon Flux at the Earth- Air Interfaces", Journal of Geophysical Research, Vol.65, No.10, October,1960.
7. J.K.Basson et al, "Lung Cancer and Exposure to Radon Daughters in South African Gold/Uranium Mines", Atomic Energy Board, Felindaba, Republic of South Africa, March,1971.

RADON EMANATION IN JADUGUDA URANIUM MINE



DISTRIBUTION OF AIRBORNE ACTIVITY IN  
A URANIUM MILL USING COBWEBS

Giridhar Jha and K.P. Eappen  
Health Physics Division  
Bhabha Atomic Research Centre  
Bombay-400085, India

ABSTRACT

Cobwebs found in the different sections of the uranium mill at Jaduguda were observed to trap dust particles in the respirable size range (about 60% respirable). As such these were taken to represent the dust particles inhaled by the workers. The dust collected from the cobwebs were analysed for U(nat), thorium ( $^{230}\text{Th}$ ), radium ( $^{226}\text{Ra}$ ) and polonium ( $^{210}\text{Po}$ ). The results indicate that equilibrium in the airborne activity exists only in the initial stages of operations, viz during crushing and screening. In other sections of the mill the equilibrium is found to be disturbed to different degrees.

INTRODUCTION

For a realistic estimation of inhalation hazard associated with uranium ore milling, the activity fraction of the constituent long-lived alpha emitters in respirable dust must be known. This may be accomplished by sampling and analysing the activity content in the respirable fraction of the airborne dust.

It was observed that cobwebs present at the plant site trap very fine dust particles. The purpose of the present study is to determine the size distribution of the particles thus trapped and to estimate the respirable fraction to decide whether the dust on cobweb can be taken to represent the dust inhaled by the workers. Further cobweb samples collected from different operation sites have been analysed chemically for the individual long-lived alpha emitters to study the distribution of alpha activities in the airborne dust.

EXPERIMENTAL DETAILS

The Cobweb

The cobweb found at the plant are mostly woven by an orb weaving spider "Microthema Gracilis". The cobwebs have the following properties:

Diameter of the net	- 25 - 30 cm
Spiral spacing	- 3 - 5 cm
Number of support thread	- 22 - 28
Diameter of support thread	- 1.25 - 1.75 $\mu\text{m}$
Diameter of spiral thread	- 1 - 1.5 $\mu\text{m}$
Shape of the frame	- hexagon

The cobwebs were collected from different units of the plant like Crushing and Screening, Grinding, Dewatering, Filtration and Clarification, Precipitation and Recovery and Tailings Treatment plant. Five samples were collected in each area at intervals of three months.

### Particle Size Measurements

The dust adhering to the cobweb samples was stripped using distilled water to which a few drops of 0.01 N sodium hydroxide was added. An aliquot of the dust laden liquid was transferred to an A.O. Spencer Bright Line Counting Chamber. After about thirty minutes delay (for complete settling of the suspended dust) the dust particles were sized using May's type graticules. On an average more than two hundred particles were sized in each case.

### Estimation of Respirable Fraction

In order to make the necessary calculation to estimate the respirable fraction of the dust collected on the cobweb we have followed the method suggested by Schulte<sup>1</sup>. The respirable fraction is obtained as a product of the mass frequency and the corresponding lung deposition factor obtained from the LASL curve.

### Chemical Analysis

The sample was dried and weighed. Organic material was oxidised by repeated treatment with conc. HNO<sub>3</sub>, leached and taken up in 4 N nitric acid. Estimation of U (nat), <sup>230</sup>Th, <sup>226</sup>Ra and <sup>210</sup>Po were carried out taking suitable aliquots of the prepared solution.

Percentage distribution of the long-lived alpha activities were determined by taking the ratio of the counts due to the individual isotope to that of the total.

## RESULTS

### Size Distribution and Respirable Fraction

The dust particles obtained from cobwebs were found to follow log normal distribution. Representative data on the size distribution and respirable fraction are given in Table 1 and 2.

Table-1  
Size Distribution and Respirable Fraction

Diameter		Frequency		Lung Depo- sition % (LASL Curve)	Respirable Fraction	Remarks
Projected area D <sub>p</sub> (μm)	Aerodynamic D <sub>ae</sub> (μm)	No.	Mass %			
0.68	0.75	56	1.2	100	1.2	
0.10	1.18	61	5.2	100	5.2	
1.56	1.67	54	13.0	100	13.0	
2.20	2.36	27	18.3	80	14.6	CMAD-1.07
3.10	3.33	12	23.1	55	12.7	σ <sub>g</sub> - 1.77
4.40	4.73	4	22.1	30	6.7	MMAD-2.85
6.25	6.70	1	16.8	10	1.7	

CMAD = Count Median Aerodynamic Diameter

MMAD = Mass Median Aerodynamic Diameter

σ<sub>g</sub> = Geometric standard deviation

Table-2

Statistical Parameters and Respirable Fraction  
Of Cobweb Dust at Different Operational Stages

Operation	Bulk Density of the dust	OMAD	Geometric Standard Deviation	MMAD	% respirable Schulte Method <sup>1</sup> Regression graph <sup>2</sup>	
Crushing and Screening	2.7	1.08	1.79	2.99	51.4	58
Grinding	2.7	0.99	1.95	3.75	44.0	48
Dewatering	2.7	1.12	1.79	3.00	59.9	58
Filtration and Clarification	2.7	1.18	1.69	2.70	57.6	65
Precipitation and Recovery	5.4	1.67	1.94	6.24	14.5	25
Tailings treatment	2.7	1.22	1.72	2.95	55.0	60

Distribution of Long-lived Alpha Emitters

Percentage contribution of long-lived alpha activities in air during different operations are given in Table-3 and shown in Figure-1.

Table-3

Activity Distribution of Long-lived Alpha Emitters

Operation	Percentage of gross long-lived alpha emitters			
	U(nat)	<sup>230</sup> Th	<sup>226</sup> Ra	<sup>210</sup> Po
Crushing and screening	39.50	20.50	20.30	19.70
Grinding	45.50	16.50	20.30	17.70
Dewatering	58.00	15.00	12.00	15.00
Filtration and clarification	72.00	11.30	6.40	10.30
Precipitation and recovery Sec.	91.00	6.90	1.10	0.80
Tailings treatment	30.50	21.70	23.90	23.90

DISCUSSION OF RESULTS

For calculating the respirable fraction of dust trapped in cobweb, the schulte method<sup>1</sup> has been followed and the results have also been verified by an alternative method<sup>2</sup>, following a theoretical curve between MMAD and respirable fraction for given standard geometric deviation(Table-2).

Chemical analysis of cobweb dust collected from different stages of operation indicate that during the initial stages i.e. crushing and screening radioactive equilibrium exists in the airborne activities(Table-3, Fig.1). Airborne uranium activity in this section is about the same as the rest of the radionuclides of interest. But during subsequent operations, uranium activity is predominant as compared to the rest.



Higher values of uranium in the airborne dust during grinding and dewatering stages is probably due to the addition of recycled iron cake and slurry containing significant amounts of uranium.

After leaching of the crushed ore most of the activities except uranium remain with the waste cake which is separated during filtration and sent for tailings treatment. One would expect airborne uranium in this section to be less than the other three alpha emitters considered. But in practice this was found not to be the case.

The predominance of uranium at the stages coming after filtration is easily explained, since the solutions and solids handled are rich in uranium as compared to other alpha emitters. But it is seen that the airborne uranium is higher also in the filtration section contrary to expectations. This is possibly due to cross contamination from precipitation and recovery areas; since both the sections are housed in the same building without partition between them. During tailings treatment which is separately housed, as expected uranium values are less than radium, polonium and ionium.

#### CONCLUSION

Cobweb dust sampling provides a long-term air sample of particles that are most likely to be inhaled by the workers. This has an edge over the conventional air sampling techniques with cyclone or other size discriminating devices, which are suitable only for grab sampling and hence are inadequate as far as average conditions are concerned. Analyses of cobweb samples for long-lived alpha emitters give fairly good idea about the activity distribution during different stages of operation.

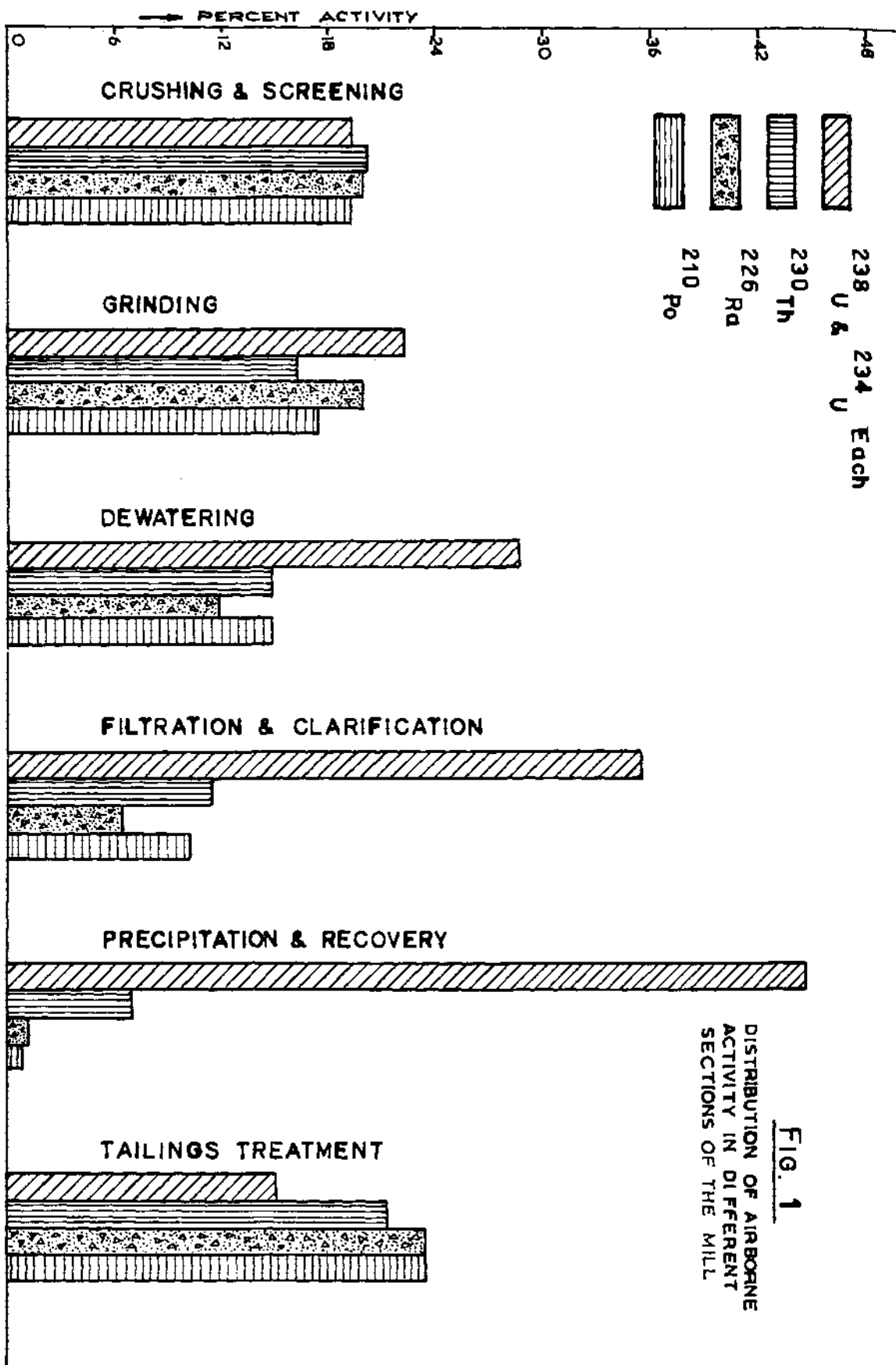
Since the  $(MPC)_a$  for gross long-lived alpha activities depends on the state of radioactive equilibrium of the constituent nuclides in air, we propose to revise the existing  $(MPC)_a$  value on the basis of the above findings.

#### ACKNOWLEDGEMENT

Our sincere thanks are due to Dr.A.K.Ganguly, Director, Chemical Group, Mr.S.D.Soman, Head, EHC Section, Dr.P.Kotrappa and Mr.M.Baghavayya for their interest and encouragement at every stage of this work. The authors wish to acknowledge the cooperation and assistance of the H.P.Unit staff at Jaduguda, Bihar.

#### REFERENCES

1. E.C.Hyatt, et al - "A Study of Two Stage Air Samplers Designed to Simulate the Upper and Lower Respiratory Tract", Proceeding of 13th International Congress on Occupational Health, (1960).
2. Kotrappa,P,et al - "HASL Cyclone, An Instrument for Measuring Aerosol Parameters for New Lung Model". To be presented at Third International Congress of the International Radiation Protection Association,(1973).



AN INSTANT WORKING LEVEL METER WITH AUTOMATIC INDIVIDUAL  
RADON DAUGHTER READOUT FOR URANIUM MINES<sup>‡</sup>

Peter G. Groer\*, Donald J. Keefe†, William P. McDowell†  
and Richard F. Selman\*

\* Radiological and Environmental Research Division

† Electronics Division

Argonne National Laboratory

Argonne, Illinois

Abstract

The Instant Working Level Meter (IWLM) evaluates the Working Level and the individual Rn-daughter concentrations in an uranium mine atmosphere within five minutes. The instrument is portable and fully automatic. The WL and the Ra A, Ra B and Ra C concentrations (pCi/liter) are displayed in digital form. Calculation of these quantities is performed by a pre-programmed CMOS calculator chip using the counts observed in the instrument's three channels (Ra A, Ra B + C, Ra C'). The Rn-daughters are collected on a membrane filter at a flowrate of 12 liter/min.  $\alpha$ -spectroscopy is performed with a silicon surface barrier detector, the  $\beta + \gamma$  - counts are detected with a plastic scintillator plus PM tube. No assumptions about Rn-daughter equilibrium are made. Only constancy of the Rn-daughter concentrations during the time of sampling (2 minutes) is assumed. The unit is entirely solid state with exception of the photomultiplier. The range of the instrument is 0.01 - 100 WL.

Introduction

The commonly used methods<sup>1-3</sup> to determine the WL (Working Level) and the short-lived Rn-daughter concentrations in uranium mine atmospheres suffer from several shortcomings. It takes a minimum of 17 minutes to complete the measurements using the fastest of these methods and in the most frequently used procedure<sup>1</sup> the Rn-daughter equilibrium and the influence of the build-up time of the activity on the resulting WL is neglected. The first attempt to solve these problems<sup>4</sup> produced an IWLM (Instant Working Level Meter) which was capable of automatic WL determination but was limited by the low air sampling rate and the high  $\gamma$  -sensitivity of its  $\beta$  -detector. The pseudo-WL due to  $\gamma$  -background has been reduced by about a factor of fifty in the instrument described in this paper. This was achieved by increasing the sampling rate to 12 liter/minute, use of a thinner scintillator (0.003 in.) and shielding of the  $\beta$  -detector.

---

<sup>‡</sup> This research was sponsored by the U.S. Bureau of Mines under Contract No. H0122106.

## Description

### Mechanical Assembly

A schematic drawing of the air sampling system is given in Fig. 1.

After a one minute background counting period and a two minute sampling period, the filter paper tape (Gelman, Acropor, pore size  $0.8\mu$ ) is moved from the air intake by spring tension to position the active area between the  $\alpha$ -detector (ORTEC, silicon surface barrier detector) and the  $\beta$ -scintillator (NE 102). After a delay of three seconds, the two minute counting period starts automatically. After this period, the WL and the RaA, RaB and RaC concentrations in pCi/liter are read out on command. After completion of the measurement the active spot is moved from the counting position and discarded. We found a self-absorption of 0.4% in the filter paper (Gelman, Acropor) using the method of J. Shapiro<sup>5</sup>. A carbon vane pump (GAST) driven by a printed circuit motor delivers a sampling rate of about 12 liter/minute. A tachometer senses the revolutions per minute and is part of a feedback system ensuring constant flow-rate.

### Electronic System

The electronic circuitry consists of three major subsystems: the detection subsystem, a control-computer subsystem and a power subsystem. (See functional block diagram.) The detection subsystem is further broken down into the  $\alpha$ -detection channel and  $\beta$ -detection channel, the pump motor regulator and drive circuit, the high voltage regulator and the solenoid actuator for the paper drive system.

Detection Subsystem. The  $\alpha$ -detection channel consists of a surface barrier detector, a high gain charge sensitive preamplifier, a pulse amplifier, and a single channel analyzer to separate RaA from RaC'. The overall  $\alpha$ -detection gain is 5 volts per pC.

The  $\beta$ -detection channel consists of a 10 stage low noise high gain photomultiplier, a NE 102 scintillator, a high gain charge sensitive preamplifier and a discriminator.

Power Subsystem. The power subsystem consists of 13 rechargeable Gel cell batteries and a connector which selects either individual cells for the external charging circuit or combines the batteries into the power pack which consists of an 18 V, 3 Ah battery for operation of the pump, a 6 V, 3 Ah battery for the digital components and  $\pm 12$  V, 1 Ah batteries for the linear components.

Control-Computer Subsystem. The control section of the control-computer subsystem consists of a CMOS driven sequential control circuit which applies the timing pulses for the fully automatic operation.

Also included in the control section are three burst generators, one for each output (RaA, Ra (B+C) and Ra C'), used to generate double pulses for each single input pulse during the background counting period. These doubled background counts are subtracted from the normal sample count through the use of

up-down counters (accumulators) to compensate for the  $\gamma$ -background. Gamma shielding is achieved through the use of a lead shield (see Fig. 1) and by positioning the batteries around the detector section. The control section also controls the input gating, signal routing and digital resets of the computer section to provide the proper compensated two minute sample count to the accumulators.

The computing subsystem is essentially a pre-programmed buss oriented digital processor. This processor accepts digital data from the accumulators, combines it with various stored constants and from this data calculates Working Level and the individual Rn-daughter concentrations. The processor consists of a MOS calculator circuit, a program memory, a constant memory, a system clock generator, accumulators, calculator driver, display driver, and display as shown in Fig. 2.

The entire circuit except for the memories is assembled from CMOS digital integrated circuits which offer very low power consumption and high immunity to electrical noise.

In order to understand the operation of the circuit, a brief explanation of it's component parts follows:

- 1) The program control section consists of two programmable read only memories (PROM's) which store the program steps, a program counter which advances once for each program step, and a 4 line to 16 line decoder which translates part of the digital word from the memory into individual commands such as add, enter accumulator A, enter constant 5, multiply, etc. The data lines from the PROM's are also used to select the location of a particular constant in the constant memory.
- 2) The constant memory consists of two PROM's which hold the 12 constants required for the calculations; a memory address register which locates and clocks out the constant requested by the program; a set of clock controls; and a set of transmission gates which tie the memories to the data buss. The constant consists of six information words which may include a decimal point. The use of PROM's allows a field change of constants should changes in counting efficiency or flowrate alter the equations.

The information is entered onto the data-buss in a bit-parallel, digit serial fashion. The system clock provides the timing information required to serialize the data. In a particular timing sequence, C0 through C15, the clock pulse C1 steps the program memory register to the next location. If the command at this location calls for entering data from an accumulator, the constant memory or the flowrate correction switch, then clock pulses C2 through C15 enter the data into the calculator. If the new program step called for an operation such as multiply or add, then this operation would be entered into the calculator at time C1. When C1 appears for the second time, the program again advances and new data is entered or a new operation performed.

The clock generator consists of an oscillator driving a binary scaler

whose output is decoded in a 4 line - 16 line decoder to generate clock lines  $C_0 - C_{15}$ . These lines are then routed to the proper locations in the system.

The data buss is terminated in the calculator driver card which shifts the voltage levels from those required by the PROM's to those required by the calculator circuit. After the level shifting, the data buss is decoded in a 4 line to 10 line converter to obtain digit information for the calculator while a separate conversion is performed to obtain the decimal point information. These decoded data are then used to control information inputs to transmission gates which enable the calculator inputs. The calculator is interfaced to a Light Emitting Diode (LED) display for the presentation of data.

### Theory and Calibration of the IWLM

The WL is a linear combination of the short-lived Rn-daughter concentrations as shown in the following equation:

$$WL = 1.052 \times 10^{-4} N_A + 5.908 \times 10^{-5} (N_B + N_C) \quad (1)$$

Therefore, the three unknowns  $N_A$ ,  $N_B$ ,  $N_C$  (atoms/liter) have to be determined to evaluate the WL. This is done by relating these quantities to the counts observed in the three channels of the instrument, as shown below:

$$\begin{aligned} A &= 0.580386 E_A V N_A \\ B + C &= (0.036204 E_B + 0.001584 E_C) V N_A + \\ &+ (0.098134 E_B + 0.006941 E_C) V N_B + \\ &+ 0.131000 E_C V N_C \\ C' &= (0.001584 N_A + 0.006941 N_B + 0.131000 N_C) E_A V \end{aligned} \quad (2)$$

- A =  $\alpha$ -counts in RaA - channel
- B+C =  $\beta$ + $\gamma$  -counts from RaB and RaC
- C' =  $\alpha$ -counts in RaC' - channel
- V = flowrate (liter/minute)
- $E_A$  = detection efficiency for RaA and RaC'
- $E_B$  = detection efficiency for RaB
- $E_C$  = detection efficiency for RaC

The numerical coefficients in (2) follow from the laws of radioactive series decay. The half-lives used are:

$$\begin{aligned} \text{RaA: } T_{1/2} &= 3.05 \text{ min.} \\ \text{RaB: } T_{1/2} &= 26.8 \text{ min.} \\ \text{RaC: } T_{1/2} &= 19.7 \text{ min.} \end{aligned}$$

For example, the numerical coefficient (0.580386) in the equation for A follows from:

$$(1 - \exp(-\lambda_A t_B)) \exp(-\lambda_A/20) (1 - \exp(-\lambda_A t_D)) / \lambda_A \quad (3)$$

with:

$$\begin{aligned} \lambda_A &= \text{decay constant of RaA} = 0.227621 \\ t_B &= 2 \text{ min. sampling time} \\ t_D &= 2 \text{ min. counting time} \end{aligned}$$

The first term in (3) describes the build-up, the second the decay of the RaA activity during the 3 sec. delay and the third the accumulation of counts during the counting period. The analogous coefficients for daughter and granddaughter products are more complex and are not given here.  $E_A$  is determined by comparison with a calibrated hemispherical gas-flow proportional counter.  $E_B$  and  $E_C$  are calculated in the following manner. First  $N_A$ ,  $N_B$  and  $N_C$  are determined from the  $\alpha$ -counts observed using the following equations:

$$\begin{aligned}
 N_A &= 0.926838 E_A V A(5) \\
 N_B &= (-0.879403 A(5) - 11.12606 C'(5) + 2.752840 C'(30)) E_A V \quad (4) \\
 N_C &= (0.049957 A(5) + 4.232080 C'(5) - 0.251541 C'(30)) E_A V \\
 A(5) &= \text{RaA counts observed during five minutes starting three} \\
 &\quad \text{seconds after the end of the two minute sampling time.} \\
 C'(5) &= \text{RaC' counts observed during the same time interval as above.} \\
 C'(30) &= \text{RaC' counts observed during thirty minutes starting three} \\
 &\quad \text{seconds after the end of sampling.}
 \end{aligned}$$

The numerical coefficients in (4) follow again from the laws of radioactive series decay. Since their derivation is straightforward but lengthy, it is not given here. With  $N_A$ ,  $N_B$  and  $N_C$  known  $E_B$  and  $E_C$  can be determined from the following equations:

$$\begin{aligned}
 BC(5) &= (0.127907 N_A + 0.236138 N_B) V E_B + \\
 &\quad + (0.010200 N_A + 0.028418 N_B + 0.311000 N_C) V E_C \\
 BC(30) &= (0.981722 N_A + 1.050628 N_B) V E_B + \\
 &\quad + (0.385895 N_A + 0.478116 N_B + 1.256959 N_C) V E_C \quad (5) \\
 BC(5) &= \text{total } \beta \text{-counts observed during five minutes starting three} \\
 &\quad \text{seconds after the end of sampling.} \\
 BC(30) &= \text{total } \beta \text{-counts observed during thirty minutes starting} \\
 &\quad \text{three seconds after the end of sampling.}
 \end{aligned}$$

With  $E_A$ ,  $E_B$  and  $E_C$  so determined, equations (2) can be inverted. Properly scaled the inverted equations give the Rn-daughter concentrations in pCi/liter and the WL as a linear combination of A, B + C and C'. These inverted equations are programmed in the calculator subsystem of the IWLM. It is clear from this description of the calibration that the IWLM determines the Rn-daughter concentrations and the WL without any assumptions about Rn-daughter equilibrium. Since all weighing coefficients are strictly proportional to  $1/V$ , a flow-rate variation can be easily corrected for. This correction is accomplished by setting the ratio (calibration flowrate/observed flowrate) on a thumb wheel switch indicated in Fig. 2. Recalibration of the IWLM, if used at different elevations or with different flowrates, is therefore unnecessary.

### Tests

We tested a prototype IWLM in the experimental Dakota Mine in New Mexico. The results are shown below:

Test	IWLM				$\alpha$ -Spectroscopic Method (See Eq. (3))				Kusnetz Method
	WL	RaA (pCi/litre)	RaB (pCi/litre)	RaC (pCi/litre)	WL	RaA (pCi/litre)	RaB (pCi/litre)	RaC (pCi/litre)	WL
1	0.88	173	85	68	0.83	171	80	66	0.85
2	1.44	231	129	140	1.51	233	150	137	1.58
3	1.11	219	104	92	1.12	217	109	91	1.12
4	1.47	292	149	105	1.44	296	146	107	1.43
5	0.71	110	69	63	0.76	142	73	64	0.71
6	0.32	74	33	21	0.31	76	30	20	0.31
7	0.52	89	62	28	0.40	90	38	29	0.44

WL, RaA and RaC concentrations determined by the IWLM and the spectroscopic method are in good agreement. The same is true for the RaB concentrations in most cases. The reasons for deviations like in Test 2 and 7 are not clear. More tests are needed to clarify these discrepancies.

#### References

1. H. L. Kusnetz, Am. Ind. Hyg. Assoc. Quart. 17, 85 (1956).
2. E. C. Tsivoglou, H. E. Ayer and D. A. Holaday, Nucleonics 11, 40 (1953).
3. J. Thomas, Health Physics 19, 691 (1970).
4. P. G. Groer, R. D. Evans and D. A. Gordon, Health Phys. 24, 387 (1973).
5. J. Shapiro, Univ. of Rochester, Report UR-298 (1954).

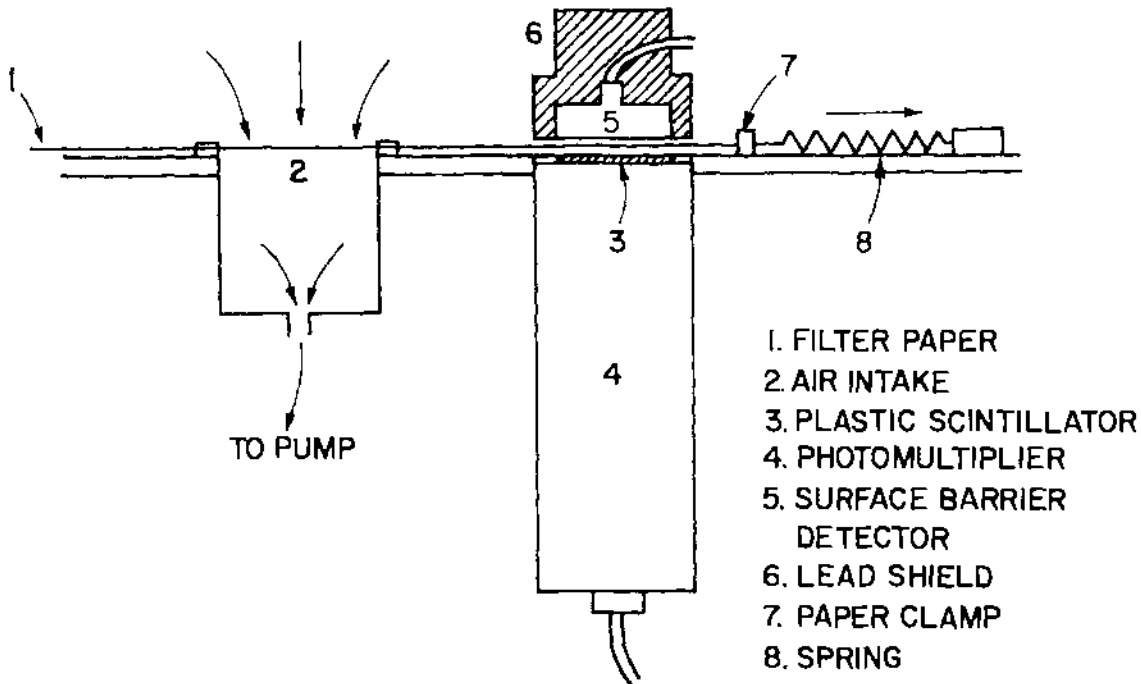


Fig. 1 Detection, air sampling and paper transport mechanisms of the IWLM.



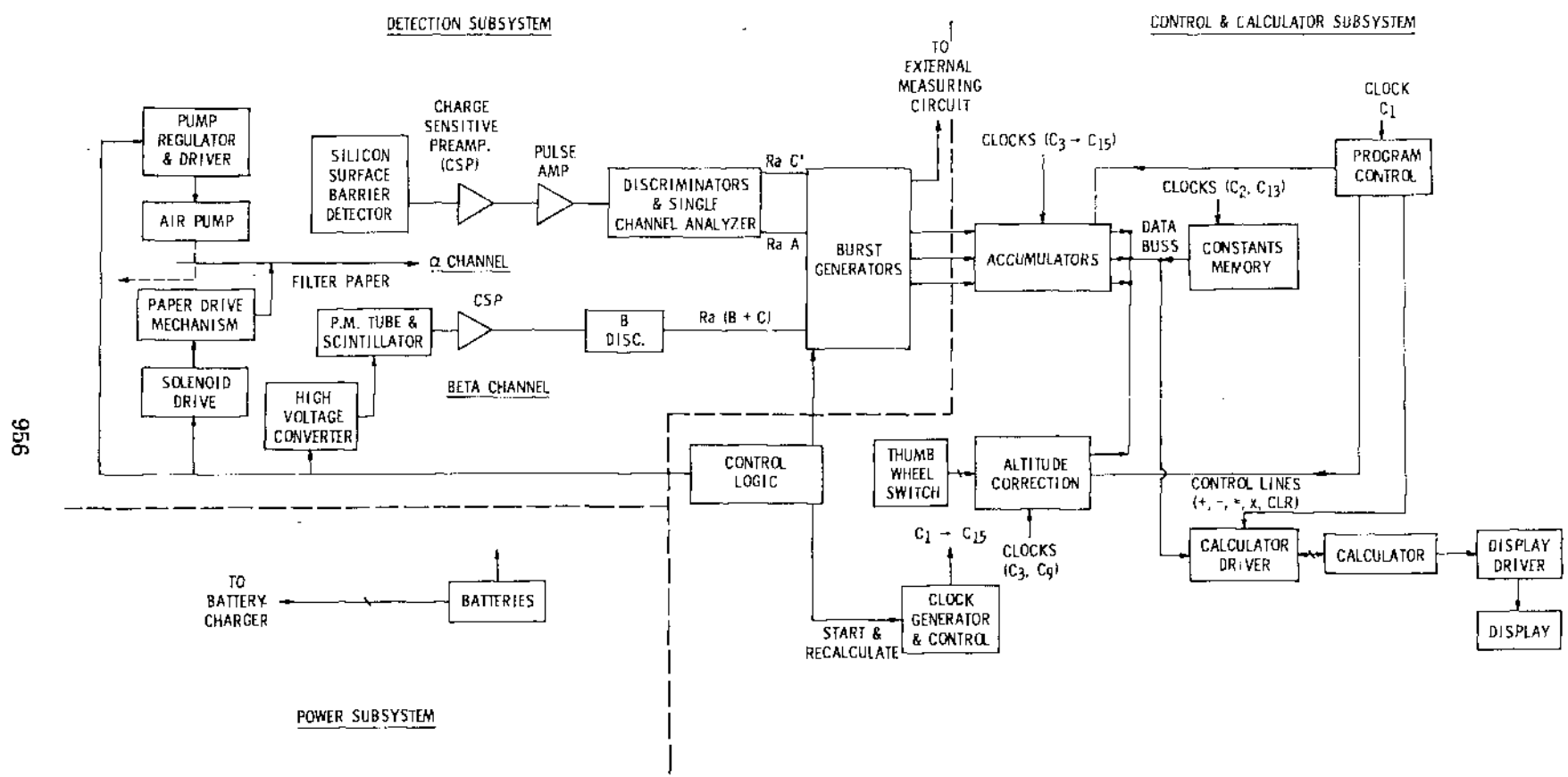


Fig. 2 Functional block diagram of the IWLM.

## IN VIVO MEASUREMENTS

### MONITORING OF LOW-ENERGY X-RAY RADIONUCLIDE CONTENT IN HUMAN BODY

E.I. Dolguirev, G.N. Kajdanovsky, N.V. Porozov, V.M. Shamov

Institute of Radiation Hygiene

Leningrad

USSR

#### Abstract

A 15 cm -dia 0.1 cm thick NaI detector and an Ar-CH<sub>4</sub> proportional counter with a 300 cm<sup>2</sup> window are used for in vivo measurements of <sup>210</sup>Pb, <sup>239</sup>Pu and <sup>241</sup>Am content in human body. The subject's background was determined from the correlation of the counting rates in two channels, e.g. for <sup>210</sup>Pb: 30-55 and 100-150 keV, respectively. The detectors were calibrated on an anthropomorphic phantom with a thickness of the tissue absorption layer varying from zero to 4 cm.

Calculation methods for nuclide content in critical organs and cases illustrating different types of radionuclide distribution in human body are given.

#### Introduction

The amount of internally deposited radionuclides in personnel is accepted by present-day international and national standards as one of the basic criteria used in the sphere of radiation protection. However, the evaluation of human body burdens of incorporated <sup>210</sup>Pb, <sup>239</sup>Pu and <sup>241</sup>Am is materially complicated by the difficulties involved in obtaining data on the distribution of these radionuclides in individual critical organs. The difficulties involved in detection are due to a number of factors and particularly, to the low-energy radiation of radionuclides assessed. The possibility of obtaining such information is determined to a large extent, by proper choice of techniques of in vivo and calibration measurements which depend on the pattern of radionuclide distribution among and in the subject's organs.

There are three main types of distribution of the above radionuclides in human organism: lung, osteohepatic and a combined one. The specific pattern of distribution is determined by the rhythm and route of the nuclide administration, the chemical form of the deposited compound and some other factors. The combined type of radionuclide distribution is the most general and complicated one, as far as direct measurements are concerned.

Many important aspects of in vivo assessment of low-energy radionuclide content in human body have been discussed in the papers<sup>1-6</sup>. This work deals with the methods and illustrations of measurement procedures for <sup>210</sup>Pb, <sup>239</sup>Pu and <sup>241</sup>Am for different

patterns of their distribution in the body.

### Equipment

The unit comprises a detection system and a measurement panel with channels for registering energies in the ranges of 10-25, 30-55, 35-70 and 100-150 keV. The detection system consists of two proportional 7.5 cm Ar-CH<sub>4</sub> gas filled counters with a 300 cm<sup>2</sup> window each and two scintillation counters with 15 cm-dia 0.1 cm thick crystals of NaI(Tl). The counter window panes are made of 0.03 cm-thick beryllium plates. The counters are mounted in an iron chamber 2x1.5x1.5 m with a wall thickness of 15 cm. The scintillation counter background is 20 cpm (10-25 keV) and 65 cpm (35-70 keV). The proportional counter background is 3 cpm (10-25 keV). The counter sensitivity is characterized by the magnitude of calibration coefficients (see Fig.1).

### Prediction of the Subject's Background

The sensitivity and accuracy of in vivo measurements of low-energy internally-deposited nuclides are determined, to a great extent, by the individual subject's background level and precision of its measurement. It is impossible to measure directly the specific background in a given contaminated subject because the continuous distribution of pulses of the diffused radiation of high-energy radionuclides <sup>40</sup>K, <sup>137</sup>Cs, etc. and the photopeak of the low-energy radionuclide measured, e.g. <sup>210</sup>Pb, <sup>239</sup>Pu, <sup>241</sup>Am are registered in the same energy interval. We suggested a method of prediction of the subject's background which is based on the probable correlation between the background counting rate in the main channel where the photopeak of the nuclide measured is registered, and the background counting rate in the reference channel, where this peak does not occur. The correlation between the background counting rates in these two channels was derived from our measurements carried out in a group of people of the different build and with different body contents of high-energy radionuclides. The experimental data processed by the method of least squares were approximated by the following equation:

$$N^M = \alpha + \beta N,$$

where  $N^M$  - expected background counting rate, cpm, in the main channel in the energy interval of  $E_1 - E_2$ ;  $N$  - registered counting rate in the reference channel in the energy interval of  $E_3 - E_4 = 100 - 150$  keV;  $\alpha$  and  $\beta$  - parameters whose numerical values for different energy intervals are given in the Table:

Radionuclide	$E_1 - E_2$ keV	Position of Detector	$\alpha$	$\beta$
<sup>210</sup> Pb	30 - 55	Over chest	42.5	1.37
		Over skull	14.80	0.50
<sup>239</sup> Pu	10 - 25	Over chest	9.7	0.19
		Over skull	0.19	0.33
<sup>241</sup> Am	35 - 70	Over chest	18.6	1.33
		Over skull	11.5	0.88

### Methods

Since the radionuclides of lead, plutonium and americium are

generally deposited in the lungs, liver and skeleton. The detectors were placed over the lungs and the liver in front of the subject's chest and over the frontal bone of the skull, a location most convenient for skeletal measurements.

The body contents of  $^{210}\text{Pb}$  and  $^{241}\text{Am}$  were measured by scintillation counters, while that of  $^{239}\text{Pu}$  - by both scintillation and proportional counters.

The set of equations for the values of the activity of the radionuclide deposited in each of the critical organs is:

$$J_i = \sum_{j=1}^4 q_j \varepsilon_{ij} \quad (i=1,2,3,4) \quad (\text{Eq.1})$$

where  $J_i$  - the counting rate of the detector over the  $i$ -organ, which is determined by the measured nuclide radiation;  $q_j$  - the desired activity of the radionuclide deposited in the  $j$  - organ;  $\varepsilon_{ij}$  - calibration coefficient which is numerically equal to the counting rate measured by the detector over the  $i$ -organ for unit activity of the radionuclide in the  $j$ -organ of the phantom. The indices used of the equation set (Eq.1) are: 1 - the right lung 2 - the lung; 3 - the liver and 4 - the skeleton.

The value  $J_4$  includes the counting rate measured over the skull, while  $\varepsilon_{44}$  - the same for unit activity of the radionuclide content in the entire skeleton.

Radionuclide quantum radiation was measured in the intervals: 10 - 25 keV for  $^{239}\text{Pu}$ ; 10-25 keV and 35-70 keV for  $^{241}\text{Am}$  and 30-55 keV for  $^{210}\text{Pb}$ . The calibration coefficients  $\varepsilon_{ij}$  (Fig.1) were obtained by means of an anthropomorphous phantom which comprised the skull, trunk and arms and legs.

Since the actual distribution of deposited radionuclides involves the values of  $\varepsilon_{41}$ ,  $\varepsilon_{42}$  and  $\varepsilon_{43}$  close to zero, these values were disregarded in the calculations, too.

Solutions to the equation set (Eq.1) were found from  $q_j = \Delta_j / \Delta$  where  $\Delta$  is the determinant of the system and  $\Delta_j$  is the determinant of the undetermined value  $q_j$ .

The fourth equation was excluded from the set of equations (Eq.1) and the terms containing  $\varepsilon_{14}$ ,  $\varepsilon_{24}$  and  $\varepsilon_{34}$  - from the three first equations, when  $^{239}\text{Pu}$  content was calculated, on the assumption that the X-ray radiation of  $^{239}\text{Pu}$  is nearly completely absorbed by skeletal bones.

In assessing  $^{210}\text{Pb}$  or  $^{241}\text{Am}$ , the counting rate  $J_4$  determined by the measured radionuclide radiation was found to be equal to that registered by the detector less the background value. In the measurements of  $^{239}\text{Pu}$  content this equation may be upset due to the presence of the impurities of  $^{241}\text{Am}$ , the energies of the X-ray lines of which lie within the range of  $^{239}\text{Pu}$  radiation. In such case, the values of  $J_i$  were obtained from the expression:

$$J_i = I_i - \sum_{j=1}^3 q_j^{Am} \varepsilon_{ij}^{Am} \quad (\text{Eq.2})$$

where  $I_i$  - the counting rate detected over the  $i$ -organ less the background value;  $q_j^{Am}$  - the amount of the activity of  $^{241}\text{Am}$  deposited in the  $j$ -organ;  $\varepsilon_{ij}^{Am}(10-25)$  - calibration coefficients in the interval of 10-25 keV obtained experimentally as a result of insertion of  $^{241}\text{Am}$ -emitters into the phantom organs. Values for  $q_j^{Am}$  were computed from the data of measurements of the gamma-radiation of  $^{241}\text{Am}$  in the energy interval of 35-70 keV.

### In Vivo Measurements Examples

#### Case of Lung Distribution of $^{241}\text{Am}$

Four people were exposed to the radiation from an americium

source as a result of a failure of containment during experiments. The nuclide was inhaled in the form of insoluble americium dioxide. 0.4 to 2.2 nCi<sup>241</sup>Am was found in the lungs. Fig.2 shows the spectrum of <sup>241</sup>Am radiation recorded over the lungs of one of the subjects three months after the exposure. Distinct gamma-spectra recorded over the subject's head were detected at the beginning of measurements only (Fig.3). As they were not detected after repeated decontamination measures had been taken, it was an indication of the surface contamination of the skin and hair. Therefore, it provides evidence that it was a case of lung distribution of <sup>241</sup>Am.

#### Case of Osteohepatic Distribution of <sup>210</sup>Pb

The measurements were carried out in a group of volunteers who took a hydrochloric acid solution of <sup>210</sup>Pb, at pH = 3. The gamma-spectra recorded over the subjects' head and liver are given in Fig.4 and 5. The activity Q deposited in the whole skeleton was assessed with due regard to the equation:  $Q = q \cdot k(t)$ , where q is the activity deposited in the skull; k(t) is the coefficient of conversion of the skull activity to that of the skeleton which allows for both the ratio of the masses of the skull and the whole skeleton and the nuclide distribution in the compact and trabeculate tissue portions of skeletal bones. The value k(t) varies with time from 16.5 on the first day of radio-nuclide administration to 12.3 on the 100th day, owing to the continuous re-distribution of bone-seeking elements.

The measurement results showed the skeleton/liver ratio to be 3 to 4 in the average of four subjects on the 100-th day of <sup>210</sup>Pb administration, which points to the skeletal distribution of lead chloride in human body.

#### Case of Combined Lung-Osteo-Hepatic Distribution of <sup>239</sup>Pu and <sup>241</sup>Am

Measurements in man usually fail to produce statistically significant spectra of plutonium and americium radiation. Such spectra, however, may be obtained by long-term measurements, if the thickness of the muscular, fat and skin tissues of the subject is not great. Fig.6 shows the spectra recorded by a scintillation counter placed over the lungs, liver and skull of the subject. The peaks of the pulse amplitude distribution corresponds to the energies of 17 keV and 60 keV, which indicates at the incorporated plutonium and americium. Fig.7 shows the spectra obtained by means of a proportional counter placed over the skull and the right lung of the same subject. It also shows three characteristic peaks which make it possible to identify the X-ray radiation of plutonium with energies of 13.6 keV, 17.4 keV and 20.4 keV respectively. The said spectra provided evidence that this is a typical case of the lung-osteo-hepatic type of radio-nuclide distribution.

#### References

- 1.R.Ehret, H.Kiefer, R.Maushart and G.Möhrle, Assessment of Radioactivity in Man, Vol.1, I.A.E.A., Vienna, 1964.
- 2.D.Ramsden and R.Speight, AEEV-R494, 1967.
- 3.B.Taylor, Health Physics, v.17, No.1, 1969.
- 4.T.Tomitani and E.Tanaka, Health Physics, v.18, No.3, 1970.
- 5.J.Rundo, K.Rudran and B.Taylor, Health Physics, v.17, 1969.
- 6.M.Eisenbud et al., Health Physics, v.16, 1969.

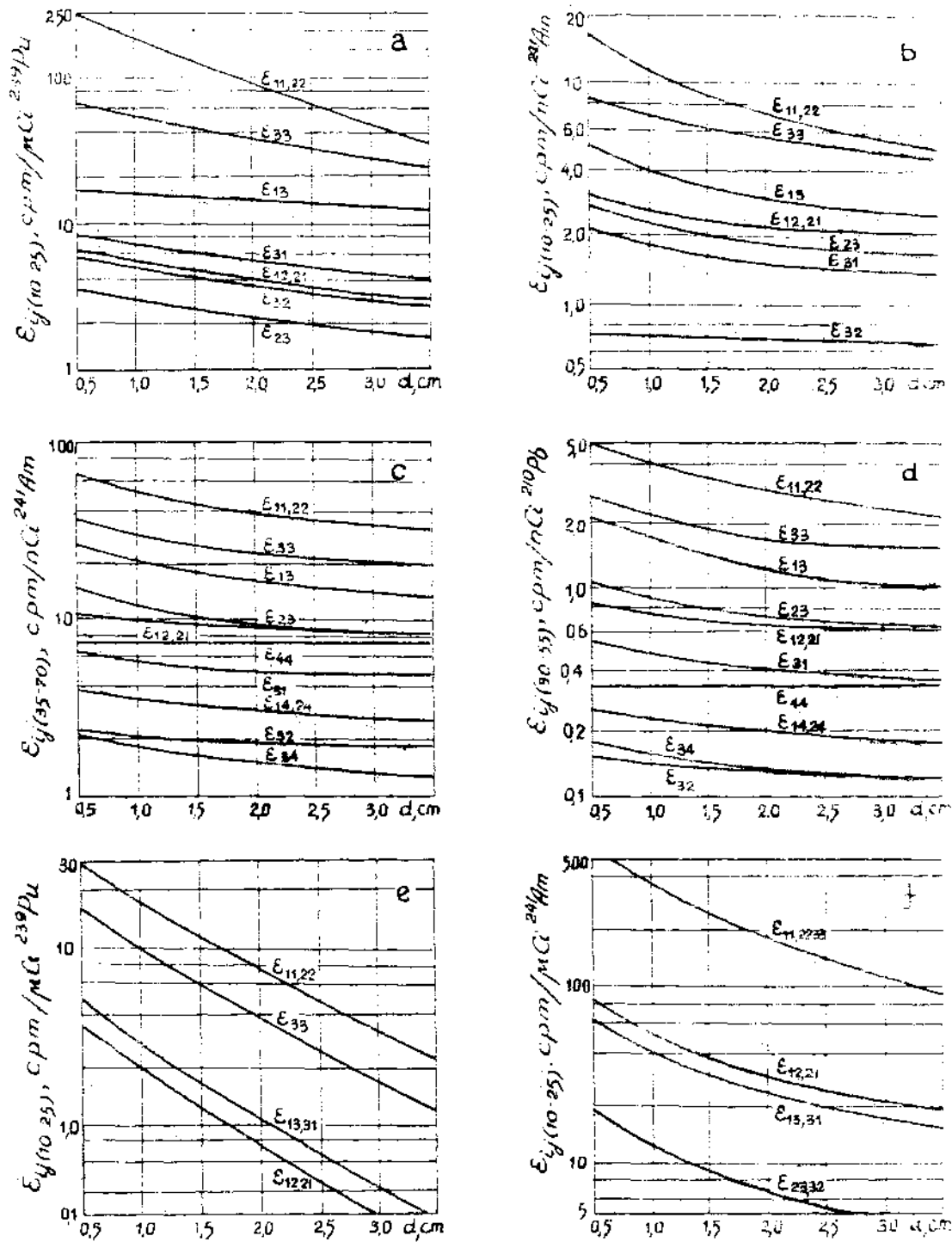


Fig. I. Calibration coefficients  $\epsilon_{ij}$  vs soft tissue thickness for scintillation counter in the energy range: a) 10-25keV  $^{239}\text{Pu}$ , b) 10-25keV  $^{241}\text{Am}$ , c) 35-70keV  $^{241}\text{Am}$ , d) 30-55keV  $^{210}\text{Pb}$  and for proportional counter in the energy range 10-25keV: e)  $^{239}\text{Pu}$ , f)  $^{241}\text{Am}$ .

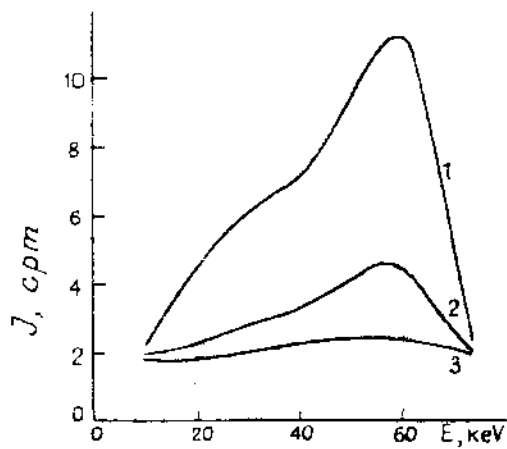


Fig. 2. Spectra  $^{241}\text{Am}$ : 1-fantom (10nCi), 2-contaminated subject (2nCi), 3-control subject.

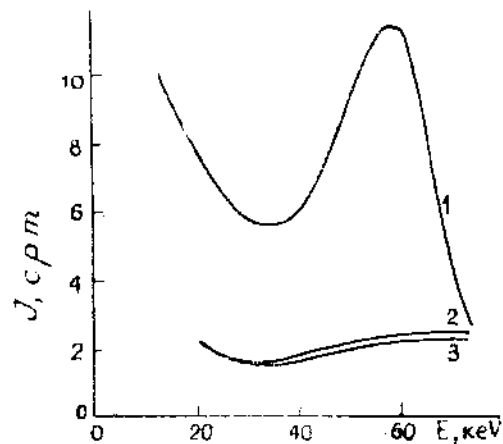


Fig. 3. Spectra  $^{241}\text{Am}$ : 1-contaminated 3.10.72; 2-6.22.72; 3-control subject.

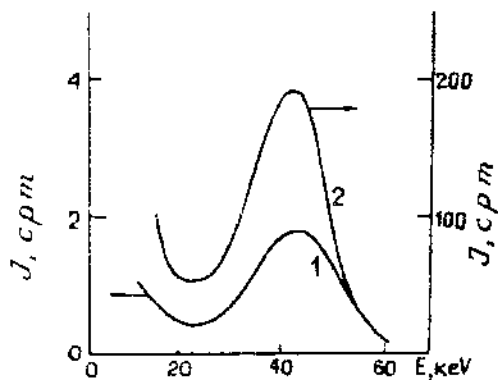


Fig. 4. Net spectra  $^{210}\text{Pb}$ : 1-over head contaminated subject; 2-over skull of fantom.

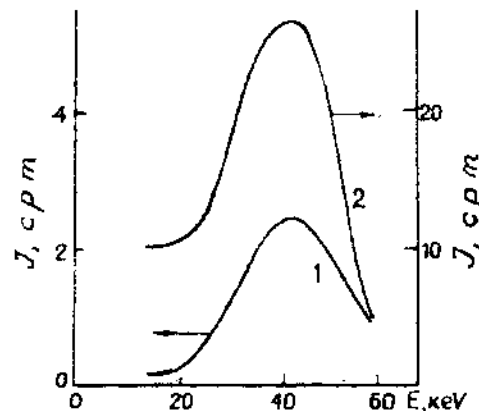


Fig. 5. Net spectra  $^{210}\text{Pb}$  over liver: 1-contaminated subject, 2-fantom.

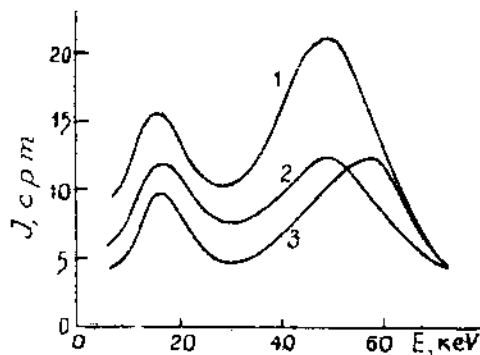


Fig. 6. Spectra Pu+Am over: 1-right lung, 2-liver, 3-Head of contaminated subject.

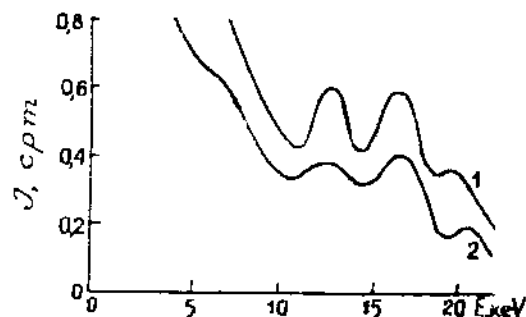


Fig. 7. Spectra Pu+Am over: 1-right lung, 2-head contaminated subject.

ОПТИМИЗАЦИЯ УСЛОВИЙ ГАММА-СПЕКТРОМЕТРИЧЕСКИХ ИЗМЕРЕНИЙ  
В РАДИАЦИОННОЙ ЗАЩИТЕ

О.В. Лебедев, В.М. Малыгин  
Институт радиационной гигиены  
Ленинград  
СССР

Аннотация

На основе нового критерия радиометрического качества и анализа параметров около 100 спектрометров гамма-излучения человека рассматриваются вопросы оптимизации измерения радиоактивности препаратов и организма человека.

Abstract

Some problems of sample and human body radioactivity measurement optimization are discussed on the basis of new radiometric quality criteria and analysis of about 100 whole-body counters' data.

Определение оптимальных условий спектрометрических измерений малых активностей производится с помощью критерия радиометрического качества (КРК), максимальное значение которого соответствует параметрам, обеспечивающим наивысшую чувствительность методики. Ранее было показано<sup>1;2</sup>, что используемые на практике КРК в виде комбинаций скорости счета препарата  $n_0$  и фона  $n_\phi$  ( $n_0^2/n_\phi; n_0$ ) дают неточный оптимум режима, т.к. действительны только в экстремальных случаях  $n_0 \ll n_\phi$  и  $n_0 \gg n_\phi$ <sup>3;4</sup>. Истинный оптимум при любом соотношении  $n_0/n_\phi$  указывает КРК  $Q = 1/\varepsilon^2 T$ , где  $\varepsilon$  - относительная ошибка и  $T$  - продолжительность измерения препарата. Так, например, для исходного  $n_0/n_\phi = 5$  КРК  $Q$  указывает, что без ухудшения чувствительности методики допустимо увеличение фона в  $n_{\phi 2}/n_{\phi 1} = 20$  раз, если при этом эффективность возрастает в  $n_{0 2}/n_{0 1} = 3$  раза, (см.рис.1), тогда как КРК  $n_0^2/n_\phi$  допускает возрастание фона только в 9 раз. Вычисление  $Q$  производится с помощью набора кривых  $Q = \text{const}$ , в координатах  $n_0; n_0/n_\phi$  или по выра-



жению  $Q$  через эти параметры. Кривые зависимости  $Q$  от активности  $A$  препарата дают сопоставление приборов по чувствительности с учетом их эффективности и фона, а значение  $Q$  указывает степень трудоемкости радиометрии определенных уровней активности. Соотношение  $Q$ ,  $A$ ,  $E_\gamma$ , ширины канала регистрации определяет оптимальный режим и реально измеряемую активность. Энергетическая зависимость  $Q$  близка к обратной пропорциональности в диапазоне  $0,3+0,6$  Мэв для  $A$  от  $1$  до  $50$  пкюри, имеет плоский минимум в области  $0,7+0,8$  Мэв с нарастанием при  $E_\gamma = 1$  Мэв, существенный при  $A \ll 1$  пкюри; при  $A$  от  $0,05$  до  $1$  пкюри  $Q$  уменьшается вплоть до  $E_\gamma = 1,5$  Мэв. Для спектрометров с кристаллами  $NaJ$   $\varnothing 40 \times 40$  мм и  $\varnothing 70 \times 70$  мм  $Q$  изменяется в диапазоне  $E_\gamma (0,06+2,5$  Мэв), соответственно, в  $25$  и  $10$  раз. При этом отличие  $Q$  от упрощенного КРК  $-n^2/n_\varphi$  достигает  $15-30$  уже при активности препаратов  $0,1+1$  икюри. Для оценки трудоемкости радиометрии препаратов различной активности в диапазоне  $E=0,1+1,5$  Мэв были рассчитаны значения  $Q$  для различных вариантов геометрии измерения. На рис.2 приведена номограмма для кристалла  $NaJ$   $\varnothing 70 \times 70$  мм в чугунной защите толщиной  $10$  см и препаратов объемом от  $5$  до  $2000$  см<sup>3</sup>, позволяющая оценить степень трудоемкости радиометрии препаратов активностью от  $0,01$  до  $10$  икюри. Результаты анализа показывают, что обобщенный КРК  $Q$  уточняет до  $2-7$  раз оценки детектируемых уровней активности, которые можно получить из упрощенных КРК. С применением КРК  $Q$  построена номограмма (рис.3), связывающая значения  $A$ ,  $n_\varphi$ , эффективности ( $\mathcal{E} \%$ ), полной продолжительности измерения  $T$  и относительной ошибки ( $\mathcal{E} \%$ ) в диапазоне  $A=0,5$  пкюри+ $0,1$  мкюри;  $n_\varphi=0,01$  +  $1000$  имп/мин;  $\mathcal{E}=0,001+100\%$ ;  $T=1+1000$  мин.;  $\mathcal{E}=2+50\%$ . Номограмма весьма удобна при планировании экспериментов с радиометрическими измерениями.<sup>6</sup>

Оптимизация конструктивных параметров аппаратуры приобретает особое значение при разработке спектрометров гамма-излучения человека (СИЧ), для которых характерна высокая стоимость защитной камеры и детекторов большого объема. В данном случае из-за трудности учета всех факторов расчетные методы или моделирование не позволяют определить достаточно достоверно зависимость радиометрических параметров СИЧ от конструктивных параметров, характеризующих геометрию измерений, материал и толщину защиты и пр. Нами был предпринят анализ параметров более  $100$  СИЧ, опубликованных в сводке<sup>7</sup>. Рассматривались дифференциальные и куму-

лятивные распределения параметров, исследовалась связь между параметрами, определялись значения для каждого СИЧ. Несмотря на существенные различия конструкции СИЧ, анализ выявил отдельные закономерности.

Ниже приводятся результаты относящиеся к оценке толщины  $d$  защиты и объема кристаллов детектора. Ранее было показано <sup>8</sup>, что для сравнения качества защит следует пользоваться плотностью фона (отношением скорости счета фона  $N_p$  к интервалу энергий  $\Delta E$  и полной поверхности  $S$  кристалла), которая в широком диапазоне энергий гамма-излучения не зависит от размеров кристалла. Распределение СИЧ по величине плотности фона в области 0,66 и 1,46 Мэв показано на рис.4. На рис.5 представлена зависимость от толщины  $d$  средней плотности фона для 4 групп стальных защит в диапазонах толщин  $d=10+12,5$ ;  $14+16$ ;  $16-19$  и  $20$  см. Из рис.5 видно, что увеличение толщины защиты от 15 до 20 см сопровождается уменьшением фона порядка 30%, т.е. снижением минимальной обнаруживаемой активности лишь на 15-20%. Поэтому 15 см следует считать оптимальной толщиной стальной защиты. При такой толщине больший эффект дает дополнительные затраты, направленные не на усиление защиты, а на увеличение объема кристалла детектора. По средним значениям плотностей фона в области 0,66 и 1,46 Мэв защиты из свинца толщиной 10-12 см оказались эквивалентны 20 см стали, а толщиной 5-6 см - 15 см стали и на 30-35% эффективнее стальных защит толщиной 10-12 см. Поэтому облегченные защиты транспортных СИЧ целесообразно изготавливать из свинца.

На рис.6 показаны кумулятивные распределения СИЧ по величине относительной статистической ошибки  $\Delta K/K$  определения содержания калия (150 г) в организме человека за 20 минут (нижняя кривая) и по величине активности  $A_0$  инкорпорированного цезия-137, определяемой с точностью 20% за 5 минут (без учета вклада K-40) - верхняя кривая. Распределения показывают, что 90% СИЧ способны измерять за 5 минут менее 10 нкюри цезия-137 и определять за 20 минут среднее содержание калия с точностью лучше 8%. Зависимость от суммарного объема  $V$  кристаллов СИЧ средних значений  $A_0$  и  $\Delta K/K$ , полученных для отдельных групп СИЧ с типичными значениями  $V$ , представлена на рис.7 (здесь опущены несколько СИЧ с малым объемом кристаллов, для которых  $A_0$  и  $\Delta K/K > 20$ ). Видно, что радиометрические качества СИЧ резко ухудшатся при  $V < 1500$  см<sup>3</sup>. Однако, с увеличением суммарного объема

кристаллов от 2000 до 6800 см<sup>3</sup> чувствительность возрастает в среднем лишь в 1,7 раза. Поэтому с точки зрения радиометрического качества оптимальным является применение одного кристалла  $\varnothing 15 \times 10$  см ( $V \sim 1800$  см<sup>3</sup>) или  $\varnothing 20 \times 10$  ( $V \sim 3200$  см<sup>3</sup>). Увеличение суммарного объема кристаллов свыше 3500 см<sup>3</sup> целесообразно лишь в том, случае, если применяют несколько детекторов для улучшения изочувствительности геометрии измерений.

#### ЛИТЕРАТУРА

1. R. Loevinger, M. Berman. *Nucleonics* 1951, 8, 26.
2. В.М. Малыгин В сб. "Радиоактивные изотопы в атмосфере и их использование в метеорологии" Атомиздат 1965, 456-460.
3. M.A. Greenfield et al, *Int. Journ. Appl. Radiat.* 1960, 10, 205.
4. В.М. Малыгин, *Измерительная техника* 1966, 12, 84-85.
5. В.М. Малыгин, *Атомная энергия*, 1965, 1, 78.
6. В.М. Малыгин, *Приборы и техника эксперимента*, 1969, 3, 87-88.
7. *Directory of Whole Body Radioactivity Monitors.* IAEA, Vienna 1970.
8. Ю.С. Белле, О.В. Лебедев В сб. "Методы определения радиоактивности". Изд. АН УССР, 1972, 109-124.

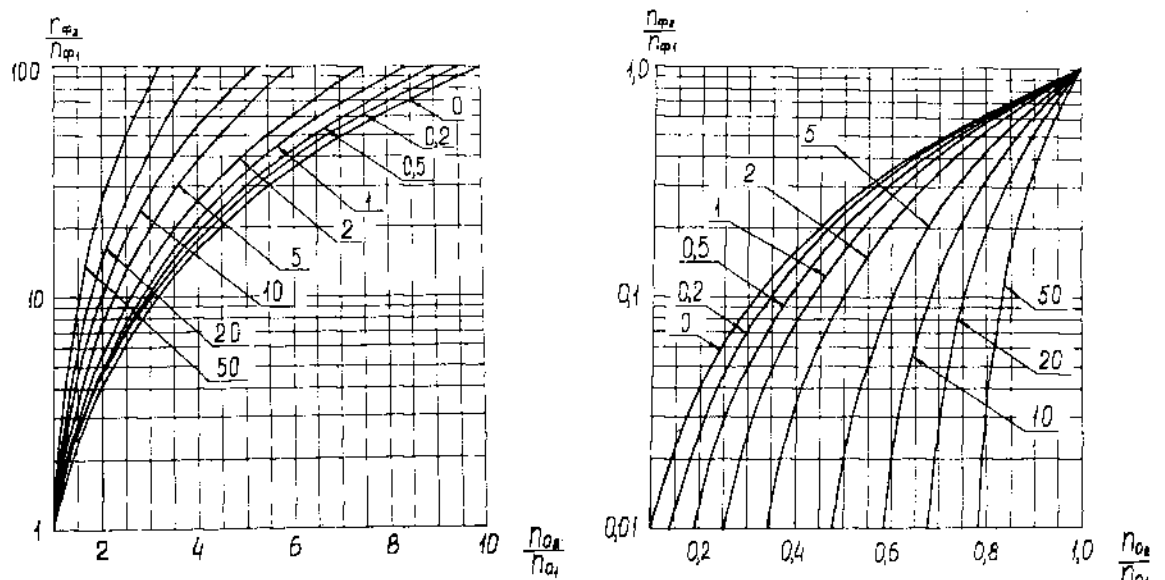


Рис. 1.

ПОДПИСИ К РИСУНКАМ.

- Рис.1. Допустимая кратность изменения фона  $n_{\phi 2}/n_{\phi 1}$  при изменении эффективности регистрации в  $n_{o2}/n_{o1}$  раз, не вызывающая ухудшения чувствительности методики. Параметр у кривых соответствует исходному значению  $n_o/n_{\phi}$ .
- Рис.2. Степень трудоемкости радиометрии проб объемом от 5 до 2000 см<sup>3</sup> с помощью детектора с кристаллом NaI Ø 70x70 мм в чугунной защите толщиной 10 см.  
 I зона:  $\epsilon \leq 10\%$  T=1 мин                      II зона:  $\epsilon \leq 10\%$  T=15 мин.  
 III зона:  $\epsilon \leq 20\%$  T=1 час                      IV зона:  $\epsilon \leq 30\%$  T=4 часа.
- Рис.3. Номограмма для определения статистических характеристик режима радиометрии.
- Рис.4. Распределение СИЧ по плотности фона в области 0,66 и 1,46 Мэв.
- Рис.5. Зависимость средней плотности фона в области 0,66 и 1,46 Мэв от толщины  $\alpha$  стальной защиты.
- Рис.6. Распределения СИЧ по величине относительной ошибки  $\Delta K/K$  определения 150 г калия в организме человека за 20 мин. (нижняя кривая) и по величине  $A_o$  активности цезия-137, измеряемой за 5 минут с точностью 20% (верхняя кривая).
- Рис.7. Зависимость средних значений  $\Delta K/K$  и  $A_o$  от суммарного объема V кристаллов СИЧ.

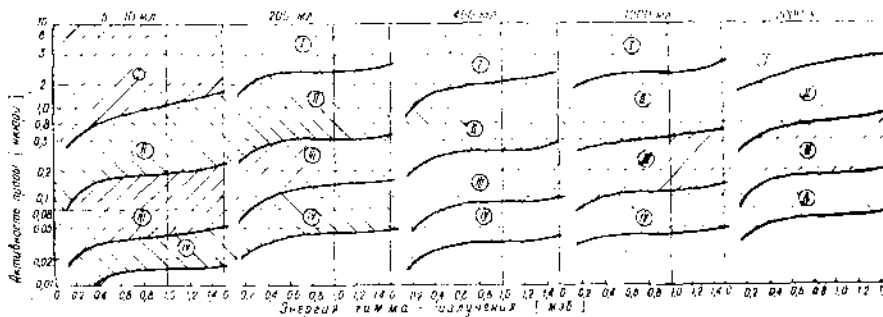


Рис. 2.

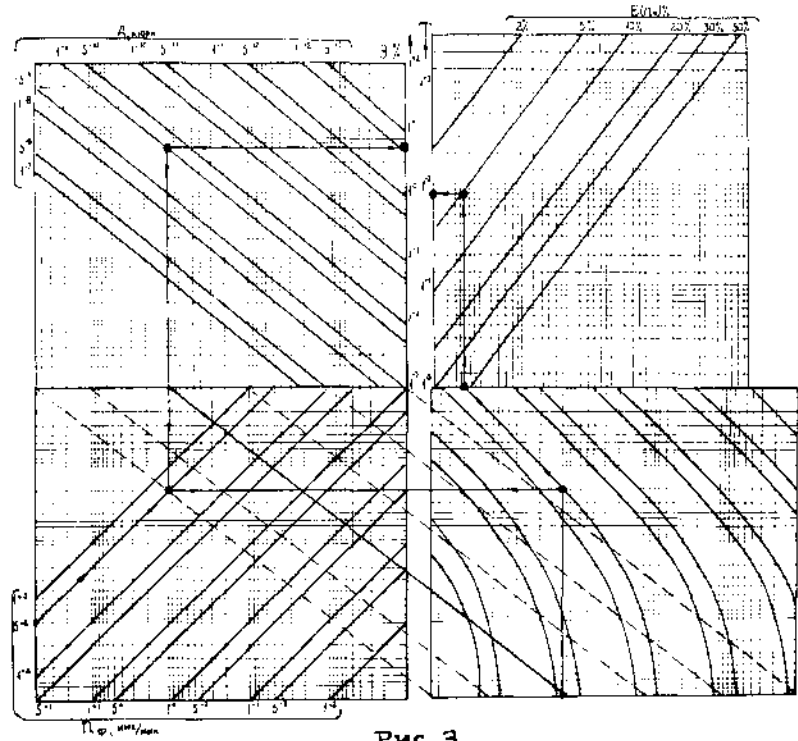


Рис. 3.

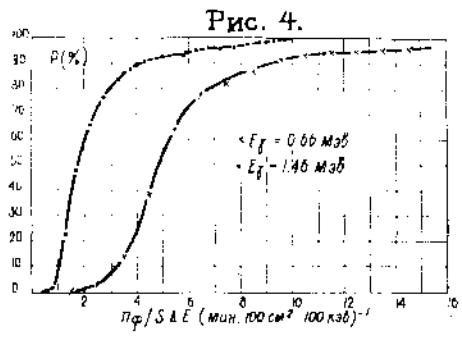


Рис. 4.

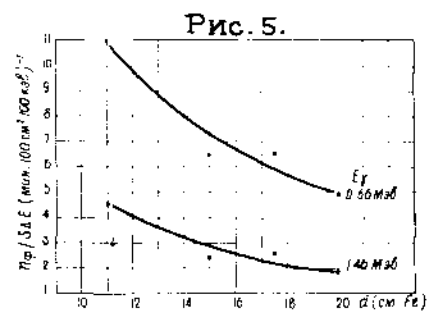


Рис. 5.

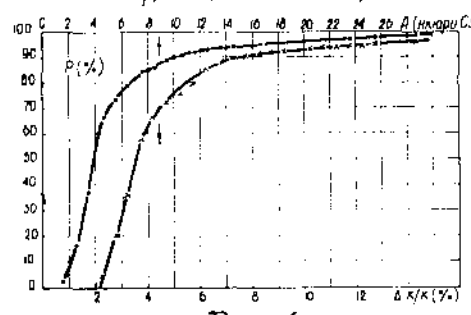


Рис. 6.

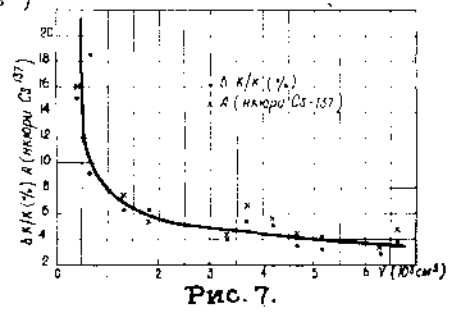


Рис. 7.

THE METHODS OF ABSOLUTE CALIBRATION OF EQUIPMENT FOR  
MEASUREMENTS OF Pb-210, Pu-239 AND Am-241 IN HUMAN BODY

E. Dolguirev, G.N. Kaidanovsky, N.V. Porozov, V.P. Shamov

The Institute of Radiation Hygiene

Leningrad

USSR

Abstract

Calibration of detectors designed for in vivo measurements of  $^{210}\text{Pb}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  in human body was made by means of an anthropomorphic phantom. Absorption of low energy photons (13 - 60 keV) in materials simulating various biological tissues was studied. The calibration coefficients for the fat tissue ( $Z=5.9$ ) were 3 times as high as those for the muscular one ( $Z=7.4$ ), when the tissue thickness was 2 cm. In this connection, the distribution and ratio of the said tissues in human body were studied, using a group of 30 men. In calibration phantom measurements, a tissue equivalent material simulating the biological tissue with 22% of fat tissue and 78% of muscular one was used. Detectors have been calibrated for measurements in the energy bands: 10-25 keV  $^{239}\text{Pu}$ , 30-55 keV,  $^{210}\text{Pb}$ , 10-25 keV and 35-70 keV  $^{241}\text{Am}$  and tissue absorber thicknesses in the range from 0.5 to 4 cm. The effect of such factors as variations in the shape and location of body organs, radiations of isotopes deposited in the skeleton and some others was taken into account.

Introduction

The interpretation of the results of in vivo measurements of the body  $^{210}\text{Pb}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  depends on the choice of calibration coefficients appropriate to a specific subject. Detectors may be calibrated, using (a) volunteers who take a safe dose of the assayed isotope <sup>1,2</sup>, or (b) anthropomorphic phantoms. The calibration made on volunteers may provide sufficiently realistic coefficient values which allow for both the complex structure of the bone and soft tissue shield and the actual distribution of inhaled aerosol throughout the lungs.

However, such factors as the availability of volunteers, the choice of isotopes to simulate a given radionuclide as well as the possibility of modelling of nothing more than a particular case of the radionuclide distribution in the lungs limit the scope of this direct method of calibration.

This paper deals with the further development and improvement of the phantom calibration technique which can provide numerical calibration coefficients for the deposited radionuclide distribu-

tion of the lungs-liver-skeleton pattern.

### Phantom Design and Tissue Equivalent Materials

The basic criteria used in the phantom development were its tissue equivalence in the 10-100 keV radiation band, modelling of different types of radionuclide distribution and simulation of different thicknesses of the tissue absorber in the region of the chest.

The complete phantom assembly consists of component phantoms, of the skull, chest and arms and legs. A natural human skull is used as the skull phantom and hollow polyethylene cylinders simulate the extremities.

The chest phantom assembly comprises a thin (0.2 cm) plexiglass shell, containing a human thoracic skeleton and man-made organs (the lungs and liver). The free space of the shell is filled with the tissue equivalent material. The shell front wall is movable and it is provided with an attachment for setting it at a desired distance from the phantom sternal ribs. The front wall and the phantom base are supplied with portholes for filling the tissue equivalent material and artificial organ replacement.

### Phantom Filling Materials

The muscular and fat tissue and the skin are the main constituents of the human tissue covering the thoracic cage. The calculated values of the mass absorption coefficients for these tissues (cm<sup>2</sup>/g) at 10-100 keV shown in Table 1 (Columns 2, 3 and 4) suggest that the fat component should be taken into account in the selection of the tissue equivalent material. (The skin, as far as its absorption properties are concerned, may be assumed to be similar to the muscular tissue).

Table 1

Tissue Energy band	Pectoral muscle	Fat	Skin	Mean tissue shield	Tissue equivalent material
10	5.249	2.406	4.854	4.579	4.520
15	1.633	0.815	1.499	1.356	1.357
20	0.789	0.409	0.722	0.684	0.673
40	0.259	0.217	0.237	0.247	0.243
50	0.221	0.195	0.199	0.212	0.209
60	0.201	0.184	0.181	0.195	0.195
100	0.170	0.161	0.151	0.166	0.163

The experimental testing of the fat and muscular tissues for their absorption properties were carried on the phantom, which was alternately filled with materials simulating each of these tissues. Attenuation curves for the X-ray radiation of <sup>239</sup>Pu emitters uniformly distributed throughout the lung model were obtained. The experimental results given in Table 2 are consistent with the data in Table 1 and support our suggestion.

Table 2

Tissue thickness, cm	1	2	3	4
The ratio of photons which passed through the "fat" to those which passed through the "muscles"	1.4	3	6	10

The ratio of the thicknesses of the fat and muscular tissues covering the thoracic cage in the region of the lungs was measured in 18 male corpses and was found to vary from 34/66 to 10/90, the mean value being 22/78.

Table 1 (columns 5 and 6) gives values of the mass absorption coefficients for the experimental mean tissue consisting of 22% fat and 78% muscles and the tissue equivalent sugar and magnesium oxide based material. The electronic densities of this material and the simulated tissue are  $3.24 \cdot 10^{23}$  and  $3.32 \cdot 10^{23}$  electron/gram, respectively.

The models of organs, i.e. the lungs and liver, were made of the tissue equivalent material to conform to the average size, shape and density of their human prototypes. The model shell was capron. The lung model shell was filled with a cotton fabric soaked in a NaCl solution to reproduce the desired values of  $Z=7.4$  and  $\rho=0.27$  g/cm<sup>3</sup>. The same material was used for filling the liver and chest shells. The experimental coefficients of self-absorption of <sup>239</sup>Pu radiation in the lungs and liver were found to be 3.4 and 5.1, respectively.

#### Measurement of Soft Tissue Thickness

Owing to a considerable attenuation of low-energy X-ray radiation in the tissues, the accuracy of calibration coefficients selected for the measurements in the monitored subject is materially dependent on the precision of measurements of the thicknesses of soft tissues covering the thoracic cage.

To develop techniques for measurement of soft tissue thicknesses, radiograms of tissues in a spacial saggital plane were made. Previously, the ratio of the average thickness across this section  $d_s$  to the mean thickness of these soft tissues across the whole area covered by the detector  $d_g$  was obtained in corpse measurements.

The equation  $d_s/d_g = 1.05 \pm 0.12$  holds for this ratio in a wide range of W/H variations of 0.37 - 0.52, where W - weight, kg; H - height, cm.

The subject's posture proved to influence the thickness of the tissue shield in the chest region. The smallest thickness of the soft tissues over the lungs was found in in vivo measurements to be, when arms are raised and placed behind the neck of the subject's head. It was shown experimentally that, for this posture, the absorber thickness decreases by 0.48 cm, as compared with a pose with arms at sides, which is consistent with the value of 0.5 cm, given in This posture was accepted as standard in all subsequent measurements.

The measurement data for soft tissue thicknesses in 26 subjects were related to different parameters of the body and approximated by means of functions, such as  $d = \varphi(W, H, C, C_1, C_2)$ , where  $C, C_1, C_2$  - circumferences of the chest, waist and hips, res-



pectively, cm; W - weight, kg, H - height, cm. These parameters were measured simultaneously with radiograms being taken. The dependence of d on the selected parameters is best approximated by the equation:

$$d = 118 W/H \cdot k_1 \cdot k_2 - 39.2, \text{ where } k_1 = C/C_2, k_2 = C/C_2$$

The graph showing this relationship is given in Fig.1.

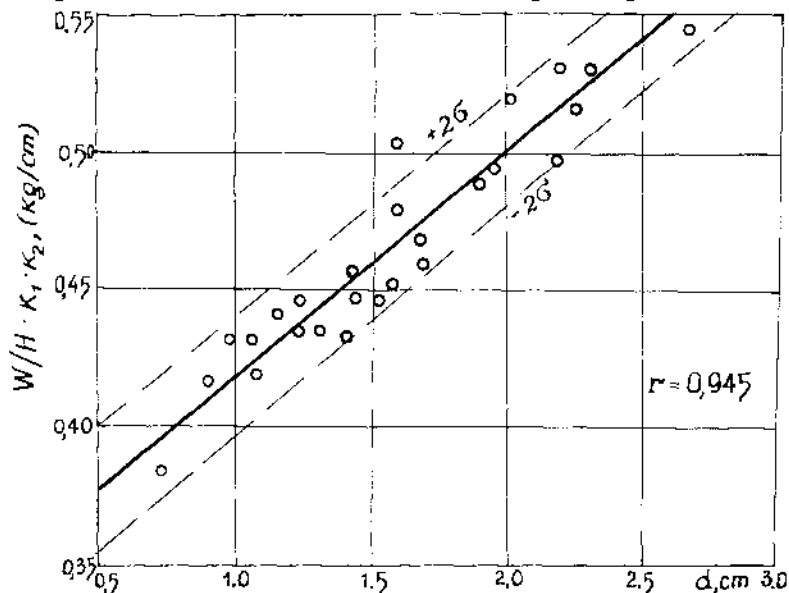


Fig.1. The correlation of soft tissue thickness d and parameters W/H,  $k_1$  and  $k_2$ .

Thus, simple measurements of the body parameters make it possible both to assess the thickness of the soft tissue layer covering the thoracic cage and to choose suitable calibration coefficients.

#### Calibration Procedure

The lungs, liver and skeleton are the chief sites of  $^{210}\text{Pb}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  deposition in human body. Suitable sets of calibration coefficients are required for the assessment of burdens of these radionuclides in the said organs on the basis of in vivo measurements.

100 point emitters of known activity were placed uniformly alternately throughout the models of the lungs and liver. The distribution of  $^{210}\text{Pb}$  or  $^{241}\text{Am}$  emitters in skeletal bones is given in Table 3. Measurements in the four points, i.e. the left lung, the right lung, the liver and the skull, were carried out for each insertion of the emitters into one of these models. The pulse count rate was registered at 10-25 keV for calibration with respect to  $^{239}\text{Pu}$ , 10-25 keV and 25-70 keV ( $^{241}\text{Am}$ ) and 30-35 keV ( $^{210}\text{Pb}$ ).

The data of the standard calibration of NaI scintillation counter ( $S=177 \text{ cm}^2$  and  $h=0.1 \text{ cm}$ ) for a 1.7 cm - thick soft tissue are given in Table 4.

Table 3.

## Distribution of activity in the skeleton

Bones of skeleton	Weight %	Quantity of sources		Activity nCi
		11 nCi	2nCi	
Skull	14.1	50		550
Chest Cage	15.3		210	588
1. Sternum	1.2		16	44.8
2. Ribs	9.1		126	352.8
3. Clavicles	1.2		16	44.8
4. Scapulae	3.8		52	145.6
Vertebral Column	9.0	38		418
1. Cervical	1.4	5		55
2. Thoracic	4.2	14		154
3. Lumbar	3.4	12		132
4. Sacrum	2.0	7		77
Pelvis	7.8	27		297
Arms	16.2	56		616
1. Radii	5.9	20		220
2. Ulnae	4.4	16		176
3. Hands	5.9	20		220
Feet	35.6	126		1386
Femora	15.7	56		616
Tibiae	14.2	50		550
Feet	5.7	20		220
Total	100	297	210	3855

The sets of calibration coefficients for soft tissue thicknesses from 0.5 to 3.5 cm for scintillation and proportional counters are given in our paper "Dosimetric Monitoring of Content of the Radionuclides with a Low Radiation Energy in the Human Organism"

The total error for calibration coefficients (Table 4) due to the variations in the organ shape, the different location of the organs in the phantom the detector displacement with respect to the phantom front wall, the counting statistics and the precision of emitters' calibration is 16% for  $^{239}\text{Pu}$ ; 8% - for  $^{210}\text{Pb}$  and 6% - for  $^{241}\text{Am}$ .

The error due to the assumption of the uniform distribution of the assayed radionuclide throughout the organ was determined separately. The values of calibration coefficients for different patterns of radionuclide distribution in the lung model are shown in Tables 5 and 6.

Table 4

Calibration Factors for Homogeneous Distribution cpm/nCi/d=1.7cm

Source position	Counter position	Radionuclides and energy band			
		$^{239}\text{Pu}$	$^{210}\text{Pb}$	$^{241}\text{Am}$	
		10-25	30-55	10-25	35-70
Left lung	left lung	0.120	3.1	8.5	40.0
	right lung	0.004	0.7	2.5	8.4
	liver	0.04	0.2	0.7	2.0
Right lung	left lung	0.004	0.7	2.5	8.4
	right lung	0.120	3.1	8.5	40.0
	liver	0.006	0.4	1.6	5.0
Liver	left lung	0.003	0.8	2.1	10.0
	right lung	0.016	1.4	3.3	18.0
	liver	0.040	1.8	5.9	22.6
Skeleton	left lung		0.22		3.6
	right lung		0.22		3.6
	liver		0.15		1.7
	skull		0.40		7.5

Table 5

Calibration Factors (cpm/nCi, d=17 cm) for  $^{241}\text{Am}$  35-70 keV

Position		Emitter distribution in the lung			
Emitters	Detectors	Uniform	Upper part	Lower part	60% in lymphatic nodes; 40%-uniform
Left lung	Left lung	40.0	35.0	43.0	42.0
	Right lung	5.4	7.9	8.7	8.3
	Liver	2.0	1.2	2.3	1.9

Calibration Factors (cpm/nCi, d=1.7 cm) for  $^{239}\text{Pu}$  10-25 keV

Position		Emitter distribution in the lung			
Emitters	Detectors	Uniform	Upper part	Lower part	60% in lymphatic nodes; 40%-uniform
Left lung	Left lung	0.120	0.085	0.130	0.105
	Right lung	0.004	0.003	0.003	0.003
	liver	0.004	0.001	0.005	0.003

### References

- 1.D.NEWTON et al., Assessment of Radioactive Contamination in Man, IAEA, Soholm, 1972.
- 2.B.T.TAYLOR Health Physics, 17, 1, 1969.
- 3.D.A.WAITE et al., American industrial Hygiene ass., v.31, 1970.
- 4.R.G.SPEIGHT et al., AEEW-R 359.

ЧУВСТВИТЕЛЬНЫЙ МЕТОД РАДИОМЕТРИИ  
ИННОРПОРИРОВАННЫХ НУКЛИДОВ ТОРИЕВОГО РЯДА

А.А.Петушков, М.Р.Зельцер, П.П.Ляской

Институт гигиены труда и профзаболеваний АМН СССР,  
Москва, СССР

The complex of the apparatus and methods for the assessment of Th-228 body burden in man was developed. The correlation is found between content of thoron ( $Q_{Th}$ )<sub>A</sub> in the breath, which was measured by a special scintillation apparatus (0.05 pCi/l sensitivity), and Ra-224 in the excretion ( $Q_{R2}$ )<sub>x</sub>, which was measured by the method of the high emanating samples preparation (0.05 pCi sensitivity), with the activity of Th-228-body burden in man.

Расширение использования в промышленности тория и различных неметаллических руд, в которых торий присутствует как примесь ведет к увеличению контингента людей, работающих в условиях контакта с радионуклидами ториевого ряда. В связи с этим понятен интерес к проблеме контроля поступления в организм и накопления в нем тория и продуктов его распада /1 ; 2 ; 3 ; 4 /.

В соответствии с указаниями МНРЗ (ICRP-10) и ННРЗ СССР (НРБ-69) такой контроль должен строиться в основном на определении индивидуального содержания в организме изотопов ториевого ряда, среди которых большое значение имеет радиоторий, ответственный вместе с дочерними продуктами за 90% энергии альфа-распада ториевого ряда.

Наиболее точную оценку содержания радиотория в организме (в пределах своей чувствительности) дает метод гамма-спектрометрии человеческого тела с помощью СИЧ, однако, как показывает практика, его чувствительность (1-5 нюри) в большинстве случаев недостаточная для проведения массовых обследований. В принципе большую чувствительность могут обеспечить методы оценки содержания радиотория в организме по активности торона в выдыхаемом воздухе и радия-224 в выделениях. Для их реализации нужно знать величины коэффициента эманирования торона и перехода эндогенного радия-224 в выделения, которые для наиболее важного в условиях профессионального контакта ингаляционного поступления тория в организм изучены недостаточно.

Чтобы получить необходимые соотношения, мы предприняли попытку с помощью аппарата камерных моделей проанализировать имеющуюся в работах / 6 ; 7 ; 8 ; 9 ; 10 и других / экспериментальную информацию о метаболизме тория и продуктов его распада в организме при их внутривенном и аэрогенном поступлении.

Анализ показал, что в случае аэрогенного поступления радиотория в растворимой, ионной форме ежедневно с экскретатами выводится доля равновесного радия-224, равная  $4 \pm 5\%$  от содержания радиотория в организме -  $\mathcal{A}_{T_2}$ , причем перераспределение со временем депонированного радиотория существенно не сказывается на этой величине; чего нельзя сказать об активности торона в выдыхаемом воздухе, которая за время  $2T$  (где  $T \sim 80$  дн - период полураспада торона из легких) изменяется от 13 до 6% от величины  $\mathcal{A}_{T_2}$ . Отметим, что скорость выведения радиотория не превышает  $0,2\% \mathcal{A}_{T_2}$  в день.

При поступлении радиотория в нерастворимой форме ежедневная экскреция равновесного радия-224 зависит от дисперсности пыли, с которой радиоторий поступает в легкие: для пыли двуокиси тория с  $СМД \sim 1 \mu$  ежедневно выводится  $\sim 1\% \mathcal{A}_{T_2}$ , а для пыли двуокиси с  $СМД = 0,2 \mu$  -  $2\% \mathcal{A}_{T_2}$ ; в то же время эмансирование торона зависит от дисперсности слабее и для указанных пылей коэффициент эмансирования  $\sim 6\% \mathcal{A}_{T_2}$  и также как и экскреция радия-224 существенно от времени не зависит.

Для определения содержания в выделениях радия-224 нами используется методика, основанная на эмансметрии сухих препаратов, коэффициент эмансирования торона из которых специальным приготовлением доведен до величины большей 0,8.

Чувствительность метода 0,05 нюри радия-224 в пробе /12/. Этот метод определения радия-224 проще альфа-спектрометрии / 13 / или метода "смешивания" / 14 /, с его помощью произведена оценка величины ежедневной экскреции радия-224 с калом -  $F$  у людей, не имевших контактов с торием,  $F \sim 1$  пюри/с.

Здесь очень чувствительный метод измерения содержания торона в выдыхаемом воздухе был предложен I. Hursh и A. Lovas / II /. Однако его реализация наталкивается на трудности поддержания высокой и стабильной сорбционной способности угольной ловушки, с помощью которой из выдыхаемого воздуха извлекается торон.

Для определения концентраций торона в выдыхаемом воздухе у людей в данной работе сконструирован удобный с эксплуатацией, автономный сцинтилляционный эмансметр, в котором регистрируется альфа-распад торона и тория-А в проходящей через камеру прибора струе воздуха.

Объем и конструкция камеры выбраны так, чтобы обеспечить оптимальную величину коэффициента использования активности эмансции  $\epsilon = 200 \frac{1/\text{час}}{\text{пюри/л}}$  при средней скорости продувки 8 л/мин.

Фон прибора, когда последний соединен с человеком, легкие которого в течение 20' очищались безрадонным воздухом, как показали специальные измерения не превышает 10-12 1/час.

Это позволяет измерять концентрацию торона в выдыхаемом воздухе  $\sim 0,05$  пюри/л.

С помощью этого прибора произведена оценка естественного содержания торона в выдыхаемом воздухе у работающих ( $0,14 \pm 0,02 \frac{\text{пюри}}{\text{л}}$  для группы 10 чел.). У курящих концентрация торона оказалась вдвое большей ( $0,28 \pm 0,03 \frac{\text{пюри}}{\text{л}}$  для группы 10 чел.), что подтверждается данными S. Jovet / 15 / о возможности накопления тория в легких при курении.

Приведенные выше соотношения, связывающие активность тория-228 в организме с активностью радия-224 в выделениях и торона в выдыхаемом воздухе, а также данные о чувствительности использованных радиометрических методик позволяют сделать заключение, что в случае ингаляционного поступления радиотория в растворимой форме целесообразно определять его содержание в организме по активности радия-224 и выделениях; предел обнаружения при этом  $\sim 20$  пюри

в организме. Если радиоторий поступает в нерастворимой форме, то лучше, по-видимому, воспользоваться эманометрическим методом (предел обнаружения  $\sim 10$  пикюри радиотория в легких).

В обоих случаях эти методики позволяют определять радиоторий на уровне естественного его содержания в организме.

Разработанные методы применяются для обследования на содержание радиотория у рабочих рудников и горнообогатительных фабрик промышленности редких металлов. У некоторых из них обнаружено повышение содержания торона в выдыхаемом воздухе в пределах 0,3 - 0,5 пикюри/л; поскольку торий в этом случае поступал в форме нерастворимой пыли, можно на основании вышеизложенного заключить, что в легких у них имеется депо радиотория с активностью 50-120 пикююри.

#### ЛИТЕРАТУРА

1. Г.Е.Наплан, Атомная энергия 5, 2, 1958.
2. Н.Ю.Тарасенко, Гигиена труда при работе с торием. М., 1963.
3. R.Du Toit, в кн. "Radiological Health and Safety in mining and milling". Vienna I. 203. 1964.
4. R.Albert, "Thorium: Its industrial hygiene aspects". N-Ч.1966.
5. Н.А.Павловская "Метаболизм тория-232 и продуктов его распада в организме животных и человека. Докт. дис. М., 1972.
6. C.Mays et al.,Rad. Res. 8. 6. 1958.
7. B.Stover et al.,Rad. Res. 26.1.1965.
8. В.Воскер, R. Thomas, Health Phys. 9. 2. 1963.
9. R.Thomas et al.,Health Phys. 9.2.1963.
10. I.Ballou, I. Hursh,Health Phys. 22, 2, 1972.
11. I.Hursh, A.Lovaas,Health Phys. 9.6.1963.
12. А.А.Патушнов с соавт. "Гигиена и санитария". 4, 1972.
13. I.Ogawa et al.,Nucl. Instr. and Met. 13, 169, 1961.
14. В. Rajewsky, W. Stahlhofen. Nature, 198, 4884, 1963.
15. G.Jovet,Experientia: 27. 1, 1971.

ASSESSMENT OF PLUTONIUM IN HUMAN LUNGS  
WITH THIN NaI(Tl) DETECTOR SYSTEMS

R.C. Sharma, S. Somasundaram, (Mrs.) T. Surendran,  
D.K. Kapur and S.P. Garg  
Health Physics Division (BBMS)  
Bhabha Atomic Research Centre  
Bombay 400 001, India

Abstract

The development of detector systems for assessment of inhaled Plutonium-Americium dust deposited in human lungs by direct detection of externally emitted low-energy photons, is a continuing programme at Trombay. This paper describes the work done with thin NaI(Tl) scintillation detector systems. The background data inside Trombay steel room in different low-energy bands for crystals of thicknesses 1, 2 and 5 mm are presented. To study the capabilities of three detector systems, each consisting of a set of crystals of the same thickness, a realistic chest phantom of an Indian adult was designed and employed. The chest phantom was constructed from a rib cage of an Indian adult enclosed in a hard polythene cover provided to simulate the chest profile. Measurements on absorption and scattering of low-energy photon (17, 22 and 60 keV) by four constructional materials were made to verify their degree of equivalence to human tissue and granular sugar was used as tissue equivalent material in phantom construction. The counting efficiencies and limits of detection of three detection systems for point sources of plutonium distributed in the central plane of each simulated lung of the designed phantom are reported.

A few normal subjects were counted with one detector system and the increase in background in low-energy region was investigated. The natural radioactivity of the subjects was monitored with a (20.32 cm dia. x 10.16 cm thick) NaI(Tl) crystal in a 50 cm Arc Chair. Finally, the effect of body build of a subject on the counting efficiency of plutonium is commented and our future programme is briefly indicated.

Introduction

At the Bhabha Atomic Research Centre, Trombay, we have an on-going programme of design and development of systems for in vivo assessment of plutonium and other transactinide elements deposited in the lungs. This paper (1) presents the results of our studies on the suitability of Trombay-produced thin NaI(Tl) crystals for this application, (2) describes the design and construction of a chest phantom and its use for calibration of the Pu lung monitor, and (3) compares our results with those of other laboratories using NaI(Tl) detectors, as reported in the literature.

Evaluation of Trombay-Produced NaI(Tl) Crystals

The Trombay-produced NaI(Tl) crystals, used in the present study, are of 7.6 cm diameter and 2 and 1 mm thick. They are canned in Al and



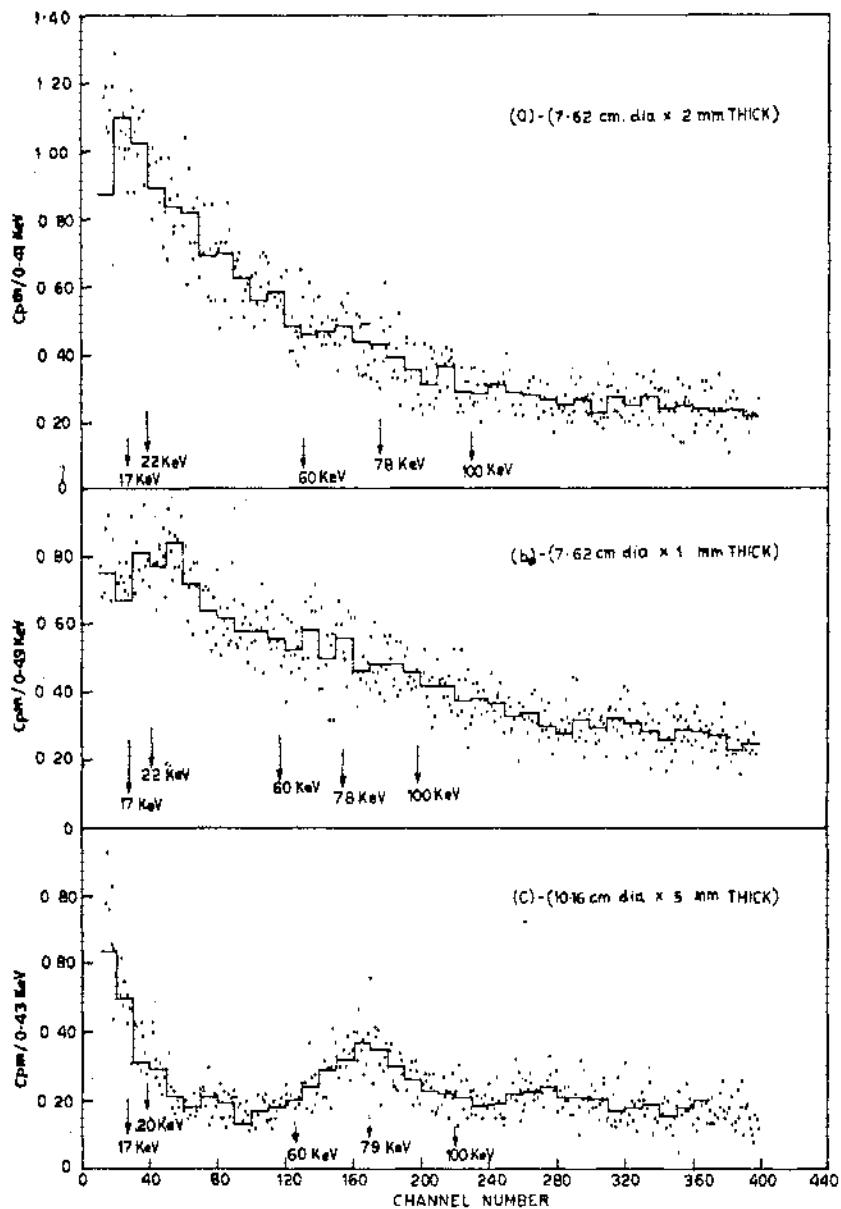


FIG. 1 - A COMPARISON OF THE BACKGROUND SPECTRA OF LOW ENERGY RADIATION INSIDE TROMBAY STEEL ROOM FOR THREE DIFFERENT THIN CRYSTAL DETECTORS. HISTOGRAMS REPRESENT TEN POINT AVERAGES.

are provided with 1 mil thick Al foil radiation entrance window and 3 mm thick glass optical window. They were coupled to selected low-noise Dumont 6363 phototubes and the output from the phototube was fed through an FET pre-amplifier to a 400-channel pulse height analyser fabricated in our laboratory.

A typical pulse height spectrum of a Trombay-produced Pu source, obtained with these crystals, showed that the L X-rays of U are not resolved and exhibited a peak at an average energy of 17 keV. A small peak at 60 keV pointed to the presence of  $^{241}\text{Am}$  in the source, the  $^{241}\text{Am}$  concentration being about 0.3% of the total Pu alpha activity. The energy resolution at 17 keV was estimated to be about 50%, compared to 46% obtained with a Harshaw integral assembly Model No. 16 MBS 5M/5A Q X (10.16 cm dia x 5 mm thick NaI(Tl) crystal, having 1 mm Be radiation entrance window and 2.54 cm thick quartz optical window) Fig.1 shows the background spectra of low-energy radiations inside the Trombay steel room<sup>1</sup>, observed with the three detectors. The histogram depicted represents ten point averages. The Harshaw assembly showed the lowest background rates in the energy bands of interest. These spectral measurements have indicated the aspects in which Trombay-produced crystals require improvement to be suitable for Pu detection<sup>2</sup>.

#### Construction of Chest Phantom

Speight et al<sup>3</sup> had suggested some tissue-equivalent materials for construction of chest phantoms. Due to non-availability of Lincolnshire bolue and Mix D and the handling problems of water, it was decided to test three probable constructional materials, viz. masonite, perspex and sugar, for their degree of tissue-equivalence. The effective atomic number (Z) and electron density (n) for tissue are quoted as 7.33 and  $3.32 \times 10^{23}$  respectively and for water as 7.42 and  $3.36 \times 10^{23}$  respectively. Perspex and masonite have much higher values of Z and n than tissue or water. The half-value thicknesses for water, sugar, perspex and masonite were found as 6.0, 6.0, 7.0 and 5.25 mm respectively at 17 keV, and 14.0, 14.0, 16.75 and 12 mm respectively at 22 keV<sup>2</sup>. The effects of forward scattering by almost equal thicknesses of the four materials as measured by energy shift, degradation of spectral resolution and variation of the ratio of count-rate in the 24-43 keV energy band to that in the 60 keV peak (43.5 - 76 keV) were all found to indicate a similarity of behaviour between water and sugar<sup>2</sup>. Sugar was, therefore, chosen in the construction of the phantom. No attempt was made to simulate the presence of fat in the chest region.

The chest phantom was constructed from a thoracic cage with clavicles, scapulae and shoulder blades taken out of the cadaver of an Indian adult. The whole rib cage fixed at the bottom to a perspex sheet was first enclosed in a frame of thin perspex strips. A hard polyethylene sheet was used to cover the frame to get the chest profile. All measurements were matched to a subject. The vertebrae contained a copper rod to keep them firmly attached. This, however, would not affect the calibration for Pu, since the bone is essentially a dark body for low-energy photons. Two symmetric polyethylene bags were filled with saw dust (density 0.3 g/cm<sup>3</sup>), each weighing about 450 g. These were shaped to a human lung and inserted inside the rib cage. Prior to this insertion, point sources of Pu deposited on perspex (1 mm thick both sides) were stuck to a filter paper which was spread longitudinally in the central plane of each simulated lung. The

presence of other parts of the respiratory tract and the heart were not simulated. Granular sugar was filled inside the polyethylene enclosure surrounding the rib cage, to provide simulation of tissue. The chest phantom thus constructed had a circumference of 83 cm. The total activity of Pu incorporated in the simulated lungs was 2.5  $\mu$ Ci.

#### Multi-Crystal Arrays

The phantom described above was employed to obtain the counting efficiencies and the minimum detectable activities for Pu in lungs for a multi-crystal array geometry simulated with a single detector. The multi-crystal array was selected with a view to achieve a large coverage of the frontal area of the chest phantom. The six crystal array chosen is shown in Fig. 2 together with the phantom outline. Each crystal array consisted of detectors of the same thickness. Thus, three multi-crystal arrays were studied to assess the capabilities of these systems and to evolve an array system of Trombay-produced crystals to count suspected cases of internal contamination by Pu and Am.

Table 1 gives the results with our systems and compares them with data of various Pu-lung monitors employing thin NaI(Tl) scintillation detectors as reported in the literature. The systems compared are those of Swinth & Griffins<sup>4</sup>, Ishihara et al<sup>5</sup>. The first column lists the various monitor parameters. In the net spectrum of radiation from point sources of plutonium distributed in the central plane of each lung longitudinally of the chest phantom we observed that attenuation of 13.6 and 16.9 keV results in shifting of the 17 keV peak to about 20 keV. The presence of <sup>241</sup>Am was indicated by peaks at 29 keV (26 keV + escape) and then at about 57.5 keV. A major conclusion drawn from Table 1 is that Pu lung monitors employing thin NaI(Tl) crystals in different configurations are not capable of achieving the MDA for plutonium desirable for routine monitoring. We achieved the best results when <sup>241</sup>Am is used as a tracer for Pu. The use of <sup>241</sup>Am is certainly not valid for soluble Pu. Nevertheless an immediate estimate of lung burden of Pu for a subject involved in an accident may be derived on this basis if isotopic composition of the contaminant is known. The MDA quoted for our systems are for very low <sup>241</sup>Am (0.5% of total alpha activity) content in Pu.

The MDA quoted for the multi-crystal arrays are all based on the statistical criterion only, i.e. three times the standard deviation in background rate for a given time of counting. Each subject would increase the background in the low energy region of the array, depending upon the level of internal contamination by other radionuclides and the thickness of the detectors employed. For the 5 mm crystal array, we observed that a subject with 125 gm K and 1 nCi <sup>137</sup>Cs increased the background by 20 cpm (12-25 keV) region and by 120 cpm in (43.5 - 76 keV) band. <sup>137</sup>Cs and K contents of the subjects are determined by whole body counting in 50 cm arc chair using a 20.32 cm x 10.16 cm NaI(Tl) detector. Collection of these data is continuing with a view to derive some useful correlations.

The counting efficiencies and MDA for Pu for the three six crystal arrays reported in Table 1 are thought to be valid for humans having body build and distribution of Pu in lungs similar to that of phantom. Several methods to correct the calibration factor for differing body builds have been proposed<sup>6,7</sup>. We have found it advantageous to use the concept of

Table 1  
COMPARATIVE DATA OF VARIOUS Pu-LUNG MONITORS EMPLOYING THIN NaI(Tl) SCINTILLATION DETECTORS

Sl.No.	Monitor Parameters	Swinth and Griffin	Ishihara et al	Our Multi-Crystal Array
1.	Detector size employed and geometry used	Array of 12 NaI(Tl) (2" dia x 1" mm each) four boxes of 3 detectors each. Two boxes positioned on the chest of the reclining subject. Other box edge 2" below the top of the chest. The other one on each side of the armpits	Single (8" x 4" mm) NaI(Tl) on the centre axis of the phantom and at a distance of 1.4 cm from the top plate to the crystal face	(10.16 cm x 5 cm); (7.6 cm x 2 cm); (7.6 cm x 1 cm) Three multi-crystal arrays studied each consisting of a set of six crystals of the same thickness; shielded with a single detector. Detector arrangement relative to phantom is shown in Fig.2. Crystal face in every position remains horizontal with the minimum distance from phantom surface being 1 cm.
2.	Phototubes coupled	Low noise 2" dia VPM/II crystal separately	Single 7" dia VPM/II/170 20th Century Electronics without light guide	(10.16 cm x 5 cm) NaI(Tl) : RCA-8055 through 2.54 cm of quartz pipe (7.6 cm x 2 cm) NaI(Tl) : Dumont-6363 through 0.50 cm of glass (7.6 cm x 1 cm) NaI(Tl) : Dumont-6363 through 0.50 cm of glass
3.	Phototube Noise predominance	Below 10 KeV	Below 15 KeV with no light guide	Below 12 KeV for all
4.	Total Sensitive Detector Area	15770 cm <sup>2</sup>	101.27 cm <sup>2</sup>	154.8 cm <sup>2</sup> for 5 mm crystal 57.1 cm <sup>2</sup> for the other two.
5.	Energy Bands used to cover	(a) (15.6 to 25.6 KeV) (b) (15.6 to 66.5 KeV) both X-ray and gamma covered.	(a) (16 to 31 KeV) (b) (35 to 75 KeV)	(a) (12 to 25 KeV) (b) (43.5 to 75 KeV)
6.	Energy Resolution at 17 KeV level	-	67.5% with no light guide	*46%, 50%, 48%
7.	Background counts per min	(a) Pu Ch.- 56.2±2.9 (b) Pu+Am Ch.- 204.8±2.5 (In 4" thick Pb shield lined with Cd and Cu)	(a) Pu Ch.- 51.2 (b) Am " - 135.0 (With no light guide) (In shielded chamber 20 Iron + 5 mm of Pb)	(a) Pu Ch.- 64.0, 204.0, 132.0 } For six crystal (b) Am Ch.- 105.0, 246.0, 254.0 } array In shielded chamber 20 cm Iron + 5 mm of Pb.
	With distilled water phantom	(a) 74.5 cpm (b) 388.0 cpm	-	-
8.	Calibration phantom and source position	Alderson Resab phantom filled with water. Plutonium (720 ppm Am) mixed with lung equivalent material kept in lung cavities	Phantom of Lucite sheets cluster of pt. sources placed in the model of human lungs	Chest phantom from human rib cage. Tissue equivalent material used is granular sugar. Point sources distributed longitudinally in the central plane of each simulated lung.
9.	Counting efficiency for Pu-239 in lungs counts per sec/nCi	(a) $0.641 \times 10^{-3}$ (Pu ch) (b) $4.38 \times 10^{-2}$ (X-ray and Am gammas)	(a) $1.34 \times 10^{-2}$ (b) -	* (a) Pu ch. $3.85 \times 10^{-3}$ ; $3.46 \times 10^{-3}$ ; $1.65 \times 10^{-3}$ For six crystal array each.
10.	Minimum Detectable amount of Pu	(a) 49 nCi } 20 min counting time (b) 67 nCi }	(a) 6.2 nCi of Pu-239 } 100 min (b) - } counting time.	* (a) Pu Ch. 12 nCi; 28 nCi; 34 nCi and 2.44, 10, 8 nCi if Am is used as a tracer. (For counting Pu) For six crystal array each and counting time of 100 min.
11.	Contribution from degraded Ca-137 and K-40 gammas	Mumuk subjects add in (a) Pu Ch.- 65.2 to 130.2 cpm (b) Am Ch.- 458 to 798 cpm	A subject with 150 gK add 4 nCi of Ca-137 adds in (a) Pu Ch.- 12.6 cpm (b) Am Ch.- 75.6 cpm	For 5 mm detector array only; a subject with 125 gK and 1 nCi of Ca-137 contributes: (a) Pu Ch.- 20.0 cpm (b) Am Ch.- 120.0 cpm.

\*The three successive values are for 5 mm, 2 mm and 1 mm thick detectors respectively.

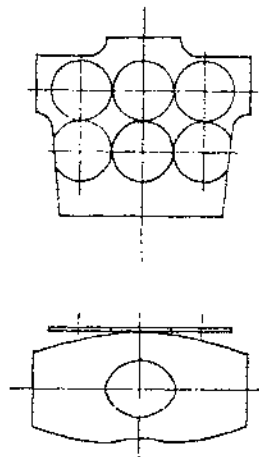


FIG.2-THE MULTI-CRYSTAL ARRAY AND THE PHANTOM  
OUTLINE

effective tissue thickness (ETT) which is defined as the thickness of a tissue equivalent material that produces for a point source of Pu, the same overall attenuation of X-rays as would occur in a subject having Pu deposited in lungs. ETT takes account of self-absorption of Pu X-rays in the lungs and the attenuation in tissue overlying the ribs. Since our Pu contains traces of  $^{241}\text{Am}$ , we determined the variations in the ratio of count-rates in Am and Pu peaks with different thicknesses of overlying water for a point source on the axis of a thin NaI(Tl) crystal at a fixed distance of 10 cm. Calculating the same ratio for the phantom, we found ETT for the phantom as 6.3 cm. This value was also confirmed by the observed shift of the 60 KeV peak of  $^{241}\text{Am}$ .

Our future programme of work in this field will include development of phoswich detectors and improvements in the methods of calibration.

#### Acknowledgement

We wish to express our sincere thanks to Shri U.R. Marwah, Technical Physics Division, BARC for the supply of thin NaI(Tl) crystals and to Ss B.J. Vaidya, G. Krishnamachari and J.M. Vidhani for the use of the 400-channel pulse height analyser and instrumentation support. The secretarial help rendered by Ss M. Idicula and G.D. Mistry is deeply appreciated.

#### References

1. Directory of Whole-Body Radioactivity Monitors (1970 Edition) IAEA, Vienna (1970).
2. Sharma, R.C. et al, BARC report (under preparation).
3. Speight, R.G. et al, in "Assessment of Radioactivity in Man", Proceedings of a symposium, IAEA, Vienna (1964) Vol. I, 118.
4. Swinth, K.L. and Griffins, B.I., Health Physics, 19 (1970) 1543.
5. Ishihara, T. et al, Health Physics, 17 (1969) 669.
6. Sharma, R.C. et al, AB Atomenergie, Studsvik Report No. AE-463 (1972).
7. Newton, D. et al, in "Assessment of Radioactive Contamination in Man", Proceedings of a symposium, IAEA, Vienna (1972) 83.

IN VIVO MEASUREMENTS FOLLOWING EXPOSURE TO  $^{133}\text{Xe}$   
AND ASSOCIATED DOSE ASSESSMENT PROCEDURES

Frances Fry  
Atomic Energy Research Establishment Harwell  
Graham Tyler  
Experimental Reactor Establishment, Dounreay

ABSTRACT

Information is presented on the retention and distribution of  $^{133}\text{Xe}$  in the human body as determined by in-vivo counting. Calculations by other workers have shown that for exposure to  $^{133}\text{Xe}$  gas the critical dose is that to the skin. It is shown here, with reference to three cases, how measurements of the body content of  $^{133}\text{Xe}$  made by in-vivo counting can be used to estimate skin doses.

By reference to actual recent cases attention is drawn to problems caused by  $^{133}\text{Xe}$  intakes in the interpretation of external contamination and plutonium-in-lung measurements.

INTRODUCTION

$^{133}\text{Xe}$  is produced in the fission of  $^{235}\text{U}$  with a total yield (direct and by chain) of about 7%. It decays with a half life of 5.27 days to stable  $^{133}\text{Cs}$  by the emission of beta particles of maximum energy 0.34 MeV, gamma rays of energy 81 KeV are emitted in 35.5% of the disintegrations and caesium K X-rays of 30 KeV energy are emitted following 56% of disintegrations. Xenon, a noble gas, which is present in the atmosphere with a partial pressure of about 8 mN/m<sup>2</sup>, is in general chemically inert but appears to combine specifically with haemoglobin (1).

$^{133}\text{Xe}$  has been used extensively in medical science for the investigation of lung function; information on the procedures and computations of the retention of  $^{133}\text{Xe}$  when used for this purpose have been presented by Matthews et al (2). Measurements of the distribution and retention of  $^{133}\text{Xe}$  in the body following both experimental and accidental inhalation in a laboratory manufacturing  $^{133}\text{Xe}$  for medical application have been reported by Venner and Devell (3). Guillot (4) has reported on retention experiments with  $^{133}\text{Xe}$ ,  $^{131m}\text{Xe}$ ,  $^{125}\text{Xe}$  and also stable xenon isotopes. The International Commission on Radiological Protection (5) state that the primary hazard from  $^{133}\text{Xe}$  is from external radiation from submersion in a cloud and calculations of internal doses arising from inhalation of radioactive noble gases presented by Whitton (6) confirm this.

Because it is a gas it is possible for  $^{133}\text{Xe}$  to leak from nuclear reactor fuel elements into operating areas. Whilst the reactor is operating any  $^{133}\text{Xe}$  will usually be accompanied by other fission product noble gases notably  $^{88}\text{Kr}$  (t<sub>1/2</sub> 2.8h) whose daughter product  $^{88}\text{Rb}$  is a solid emitting energetic beta particles; the leak will therefore usually be rapidly detected by conventional filter paper air samplers. Because of its longer half life however  $^{133}\text{Xe}$  may be released a day or so following reactor shut down virtually without the  $^{88}\text{Kr}$ . If the release is a slow one which leads to more or less uniform contamination of the air in the reactor containment conventional external radiation monitor-

ing will adequately detect and assess the hazard. Sometimes however the release may be very localised and although when the activity is dispersed throughout the operating area the resultant radiation level is very low, significant doses may be received by individuals in the immediate vicinity of the release as we will show later.

#### THE UPTAKE AND RETENTION OF $^{133}\text{Xe}$ IN THE HUMAN BODY FOLLOWING INHALATION

As stated by Matthews et al (2) the uptake of  $^{133}\text{Xe}$  into the body is a function of its solubility in blood. However this apparently straightforward situation is complicated by the presence in the atmosphere of naturally occurring stable xenon. The solubility of a gas in a liquid is a function of the partial pressure of the gas and since the normal atmosphere contains xenon at a partial pressure of about  $8 \text{ mN/m}^2$  the body will be saturated with xenon at this partial pressure.

We have made measurements of the retention of  $^{133}\text{Xe}$  in persons who have been exposed to  $^{133}\text{Xe}$  in a reactor environment. Some had been exposed to low level uniform concentrations during reactor operating periods and some to small localised clouds of high concentration following reactor shut down. Measurements made from 1 to a few hours following cessation of exposure to low level uniform concentrations showed the half life of  $^{133}\text{Xe}$  excretion to be about 2 hours. Measurements on persons exposed to small high concentration clouds were made over a longer period up to about 80 hours after exposure and showed retention curves very similar to those reported by Venner and Devell (3), viz an initial rapid excretion phase lasting about 4 hours during which the half life is less than 1 hour followed by a slower elimination rate with half life of about 6 hours for the remainder of the period.

#### DISTRIBUTION OF $^{133}\text{Xe}$ IN THE BODY FOLLOWING INHALATION

We have made measurements of the distribution of xenon-133 in the body (1) after exposure for several hours to low-level contamination and (2) after exposure to a small cloud of high concentration. Profile curves, obtained one day after exposure, are shown in figure 1. Some differences between the curves may be attributed to differences in the scanning techniques. Curve (1) was obtained by scanning with a collimated detector above the supine subject whereas a more finely collimated detector, located under the body, was used for scan (2). The 'depressions' in counting-rate in scan (2) could be due to absorption of the 80 KeV gamma-rays in bone. The 'depressions' in the counting-rate from the chest could be due to attenuation in the ribs and the decrease in counting rate at about 110 cm could be attributed to absorption in the pelvis. These effects would not be so marked with the wider-angle collimator particularly as it was used above the subject. The maxima of the distributions occur at 60-80 cm from the top of the head and could indicate accumulation of xenon-133 in liver. Neither plot shows the large depression in the chest region noted by Venner and Devell (3) in a scan made some hours after an accident inhalation.

#### CALCULATION OF SKIN DOSE FROM IN-VIVO MEASUREMENTS

The critical tissue dose from exposure to  $^{133}\text{Xe}$  is that to the skin. If however the  $^{133}\text{Xe}$  is released in the form of a small cloud normal external radiation monitoring devices will often not give a correct indication. Under those circumstances however it is possible to calculate a skin dose from

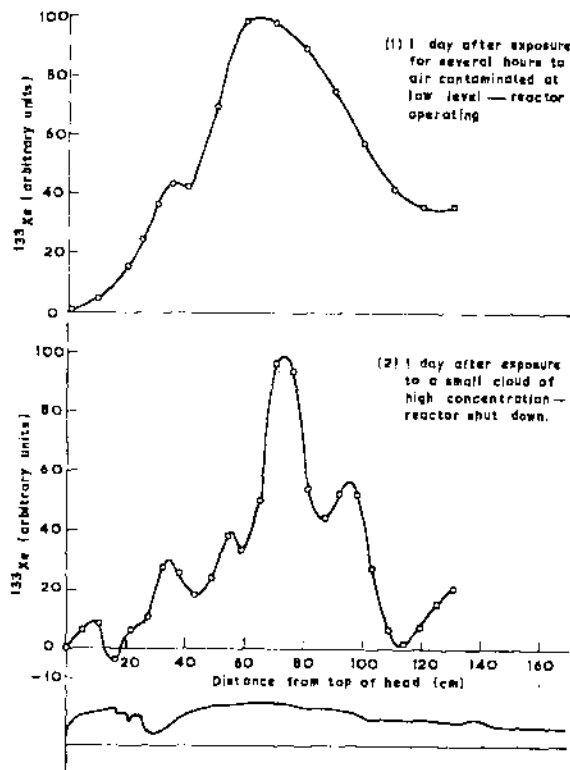


FIG 1 DISTRIBUTION OF  $^{133}\text{Xe}$  IN TWO SUBJECTS

in-vivo measurements. Although the calculation will be subject to considerable error it should be sufficiently accurate to decide whether or not an over exposure had occurred. This is illustrated by the following cases.

#### Case 1

A technician L, removing a thermocouple from a shut down reactor, was working at a glove box containing a mixture of reactor blanket gas and purging argon at a pressure just above atmospheric when a glove was snagged and torn releasing some of the gas mixture. In vivo measurements were carried out on the technician and his assistant who was standing about 2m away at the time of the incident. These both showed  $^{133}\text{Xe}$  but the intake by the assistant was only 1% of that of the technician, confirming that only a small cloud of gas had been involved. In vivo measurements were made on the technician over the period 4-75 hrs after the exposure showed a retention pattern similar to that obtained by Venner and Devell (3) after controlled inhalations; for the period 4-10 hrs the fall was more rapid by about a factor of 2. By extrapolation and interpolation of the retention curve we estimated a body content of about 9 mCi of  $^{133}\text{Xe}$  at 1 hr and 300  $\mu\text{Ci}$  at 10 hours after exposure. The Venner and Devell plot (3) shows that for controlled inhalations the retained amounts of the inhaled activity at those times are 4% and 0.5% respectively. Applying these factors to our estimated body content we obtained estimates of 200 mCi and 60 mCi respectively for the initial inhaled amount. In view of the more rapid fall from 4 hours to 10 hours in our case we chose the higher figure as being a more correct estimate (perhaps even a little on the low side). If the activity was breathed in during some unknown but short period of time, t hours (it is not necessary to know this time), whilst the subject was breathing at the standard man rate of  $1\text{ m}^3$  per hour, the activity concentration in the cloud would have been  $200/t\text{ mCi/m}^3$ . On the assumption that the radius



of the small cloud of gas was about equal to the range of the beta particles (80 cm) conventional calculation showed the beta dose rate to a plane in the centre of the cloud to be  $20/t$  rem/hr.

The gamma dose rate from a cloud of this size would be several orders of magnitude lower and was ignored. When we eliminate our unknown exposure time ( $t$ ) we obtain a skin dose of 20 rems. We were therefore able to say that, despite a film badge recorded dose of only 0.5 rem gamma and 0.4 rem beta, the technician probably received a dose to the skin of his face of more than the 13 week permitted dose (15 rem) but less than 1 year's permitted dose (30 rem) and appropriate administrative action was taken. The most significant internal organ dose from this inhaled quantity was calculated from the information given by Whitton (6) as 1.3 rem to the tracheal mucosa which is much less significant than the skin dose.

### Case 2

Several men became internally contaminated with mixed fission products during removal of a fuel element two days after reactor shut-down.

The most highly contaminated man, subject G, had a body content of 12.8  $\mu$ Ci of  $^{133}\text{Xe}$  21 $\frac{1}{2}$  hours after the release. Measurements made during the following three days indicated that  $^{133}\text{Xe}$  was being removed from the body with an effective half-life of 8.7 hours (biological half-life 9.3 hours): this is in agreement with Venner and Devell's retention curve at this time after inhalation (3).

By extrapolation of the retention curve we estimated a body content at 10 hours of 32  $\mu$ Ci  $^{133}\text{Xe}$ . According to Venner and Devell's data (3), retention of  $^{133}\text{Xe}$  at 10 hours is 0.5% and we therefore estimated the initial body burden as 6.4 mCi. It is probable that most of the intake occurred in a short period of time since the measured general air levels were high ( $>3$  nCi/ml) for about an hour and then dropped to 0.6 nCi/ml and it is also probable that a cloud of much higher concentration existed close to the source for a shorter time. As before we assumed that subject G's initial body burden was acquired in a short period while breathing at 1 m<sup>3</sup>/hr, then calculation of the  $\beta$ -dose rate at the centre of a cloud of gas, as above, showed that subject G may have received a skin dose of 0.6 rem. This is less than the 13-week permitted dose, but greater than the dose to any internal organ. The most significant internal dose was 40 mrem to the tracheal mucosa and the corresponding lung dose was 8 mrem (6).

### Case 3

$^{133}\text{Xe}$  was also identified in subject M, 2 $\frac{1}{2}$  hours after a release of mixed fission products which occurred during removal of a rig from an operating reactor. The total body content of  $^{133}\text{Xe}$  at this time was 0.5  $\mu$ Ci. Several measurements made during the next few hours indicated that xenon was being removed from the body with a biological half-life of 2 hours. However, a further measurement made three days later suggested that there was some long-term retention greater than that predicted by the Venner and Devell retention curve. The faster clearance may correspond to removal of xenon from the water-containing tissues and the longer-term clearance may represent elimination from the less well-perfused fatty regions of the body.

The initial intake was estimated by extrapolating the measured value at 2 $\frac{1}{2}$  hours to 1 hour after intake, with the observed half-life of 2 hours, and

then applying Venner and Devell's figure of 4% retention at one hour. This suggests that subject M's initial intake was 20  $\mu$ Ci. The  $\beta$ -dose to the skin of the face was estimated to be 2 mrem and internal doses to body organs were negligible.

INTERFERENCE FROM INTERNALLY INCORPORATED  $^{133}\text{Xe}$  WITH HEALTH PHYSICS MEASUREMENTS

Internally incorporated  $^{133}\text{Xe}$  gives rise to relatively small internal dose commitments as indicated in Case 1 above where the highest internal organ dose from an initial intake of 200  $\mu$ Ci was calculated as 1.3 rem. However incorporated  $^{133}\text{Xe}$  in much smaller amounts can lead to misleading results being obtained from health physics measurements as is illustrated by the following two occurrences.

i. The technician referred to in Case 1 above monitored his body immediately after the incident and believed himself to be highly contaminated (about 200 x dwl). Several unsuccessful attempts at decontamination by showering were made before a health physicist was consulted who suggested that internally incorporated  $^{133}\text{Xe}$  was the most likely cause; gamma spectrometry of a blood sample confirmed this. Later concurrent in-vivo counting and monitoring with a contamination probe showed that an internal content of 300  $\mu$ Ci gave rise to a counting rate at the surface of the body (using a thin walled, 30 mg/cm<sup>2</sup>, Geiger Muller tube of dimensions 14 cm long and 1.5 cm diameter) of 10 cps equivalent to approximately 2 dwl of skin contamination.

ii. A laboratory worker who normally worked in a laboratory handling  $^{239}\text{Pu}$  went into a reactor operating area to view an experiment on his way to keep an appointment for a  $^{239}\text{Pu}$ -in-lung measurement. This measurement was made

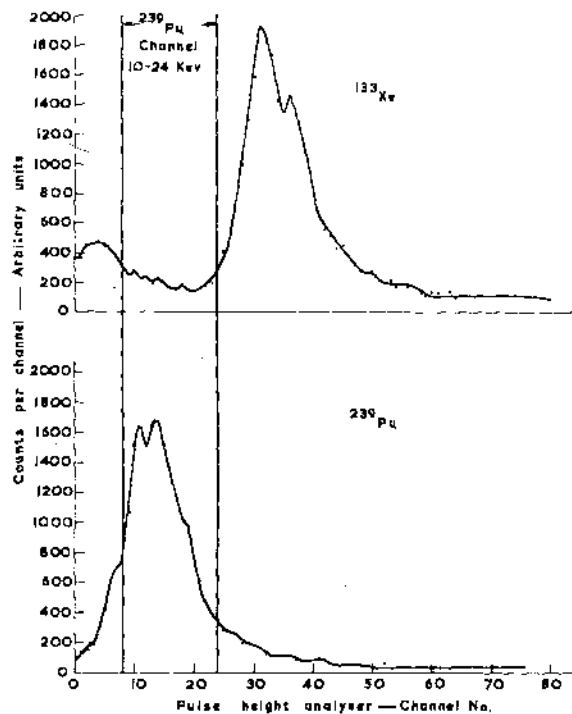


FIG. 2. PROPORTIONAL COUNTER X-RAY SPECTRA.

using a gas filled proportional counter of the type described by Taylor (7). The initial result, which was assessed by the total counting rate in the 17 KeV  $^{239}\text{Pu}$  channel, caused some concern as it was equivalent to more than 100 times the maximum permissible lung burden. Examination of the spectrum from the counter however showed peaks at about 30 KeV and 5 KeV and  $^{133}\text{Xe}$  was suspected.

A further count using a scintillation detector indicated a body content of about 0.6  $\mu\text{Ci}$  of  $^{133}\text{Xe}$  and a repeat measurement with the proportional counter a few days later showed no activity in the plutonium channel. A plot of the spectrum from  $^{133}\text{Xe}$  in the body as given by the proportional counter together with a spectrum of  $^{239}\text{Pu}$  for comparison is shown in Figure 2.

#### CONCLUSIONS

In-vivo measurements made on men exposed to air contaminated with xenon-133 in nuclear reactor environments show retention patterns similar to those reported (2). Elimination of xenon-133 is a complicated function of time, indicating that many body compartments are involved in the uptake and retention. Uptake, retention and also distribution within the body may vary depending upon the partial pressure of the inhaled xenon. Profile scanning measurements suggest that the distribution within the body is different for the two modes of uptake discussed. The reasons for this are not readily apparent, but some of the differences between the two profile curves may be due to different scanning techniques.

The critical dose from exposure to  $^{133}\text{Xe}$  is that to the skin but, as we have shown, the results of in vivo measurements of the body content and elimination rates may be used to calculate this dose.

$^{133}\text{Xe}$  incorporated in the body in amounts which give rise to trivial doses of radiation can interfere with health physics measurements and health physicists for reactor areas should be aware of the possibilities.

#### REFERENCES

1. Schoenborn, B P, Binding of xenon to horse haemoglobin. Nature 208, pp 760-763 (1965).
2. Matthews, C M E, Dollery, C T, Clark J C and West, J B, USAEC Symposium 6 Radioactive Pharmaceuticals, p567 (1966) Conf 65111.
3. Venner, L and Devell, L, Retention of  $^{133}\text{Xe}$  after inhalation. Nordic Radiation Protection Conf. Oslo 2-4 October 1968.
4. Guillot, P. Nonlinear relationship between radio xenon inhaled activity and whole-body activity; radio xenon retention differs from stable xenon retention in man and rat. Presented at 2nd European Congress of IRPA Budapest, 3-5 May 1972.
5. Recommendations of the International Commission on Radiological Protection. ICRP Publication 2. Pergamon Press (1959).
6. Whitton, J T (Mrs), Doses arising from inhalation of noble gases. CEGB RD/B/N1274 (Dec 1968).
7. Taylor, B T, Health Physics 1969, Vol 17, pp 59-69.

## In-vivo monitoring of nuclear fuel workers

C. POMROY, J. GORDON and E.G. LETOURNEAU  
Radiation Protection Bureau,  
Department of National Health and Welfare,  
Ottawa, Ontario, Canada K1A 1G1

### Introduction

As well as being a major source of uranium, Canada possesses a sizable uranium processing industry. Several plants located in Southern Ontario fabricate fuel rods for use in the Canadian nuclear power industry and also for export.

Working conditions in these plants are specified by the Atomic Energy Control Board, and monitoring of workers by means of criticality badges and urinalysis is performed by the Radiation Protection Bureau and the Ontario Department of Health.

Urinalysis results frequently show evidence of uranium ingestion, but the relation of this to total body burden is uncertain and a direct, in-vivo method of body burden measurement is required.

Coffield (1) made such measurements using a standard whole body counter in a steel room, and Quastel et al (2) of this Bureau made similar measurements, together with extensive urinalysis and other biological measurements, on 15 uranium workers. This study confirmed that body burden could not be accurately estimated from daily urinary excretion.

Unfortunately, the use of a conventional whole body counter means that the subjects have to travel to Ottawa, a distance of 250 miles from the major uranium processing plants, and while this can be arranged for small sample populations it is obviously impractical for the plants' total exposed work force. It was therefore necessary to move the equipment to the plants, and the present study was undertaken to see whether a portable system would have sufficient sensitivity to be useful.

### Theory

The usable radiations from U-Nat. are as follows:

U-238 - none	U-235 - 185 keV (54%)
Th-234 - 63 keV (3.5%) 93 keV (4%)	Th-231 - 84 keV (10%)

Since the subjects to be monitored had long exposures to uranium it was considered reasonable to assume secular equilibrium of the ingested material, which being of refined reactor grade had a very low content of radium or radium daughters. It was therefore decided to use the low energy radiations

from the thorium daughter, for which a 20 cm diameter x 3 mm thick NaI crystal, already available, would have a high efficiency. The response of this detector to a U-Nat. phantom is shown in fig. 1 as a broad peak extending from around 50 to 90 keV. The detector has negligible sensitivity to the 185 keV gammas from U-235.

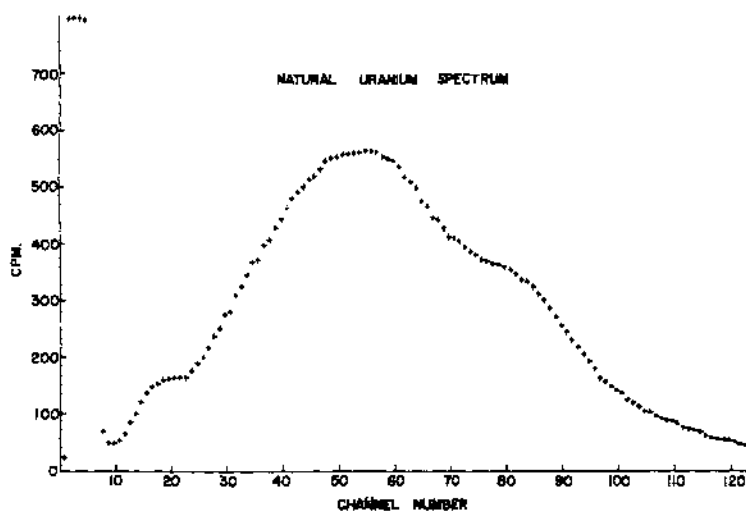


Fig. 1 Response of 20 cm x 3 mm NaI crystal to U-Nat. phantom

The phantom used for calibration purposes was a Remcal (Alderson Research Laboratories Inc.) phantom, with 9.2 g of uranium oxide dust in the lung cavities. The dust was sprayed onto adhesive coated paper tissues which were then made to adhere to the inside of four polythene bags. One bag was introduced into each of the four lung cavities and inflated with air. The basal lobes contained about four times as much uranium as the apical lobes.

The remainder of the phantom was filled with water and the net count rate observed was 3.86 cpm/mg U-Nat., which with a background of 3000 cpm gives an  $S^2/B$  ratio of 0.0049.

Using the formula of Altshuler and Pasternak (3), the minimum significant measured activity is 12.4 mg and the minimum detectable true activity is 24.8 mg, these figures being for a single 10 min. count. The crystal is also sensitive to scattered radiation from Cs-137 and K-40 to the extent of 8 cpm/nCi Cs-137 and 0.5 cpm/g K. For a normal 70 kg adult with a body burden of 5 nCi Cs-137 and containing 180 g K this would amount to a count rate of 130 cpm. Since the thin crystal is not sensitive to the primary radiation from either Cs-137 or K-40 it is not possible to measure each subject's content of these isotopes directly. The count rate from them has instead been considered part of the subject background, which is discussed further below.

#### Description of apparatus

The detector is a 3 mm thick by 20 cm diameter NaI (Tl activated) crystal coupled to 3 low-noise PM tubes. Each PM tube has its own H.V. supply, but the signals are collected by a common cable and fed into a single preamplifier and thence to the amplifier which is an integral part of the

Technical Measurement Corporation pulse height analyser. The date is recorded by either a TMC parallel printer or a Tally paper tape punch.

The settings of the H.V. Supplies are adjusted by placing a small uranium source on the crystal axis at a distance of about 40 cms. and switching on one H.V. supply at a time, and adjusting them so that the three spectra overlap as precisely as possible.

The detector is housed in a stainless steel cylinder which is lined with 1/8" lead and supported on a counterbalanced stand. See fig. 2.

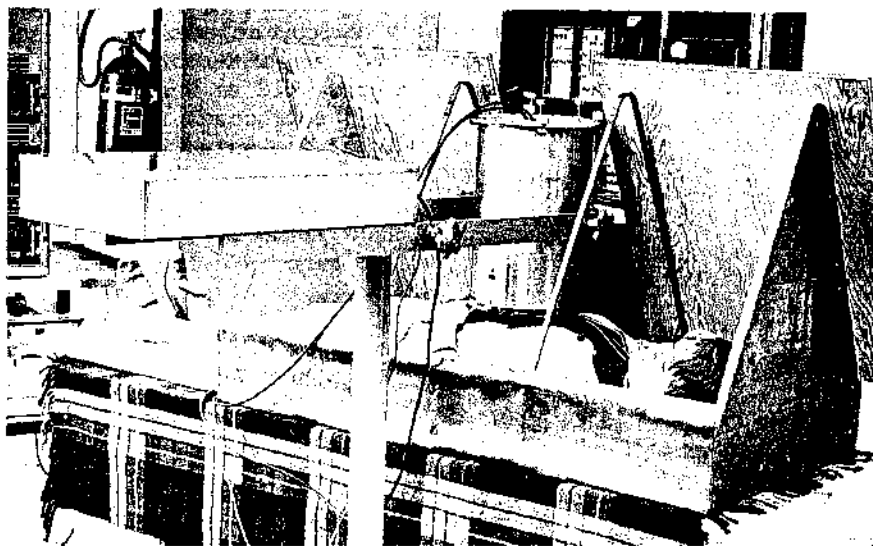


Fig. 2 Detector, counterbalanced stand and lead tent with near-side panels removed.

The shielding is based on a design by Eisenbud et al (4) and consists of 1/8" lead sheet fastened to plywood and placed over a tent shaped framework. A sheet of 1/8" lead is placed under the mattress on which the subject lies.

The effect of this shielding is to reduce the subject background in the 30-100 keV region from 9,500 cpm to 3,000 cpm.

This equipment is portable, the total weight being about 600 lbs. Two men can load it into a station wagon in about 30 mins. and on arrival at the counting location, the system can be unloaded and assembled in about an hour. A room about 10 ft square is sufficient to house the equipment and the counting bed, located away from any active area. So far no unduly high background rates have been encountered, despite the fact that the plants visited process large quantities of radioactive material.

#### Subject Background

Preliminary work in the laboratory indicated that the background count

on a subject inside the tent varied considerably, depending on his size and shape. This variation was much more than would be expected just from differences in Cs-137 and K-40 content. Therefore forty Radiation Protection Bureau staff members were studied and their backgrounds were measured together with their height, weight and "chest thickness", i.e. the front to back measurement of chest thickness made at inspiration. The latter measurement was chosen as the simplest one to give some index of the bulk of tissue under the detector.

A series of empirically chosen expressions combining the three anthropomorphic parameters were analysed using a least squares method, for their correlation with the observed count rate. The expressions were of the type  $X = (W/H) \log C$  or  $X = \exp (W/H)$  etc. and the correlation coefficients were all quite similar at around 0.84. The highest coefficient, 0.851, was obtained with the expression  $X = (W/H)\sqrt{C}$  which yielded an equation for Y, the subject background;  $Y = K (3517.4 + 259.46X)$  where K = correction factor to allow for differences in ambient background between our laboratory and other counting locations. A plot of Y vs X is shown in fig. 3, which includes the 95% confidence limits for a single estimate of Y given a value of X.

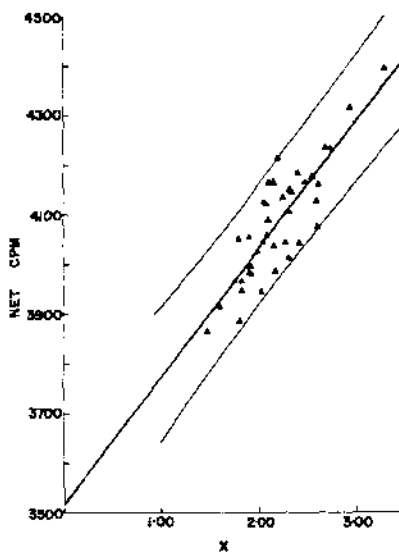


Fig. 3 Plot of observed subject background count-rate vs X where  $X = \text{weight/height} \times \sqrt{\text{chest thickness}}$ . (The 95% confidence limits are shown)

In terms of cpm these confidence limits represent  $\pm 120$  cpm on a typical subject background of 3000 cpm (K being significantly less than one at the plant where the bulk of the subjects were counted), whereas counting error for 10 min. count = 17.3 cpm. This error is more or less equal to the count rate expected from 30 mg U-Nat., the maximum permissible body burden. This means that only burdens in excess of the maximum permissible can be confidently detected. In an attempt to improve this situation, a more detailed multiple regression test was run on the data. This analysis was carried to the point where an equation containing six terms was derived, but the correlation coefficient associated with this equation was 0.8631, which was not significantly greater than with the much simpler equation originally chosen.

### Other corrections

In dealing with radiations at 100 keV or below, tissue absorption must be taken into account. On the assumption that the uranium is located in the lungs or pulmonary lymph nodes, it is necessary to make some estimate of the thickness of tissue overlying these organs. Deane (5) has measured this thickness ultrasonically and correlated it with the weight/height ratio of each subject. His formula is as follows:

$$T \text{ (thickness (mm))} = 0.071 + 0.512 \frac{W \text{ (weight (kg))}}{H \text{ (height (m))}}$$

Ramsden et al (6) did a similar study, and obtained a different formula, which included a measurement of the chest circumference (C)

$$T \text{ (cm)} = 15.3 \frac{W \text{ (kg)}}{H \text{ (cm)}} - 0.01 C \text{ (cm)} - 3.55$$

We were not able to obtain an ultrasonic device and make actual measurements, and so T was calculated both ways and the mean taken, for each subject. As there were differences between the thicknesses obtained with the two formulae, in some cases as much as 25%, a mean value was taken. This was used to read off the appropriate absorption correction from fig. 4, the error associated with this procedure being estimated as  $\pm 4\%$ , due to the relatively small slope of fig. 4.

Newton et al (7) in their work on the measurement of plutonium in the lungs, took into consideration the self-absorption of the lung tissue itself, but for the present work, in view of the absorption curve in fig. 4 this has been ignored.

### Collection of data

In the fall of 1971 a brief visit was made to a uranium processing plant in Southern Ontario. This was the first trip with the portable equipment and the main object was to see whether it could be transported and set up easily, and would be stable in operation. The time from arrival at the plant to counting the calibration standard was about  $1\frac{1}{2}$  hours, and frequent counting of the calibration standard indicated that the response of the system was very stable.

The plant management made a small room 10' x 10' available to us in the administration area of the building and the subjects arrived after a shower and change of clothing. The subjects were 3 workers who had been studied five years earlier by this laboratory (2).

The subjects were counted supine with the 8" detector in contact with the chest surface. The counting time was 10 min.

The results from this preliminary study, while showing some counts above the background of a normal subject, were calculated without the benefit of the subject background equation subsequently developed, and were therefore considered merely an indication that the system could work and that a further field trip to study a larger sample of workers would be justified.

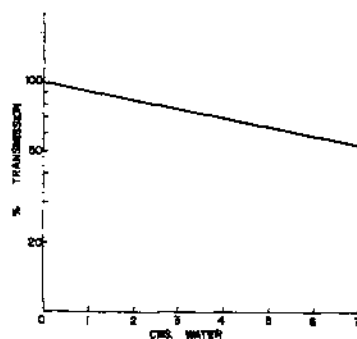


Fig. 4 Relative transmission of U-Nat. radiations in 60-90 keV region in water.



Such a trip was undertaken some time later, and involved a larger nuclear fuel processing plant. This plant contained a large stock-pile of uranium and there were some misgivings as to what the background count would be. It turned out however to be not significantly different from that at the other plant.

A total of twenty-four subjects were counted, with results as shown in table 1, calculated on the assumption that only U-Nat., in equilibrium, was present.

Table 1  
Results of Measurements in Nuclear Fuel Workers

Subject #	$X$ $(\frac{W}{H} \sqrt{C})$	Estimated background c.p.m. <sup>1</sup>	Observed count rate c.p.m. <sup>2</sup>	Net count rate c.p.m.	Absorption correction* <sup>3</sup>	mg U-Nat.
1	2.47	3056 ± 125	3298 ± 18	242 ± 126	0.78	62 ± 25
2	1.79	2927 ± 121	3401 ± 18	474 ± 122	0.87	109 ± 24
3	2.15	2995 ± 122	3005 ± 17	11 ± 123	0.82	3 ± 24
4	1.93	2953 ± 121	2975 ± 17	22 ± 122	0.85	5 ± 24
5	1.87	2941 ± 120	2981 ± 17	40 ± 121	0.87	9 ± 24
6	2.36	3035 ± 120	3047 ± 17	12 ± 121	0.81	3 ± 24
7	2.42	3046 ± 123	3011 ± 17	-35 ± 124	0.80	-
8	2.09	2985 ± 123	3244 ± 18	259 ± 124	0.83	62 ± 25
9	1.76	2920 ± 120	3090 ± 17	170 ± 121	0.88	38 ± 24
10	1.71	2912 ± 122	3018 ± 17	106 ± 123	0.87	24 ± 24
11	2.17	2999 ± 123	3217 ± 18	218 ± 124	0.82	53 ± 25
12	1.77	2919 ± 122	3204 ± 18	285 ± 123	0.88	64 ± 24
13	1.57	3075 ± 121	3041 ± 17	-34 ± 122	0.77	-
14	2.20	3005 ± 123	3291 ± 18	286 ± 124	0.86	66 ± 25
15	1.74	2917 ± 120	2984 ± 17	67 ± 121	0.89	15 ± 24
16	1.87	2941 ± 120	2968 ± 17	29 ± 121	0.87	6 ± 24
17	2.68	3097 ± 123	3326 ± 18	229 ± 124	0.77	59 ± 25
18	2.36	3035 ± 120	3349 ± 18	314 ± 121	0.80	78 ± 24
19	2.97	3151 ± 128	3282 ± 18	131 ± 129	0.75	35 ± 26
20	1.89	2945 ± 120	3211 ± 18	266 ± 121	0.86	62 ± 24
21	2.36	3035 ± 120	3374 ± 18	239 ± 121	0.80	84 ± 24
22	2.07	2980 ± 122	3047 ± 17	67 ± 123	0.83	16 ± 24
23	1.90	2948 ± 120	4908 ± 22	1960 ± 121	0.85	459 ± 24
24	2.07	2983 ± 123	2947 ± 17	-36 ± 124	0.83	-

<sup>1</sup> Errors estimated from fig. 3  
<sup>2</sup> Counting error only  
<sup>3</sup> Correction factor read from fig. 4 using tissue thickness calculated as described in text.

As will be noted some had very high values, but subsequent discussion with the plant health physicist revealed that these had worked in the UF<sub>6</sub> plant, and during the processing of this material, the thorium does not enter the gaseous phase and is left behind in the "ash". It was therefore assumed that the high readings were due, at least in part, to the inhalation of thorium 234, during the handling of this "ash", and measurements taken on some of the UF<sub>6</sub> workers six months later were down to much lower levels, which tended to confirm this assumption.

## Discussion

The results from these field trips indicate that a significant proportion of the workers examined were contaminated with uranium or its daughters. The errors, mainly due to uncertainties in the background count estimate for each subject, were such that only lung burdens greater than 30 mg could be detected with any confidence, and in some cases the material detected could have been mainly 24 day Th-234 rather than U-Nat.

The problem of identification is in fact two-fold, first to identify the degree of enrichment of the uranium, and second, to differentiate thorium from uranium. A recently acquired dual crystal (Phoswich\*) detector has been put to use in some preliminary experiments to look into these problems.

The detector is a 12.5 cm diameter, 1 mm thick NaI (Tl activated) crystal backed by a 12.5 cm diameter, 5 cm thick CsI (Tl activated) crystal, and a pulse shape discrimination circuit (Harshaw NC-25) is used to allow only those photons that are completely absorbed in the thin crystal to be recorded. The sensitivity of this detector is 1.25 cpm/mg U-Nat. (50 keV - 110 keV) for an average subject background of 600 cpm. If these figures are corrected to compensate for the smaller area of the 12.5 cm detector compared to the 20 cm detector, an  $S^2/B$  ratio of .0067 is obtained. This is better than the figure of .0049 for the 20 cm detector, but could probably be improved further by the use of a thicker front (NaI) crystal, of say, 3 mm.

The natural uranium source gives a spectrum with well-defined peaks at about 60 keV and 90 keV, and the change in relative heights of the peaks with changing enrichment was investigated, on the grounds that the Th-231 from U-235, with its gammas at around 84 keV would affect the 90 keV peak more than that at 60 keV. The peak ratio being defined as  $\frac{\text{ch. 103-162}}{\text{ch. 163-215}}$  at a gain of 0.48 keV/ch.

The plot of peak ratio vs enrichment is shown in fig. 5, and there is a marked drop in the 60 keV/90 keV ratio with increasing enrichment up to about 20%. Beyond that the curve levels out but since the material used in the plants under investigation rarely exceeds 20% enrichment, this should not detract from the utility of the curve in estimating the enrichment of the uranium present.

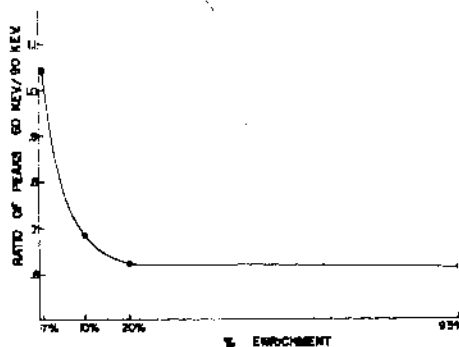


Fig. 5 Ratio of 60 keV/90 keV peaks vs enrichment

The above measurements were made with sources of high activity and so the net count rates were high. With actual subjects the net count rates would normally be much lower, and prone to the errors involved in estimating subject body background discussed already.

(\* Harshaw Chemical Co. Ltd.)

Future work with the dual crystal will include an experiment to determine the relationship between the 60 keV/90 keV peak ratio and tissue thickness, and another to explore any correlation between the body background of a normal subject in the 60 keV-90 keV region and that in the 200 keV and above region, using the 5 cm thick CsI crystal as a detector and counting all the events occurring within it. The range from 200 keV upwards would include contributions from Cs-137 and K-40 but not from the 185 keV line of U-235.

It would also be very useful if the 185 keV line of U-235 could be detected by the CsI crystal in order to distinguish between uranium and separated thorium but early results indicate that the background counts in that region are too high for sufficient sensitivity.

### Conclusions

The study shows that given certain conditions, the 20 cm x 3 mm single crystal with portable shielding can detect lung burdens in the region of 30 mg U-Nat. The conditions are that only U-Nat. is present, in equilibrium with its thorium daughters, and that no separated thorium is present.

In the major uranium processing plant studied these conditions hold for a substantial portion of the personnel, and those for which they do not hold can be identified. The system can, therefore, give useful information, and is suitable for routine monitoring within the stated limits.

A dual crystal (Phoswich) system, however, offers considerable promise for refining the method to determine enrichment and to improve the subject background estimates, and hence the sensitivity. The problem of separated Th-234 will require either a much more substantial counter of the shadow-shield type, or a method of repeated counting to detect the decay of the thorium.

### References

1. Cofield, R.E. Health Physics 2, 269 (1960)
2. Quastel, M.R., Taniguchi, H., Overton, T.R. and Abbatt, J.D. Health Physics 18, 233 (1970)
3. Altshuler, B., Pasternak, B. Health Physics 9, 293 (1963)
4. Eisaenbud, M., Laurer, G.R., Rosen, C.R., Cohen, N., Thomas, J. and Hazle, A.J. Health Physics 16, 637 (1969)
5. Deane, P.N. Report #LA-DC-72-958 (1972)  
(Los Alamos Scientific Laboratory)
6. Ramsden, D., Peabody, C.O. and Speight, R.G. UKAEA Report AEEW-R 493 (1967)  
(H.M. Stationery Office, London)
7. Newton, D., Fry, F.A., Taylor, B.T. and Eagle, M.C. IAEA-SM-150/11 Assessment of Radioactive contamination in man. p83 IAEA Vienna, 1972.

## APPLICATION OF PHOSWICH DETECTORS FOR LUNG COUNTING PLUTONIUM-238

F. Keith Tomlinson, Ralph Brown,  
Harold Anderson and Bobby Robinson  
Mound Laboratory\*  
Miamisburg, Ohio

### Abstract

Mound Laboratory's Whole Body Counter was designed and calibrated for the detection of  $^{238}\text{Pu}$  in the lungs. This paper summarizes the basic counting program since December 1969. The primary discussion is centered around the phoswich detection system. A unique triple coincidence pulse shape discrimination technique was used to reduce the background more than one order of magnitude as compared to a standard NaI(Tl) detector. Detection limits are given as a function of the subject's tissue thickness between the lungs and detectors. For a typical subject with an effective tissue thickness of 2.3 cm over the lungs, the system has a detection limit of 4 nCi.

### Introduction

Numerous laboratories have employees who work daily with plutonium. At Mound Laboratory about 450 employees are routinely monitored for  $^{238}\text{Pu}$  as part of the overall radiological health protection program. An important part of the program is the routine and special lung counting which gives a direct assessment of the most common mode of uptake - inhalation.

This paper briefly summarizes the development of lung counting capabilities since 1969 with the primary emphasis on the phoswich detector system. The sensitivity of this system lies in the pulse shape discrimination instrumentation which is used to lower the background by more than one order of magnitude compared to a standard NaI(Tl) detector. The major problems of implementing the detector/pulse shape discrimination system are also discussed. Also included is a discussion of detection limits as a function of the chestwall tissue thickness where the chestwall is the primary absorber of low energy photons emanating from the lung.

### Historical Development

Detectors The radiation safety program at Mound Laboratory was upgraded in 1969 with the completion of the Body Counting Facility. The design of the facility was reasonably standard with a Packard Instrument Company<sup>1</sup> steel room and a semi-aged air supply from the crawl space in the adjacent administration building. In 1969, two standard NaI(Tl) detectors, 10.2 cm diam by 0.4 cm thick, were coupled through amplifiers to a multichannel analyzer. Room background in the 6-27 keV band was about 0.45 count/min/cm<sup>2</sup>. A typical count on an unexposed individual was 0.542 count/min/cm<sup>2</sup> with a minimum detectable activity of about 11 nCi of  $^{238}\text{Pu}$  (approximately 3/4 m.p.l.) using a 4000-sec count.

\*Mound Laboratory is operated by Monsanto Research Corporation for the U.S. Atomic Energy Commission under Contract No. AT-33-1-GEN-53.

A diagram of the phoswich detectors purchased from Harshaw Chemical Company<sup>2</sup> in July 1969 is shown in Fig. 1. By May 1970 the two phoswich detectors were in use with a significant improvement in sensitivity over the standard NaI(Tl) detectors. Room background was about 0.0185 count/min/cm<sup>2</sup> in the 14-25 keV band; however, the pulse shape discrimination system resulted in a loss of about 6% of the detected '17 keV' photons. Even so, a typical background of an exposed individual was only 0.0346 count/min/cm<sup>2</sup> (14-25 keV) which allowed a minimum detectable activity<sup>3</sup> (3 $\sigma$  above background) of 5.1 nCi of <sup>238</sup>Pu.

In October 1972, two additional phoswich detectors, 12.7 cm diam, were purchased; the only difference was the larger diameter of the crystals. With the same pulse shape discrimination system, the room background decreased to 0.0178 count/min/cm<sup>2</sup> and the minimum detectable activity was slightly improved at 4.0 nCi of <sup>238</sup>Pu.

Calibration Procedures The most prominent photons for counting <sup>238</sup>Pu in-vivo are the <sup>234</sup>U L x-rays with an average energy of 17 keV. The total counts in the complete 17 keV region, i.e., from 5-28 keV, were originally integrated for analysis. However, it was soon discovered that 14-25 keV was the optimum area of integration. This was determined by maximizing S<sup>2</sup>/B, where S is the net count rate and B is the background. Originally, a simple two point calibration curve was used for lung deposition assessments as shown in Fig. 2.

The two points used for the exponential fit were obtained by counting the Remab phantom<sup>4</sup> full and then one-half full of liquid. This gave two chestwall thicknesses, i.e., two different thickness absorbers, at which the counting rate per unit activity was measured. This was knowingly in slight error for thin chestwalls because of the lung-to-detector distance with the phantom one-half

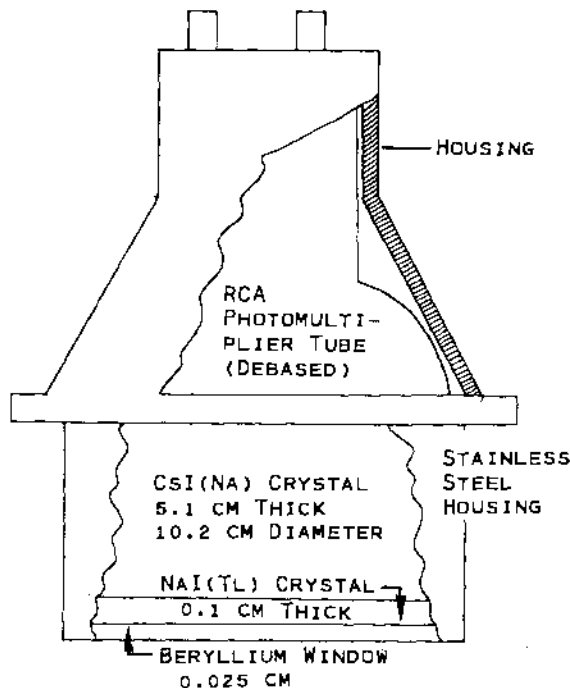


Fig. 1 Phoswich detector diagram.

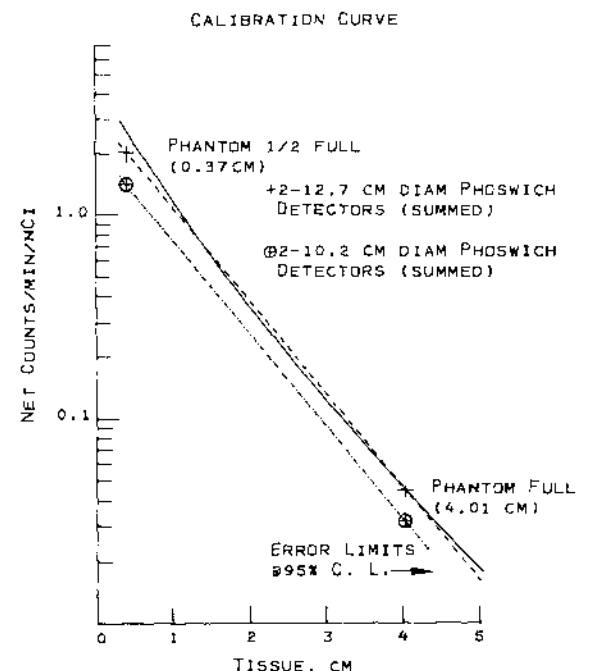


Fig. 2 Calibration curve.

full of liquid. This error was later corrected by developing the proper shaped curve using  $^{238}\text{Pu}$  doped lungs from the phantom and beefsteak absorbers.<sup>5</sup> Once the correctly formed calibration curve existed, it was overlaid and correlated to the one suitable phantom measurement where the phantom was full of liquid and had an equivalent chestwall thickness of 4.04 cm (See Fig. 2).

Counting Procedures As part of the overall radiological health program, all plutonium workers are routinely counted on a quarterly basis. Any employe is promptly scheduled for a special lung count upon discovering 200 dis/min or more on a nasal swab. More than 50% of those needing special counts have had at least a small amount of surface contamination on various parts of their bodies. One major problem encountered in special counting is determining whether the plutonium was detected from within the lungs or from contamination of the chest surface. As little as 50 dis/min on the skin's surface can cause a false reporting of a Type B Incident. Procedures used at Mound Laboratory to eliminate "false" interpretation of data are outlined below:

1. Carrying out extremely thorough surface decontamination, including "washing" the subjects chest with ethylene-diaminetetrachloroacetic acid.
2. Placing lead loaded gloves on the subjects hands and arms.
3. Counting the subject with a lead shot filled curtain around each detector for shielding.
4. Requiring a confirming lung count taken from the subjects back.

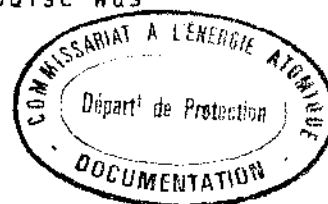
#### Instrumentation

The two phoswich detectors currently used at Mound Laboratory are summed together into the pulse shape discrimination system as shown in Fig. 3. Because of the two dissimilar crystals in a phoswich detector, each output pulse will be characteristically shaped by the crystal in which the absorbed photon lost its energy. Since the pulse shape discrimination system is aligned to accept only those low energy (17 keV) [NaI(Tl)] pulses, noise and most high energy background pulses [CsI(Na)] are rejected. About a 5-8% loss of detector efficiency has been experienced using the pulse shape discrimination system.

#### Discussion

The operation of the pulse shape discrimination system is rather unique. Initially the system was set up to discriminate by using only rise time and crossover times as shown outside the dotted lines in Fig. 3. Because the room background of 0.053 count/min/cm<sup>2</sup> (14-25 keV) was not as low as expected, the system was thoroughly reinvestigated. Extraneous background counts were caused by cosmic radiation. Typical amplifier output pulses appeared similar to those shown by solid lines in Fig. 4.

Careful examination of the cosmic ray overload pulses resulted in the discovery of a preamplifier bleed-off pulse following the initial saturated pulse by 40-80  $\mu\text{sec}$ . This bleed-off pulse was



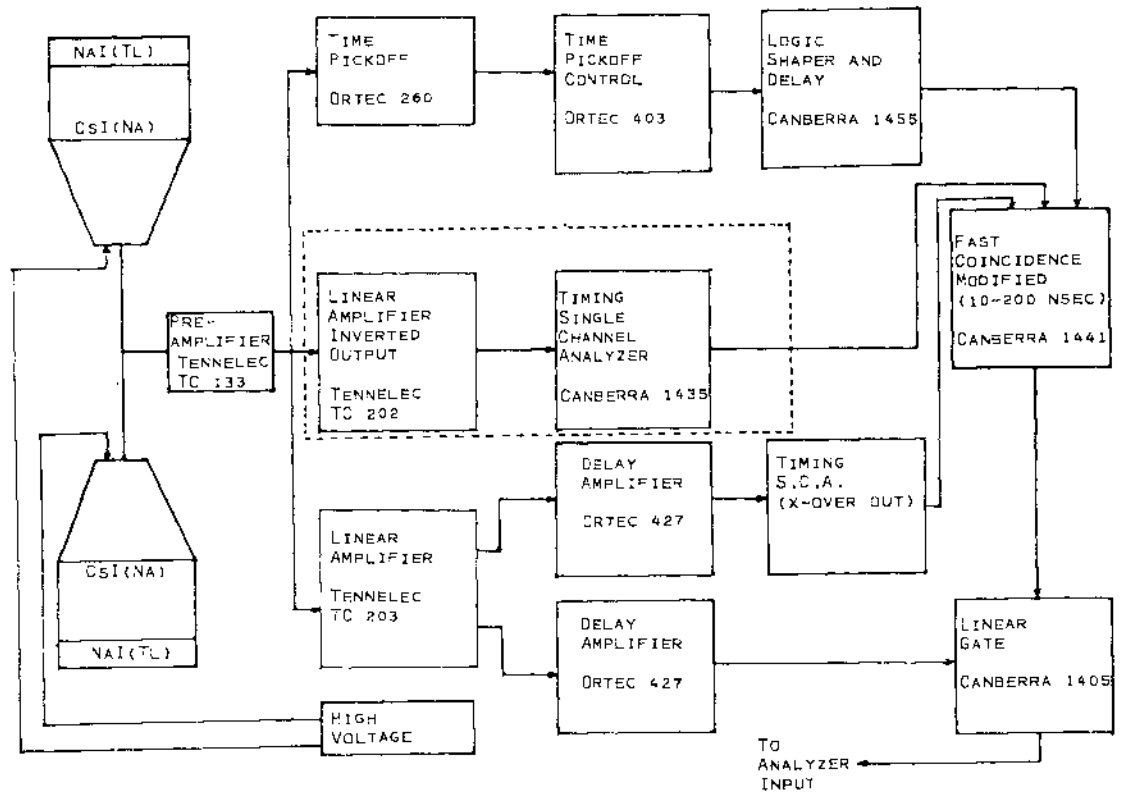


Fig. 3 Pulse shape discrimination system.

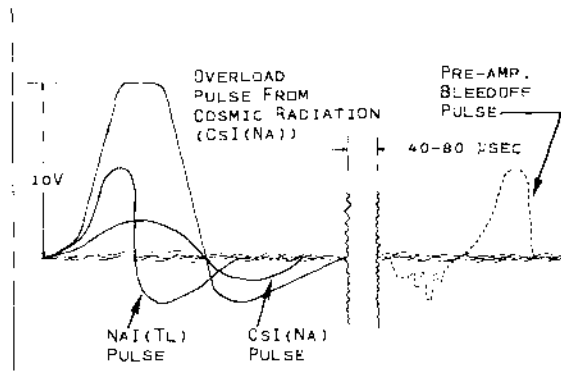


Fig. 4 Amplifier output pulses.

similar to the '17 keV' pulses from  $^{238}\text{Pu}$ . A portion of these bleed-off pulses would pass the discrimination system and cause extraneous background counts in the 17 keV region. Two solutions existed for this problem. The first and simplest was to use an integral discriminator to inhibit the system output for about 100  $\mu\text{sec}$  after the detection of any saturating pulse. The dead time is insignificant at the count rate of interest, but can be determined. The second method (in current use) to eliminate the extraneous bleed-off pulses made use of the bleed-off pulse shape. By using an inverted bipolar pulse input to a timing single channel analyzer in the leading edge mode, only those bipolar pulses with a second lobe were accepted. (See Fig. 3).

In November 1969, efficiency measurements using the internal gate of the multichannel analyzer indicated an electronic problem in the system. The efficiency for the

system decreased when the distance between the source and the detector was increased. Numerous efficiency measurements were made using the analyzer gate and then compared to results obtained under the same counting conditions with an external linear gate. The efficiency of the system using either gate was approximately 90% at counting rates of 2000 counts/min. At counting rates less than 500 counts/min, the efficiency of the system dropped to less than 50% when the analyzer gate was used, but remained at about 90% with the external gate. The unusual behavior of the analyzer gate was examined further by letting the coincidence logic pulse trigger an oscilloscope simultaneously with the linear gate in the analyzer. Observation of both the oscilloscope and the analyzer's visual display revealed that many pulses which triggered the oscilloscope did not register on the analyzer's display. Such pulses were not being stored in the analyzer's memory. It was later confirmed by the manufacturer that indeed there was a design error in the analyzer's gate. Prior to resolving the problem, abnormally low background counting rates were observed.

### Results

The comparison of a standard NaI(Tl) detector system versus a phoswich detector system of the same active area, shows an unquestionable improvement for 17 keV photons in  $^{239}\text{Pu}$  lung counting. A direct comparison is shown in Table 1.

Table 1

Standard NaI(Tl) Detectors Compared with Phoswich Detectors

Detector	Steel Room Background from 14-25 keV (counts/min/cm <sup>2</sup> )	Unexposed Person Count from 14-25 keV (counts/min/cm <sup>2</sup> )	Minimum Detectable Activity* (nCi)
Std. NaI(Tl) 10.2 cm diameter 0.4 cm thick	0.245	0.276	11
Phoswich Detector 12.7 cm diameter 0.1 cm thick NaI(Tl) 12.7 cm diameter 5.1 cm thick CsI(Na)	0.0178	0.0274	4

\*3 $\sigma$  above background (2.31 cm chestwall thickness).

The minimum detectable activity<sup>3</sup> for  $^{239}\text{Pu}$  lung counting has been improved by about a factor of three by changing to the phoswich detector system. The minimum detectable activity as a function of chestwall tissue thickness and unexposed subject counting rate is shown in Fig. 5.



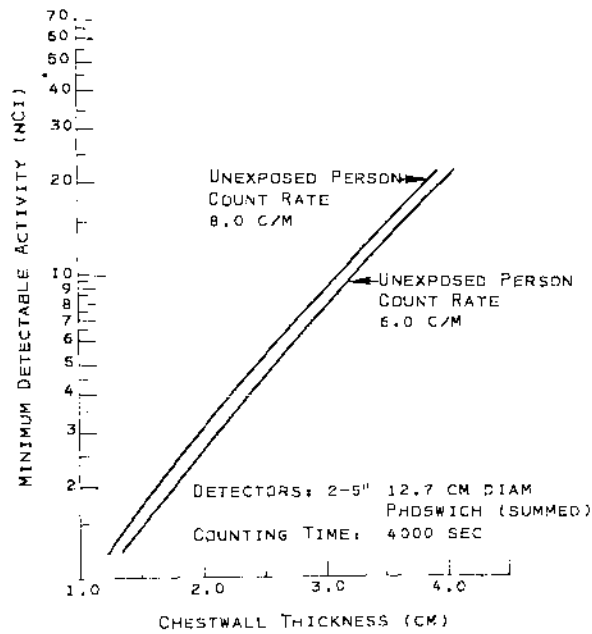


Fig. 5 Minimum detectable activity curve.

#### References

1. Packard Instrument Company, Inc., Downers Grove, Illinois, USA.
2. The Harshaw Chemical Company, Solon, Ohio, USA.
3. A Manual of Radioactivity Procedures, Handbook 80, U. S. Dept. of Commerce, National Bureau of Standards, pp 27-28 (1961).
4. Nuclear Associates, Inc., 35 Uran Avenue, Westbury, New York, 11590, USA.
5. F. Keith Tomlinson, Ralph Brown, Harold F. Anderson, and Bobby Robinson, "Chestwall Tissue Measurements for Lung Counting Applications" (Published in these proceedings.)
6. Notification, Investigation and Reporting of Occurrences, Operational Safety Handbook, U. S. Atomic Energy Commission, AEC Appendix 0502.

## CHESTWALL TISSUE MEASUREMENTS FOR LUNG COUNTING APPLICATIONS

F. Keith Tomlinson, Ralph Brown,  
Harold Anderson and Bobby Robinson  
Mound Laboratory\*  
Miamisburg, Ohio

### Abstract

Mound Laboratory's Whole Body Counter was designed and calibrated for the detection of  $^{239}\text{Pu}$  in the lungs.

Quantitative measurements depend upon the detection of the 17 keV (average) x-ray associated with the decay of  $^{239}\text{Pu}$ . Because the half-value layer for 17 keV x-rays in tissue is only 6-7 mm, the effective thickness of the tissue overlaying the lungs must be accurately determined for proper interpretation of the counting data.

The tissue thickness over the lungs is determined by ultrasonic measurements over the second, third, and fourth rib in the manner suggested by Ramsden, Peabody and Speight.

This paper presents a review of the instrumentation and technique used at Mound Laboratory to obtain the tissue thickness measurement based on our experience in making these measurements on more than 700 different persons.

### Introduction

During the last several years, various ultrasonic instruments have been used for determining human chestwall thickness. The chestwall thickness is extremely important where low energy photons are counted for lung burden assessments.

Even though reproducibility of the chestwall measurement error itself can be maintained at  $\pm 5\%$ , it can propagate very significant errors in lung burden assessments. For the case of  $^{239}\text{Pu}$  assessments where 17 keV (average) photons are counted, an error of 6-7 mm would cause the final lung count to be misinterpreted by as much as 100%. It is therefore necessary to maintain the best possible accuracy and precision in making chestwall tissue measurements for lung deposition assessments.

This paper discusses the major problems of making chestwall tissue measurements and the effect they have on making  $^{239}\text{Pu}$  lung deposition assessments. The instrumentation and techniques used at Mound Laboratory during the last four years are reviewed. A discussion of different methods of deriving the chestwall thickness that have been investigated is also included.

### Calibration

The effect of the chestwall as an absorber when lung counting for  $^{239}\text{Pu}$  must be considered during calibration. Mound Laboratory

\*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Atomic Energy Commission under Contract No. AT-33-1-GEN-53.

calibrates for lung counting using a Remab Hybrid phantom that has known quantities of  $^{239}\text{Pu}$  distributed uniformly throughout the phantom lungs.<sup>1</sup>

For  $^{238}\text{Pu}$ , the most prevalent photons available for counting are the 17 keV (average) uranium L x-rays. The three  $^{234}\text{U}$  L x-rays (13.6, 17.2, and 20.4 keV) are not attenuated by tissue at the same rate and therefore give a transmission curve similar to the beef-steak curve shown in Fig. 1.

Two 12.7 cm diam phoswich detectors were used in a "normal" counting geometry to obtain the data in Fig. 1, and thus the significant geometry effects are included.

When the statistical errors that would normally be considered in calibration procedures are disregarded, the error in relative transmission caused by an error in the chestwall tissue thickness can be readily observed from the dotted lines in Fig. 1. An error of 6 mm results in a transmission error of about 100%. The chest-wall effect on minimum detectable activity ( $3\sigma$  greater than background) is shown in Fig. 2. A typical subject with no exposure and a counting rate of 7.5 counts/min for 4000 sec is used in the example.

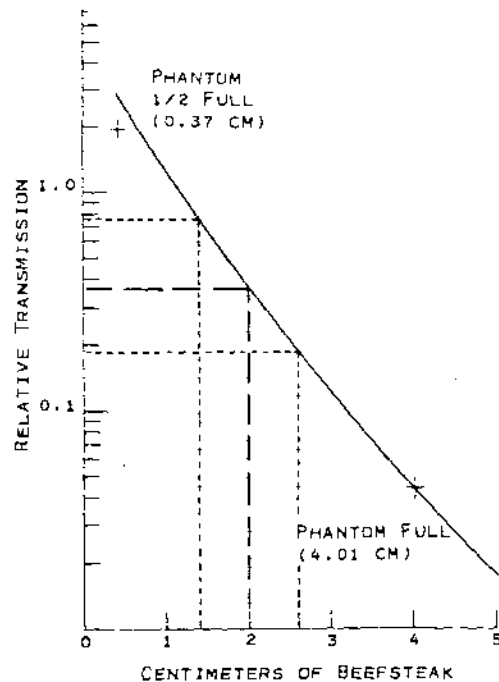


Fig. 1 17 keV (average) x-ray attenuation. [Absorber: beefsteak, ground and frozen. Source: phantom lungs ( $^{238}\text{Pu}$  spiked). Detectors: 2 each 5 in. NaI(Tl)-CsI(Na) Phoswich.]

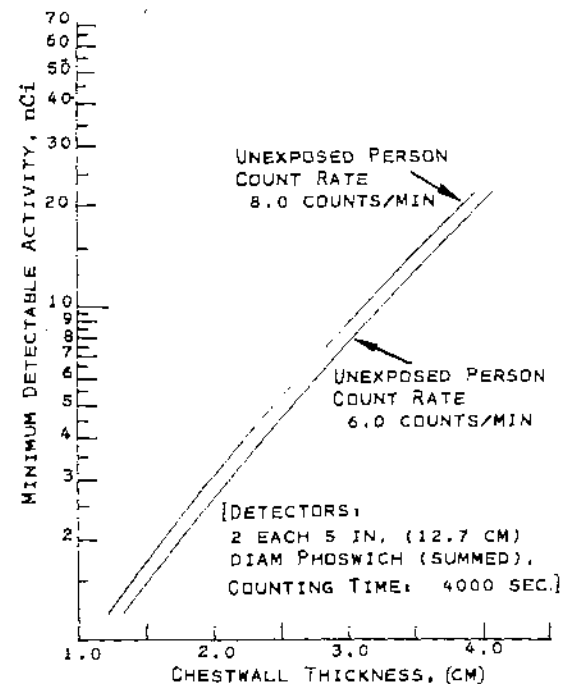


Fig. 2 Minimum detectable activity as a function of chestwall thickness

### Instrumentation

In 1968 the only known investigation into making chestwall tissue measurements had been made by Ramsden et al.<sup>2</sup> Since lung counting of employees was to be routine, it was also desirable to determine the chestwall tissue thickness rapidly and accurately.

An ultrasonic sounding instrument was purchased from Hewlett Packard.<sup>3</sup> Instruments of this type are used in the medical profession for brain and heart studies. The transducer (0.64 cm diam) is electrically pulsed and produces an acoustic frequency of 2.5 MHz which is transmitted through a coupling medium into the subject to be measured. The acoustic pulses are partially reflected at any interface where the acoustic impedance changes. The reflected portion of the pulse is then detected by the transducer and the time delay from transmission is measured. This time difference is displayed on the horizontal axis of the cathode ray tube which is calibrated in centimeters of tissue. The position of any acoustic interface is a linear function of the velocity of sound in that medium and is converted to a depth measurement from the crystal transducer. The intensity of the reflected pulse is a function of the depth and acoustic impedance mismatch and is displayed on the vertical axis of the cathode ray tube. A typical trace on the instrument is shown in Fig. 3.

This instrument has an adjustable distance marker that is adjusted via a 10-turn potentiometer, and once it is aligned with the echo of interest, it reads the distance from the transducer to the interface producing the echo. This mode of operation is commonly referred to as the "A-scan mode." A calibration block of plastic is supplied with the instrument.

#### Methods and Procedures

Ultrasonic chestwall measurements were made on cadavers using various approaches followed by sectioning of the chestwall and physical measurements made with calipers.<sup>4</sup> Rather than measure directly to the lung interface between the ribs, it was advantageous to use the tissue to rib interface since the intensity of the echo was more sharply defined. However, to do this, additional rib thickness must be added to the tissue thickness overlying the ribs. After numerous rib thickness measurements on skeletons, this rib thickness was found to range from 0.3 to 0.5 centimeters<sup>4</sup> depending on general bone structure of the skeleton.

An average thickness of the chestwall was found for the area between the detectors and the lungs. The nine measured points are located as shown in Fig. 4 and on the right side of the chest.

The photon attenuation is a function  $e^{-\mu x}$  where  $\mu$  is about  $1.15 \text{ cm}^{-1}$  for tissue and  $x$  is the absorber thickness in centimeters. Therefore, the average thickness is not a simple mean of several measured thicknesses, but instead, is an "exponential average." The true average thickness was found by measuring nine points to the rib, averaging the values of  $e^{-1.15x}$ , equating this average to  $e^{-1.15x}$ , solving for  $x$ , and then adding the rib thickness.

Asymmetry between the right and left halves of the chestwall thickness could not be defined within the precision of measurement.

A highly developed technique was necessary in order to align the transducer to achieve a well defined structure as shown in Fig. 3. The most obvious difficulties occur on subjects with appreciable amounts of fatty or muscle tissue in the chest area.

A second method of measuring the chestwall thickness between the ribs in the same chest area was also briefly investigated.

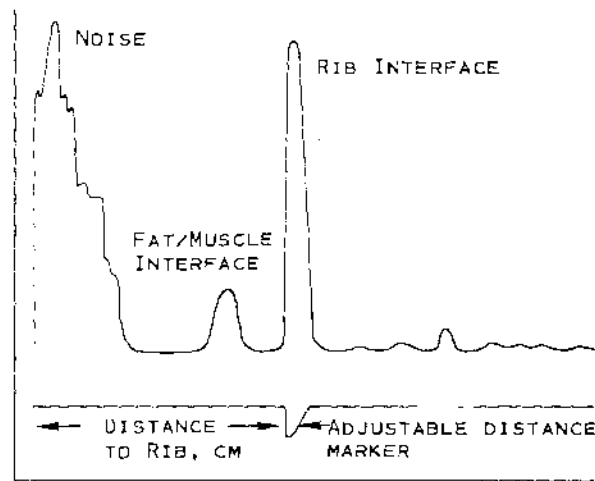


Fig. 3 Ultrasonic display of chest-wall measurement

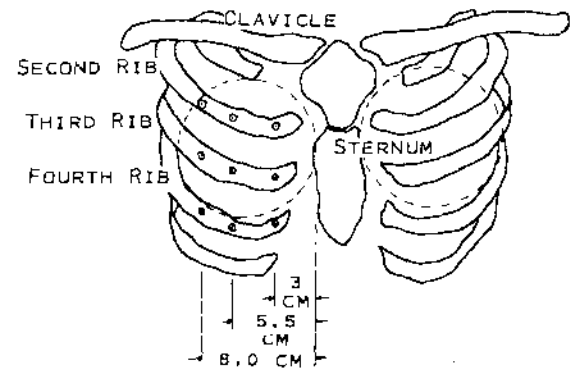


Fig. 4 Ultrasonic measurement points. (Approximate area covered by detectors shown in dotted lines.)

The same basic principles of measurement apply; however, the cathode ray tube display is slightly different.

#### Results and Discussion

After more than 1,000 chestwall determinations had been made, the possibility of discarding the procedure in lieu of a prediction method was investigated.

The average chestwall measurement for 741 different subjects was found to be  $2.28 \pm 0.748$  cm ( $2\sigma$ ). Therefore, in the simplest form, it is possible to use this value for all subjects but with an extremely large resultant error in plutonium lung assessments. The error could be in excess of 100% at the 95% confidence limit if this thickness is related back to Fig. 1.

Another prediction method used various physical body parameters, which can be quickly measured, to predict the chestwall thickness as investigated by Ramsden et al.<sup>3</sup> and Dean.<sup>5</sup> A stepwise multiple regression analysis was used to investigate different prediction equations for chestwall tissue thickness. The data used were limited to 644 different subjects for which all physical parameters were measured.

The stepwise multiple regression used is a statistical technique for analyzing a relationship between a dependent variable (chestwall thickness-T) and a set of independent variables (see Table I) in order of their importance. The criteria of importance is based upon a reduction of the total variation in the dependent variable. In each given step the independent variable most important in this reduction is entered in the regression. Unless the percentage of the total variation accounted for by an independent variable was greater than 1%, the variable was eliminated.

The results of these analyses for three major groups of independent variables are shown in Table I.

TABLE I Regression Analyses

1. Independent Variables: Weight (W), Height (H), Chest Circumference (CC), Waist Circumference (WC), Chest Thickness (CT) and Age (A)

<u>Step No.</u>	<u>Variable</u>	<u>Regression Equation</u>	<u>Total Variation Accounted for by Regression</u>	<u>Multiple Correlation Coefficient</u>	<u>Standard Error of Estimate (cm)</u>
1	W	$T = a+bW$	40.3%	0.634	0.292
2	H	$T = a+bW+cH$	46.1%	0.678	0.270
3	A	$T = a+bW+cH+dA$	51.7%	0.718	0.264

Example of Step 3 Regression Line:  $T = 4.0185 + 0.0107 W - 0.0475 H - 0.0088A$   
 Standard Error of Estimate ( $\sigma$ ) = 0.264 cm

6001

2. Independent Variables: W/H, CC, WC, CT, A

1	W/H	$T = a+b(W/H)$	43.1%	0.657	0.285
2	A	$T = a+b(W/H)+cA$	48.2%	0.694	0.273

Example of Step 3 Regression Line:  $T = 0.8388 + 0.6880 W/H - 0.0082A$   
 Standard Error of Estimate ( $\sigma$ ) = 0.273 cm

3. Independent Variables  $(W/H)^{\frac{1}{2}}$ , CC, WC, CT, A

1	$(W/H)^{\frac{1}{2}}$	$T = a+b(W/H)^{\frac{1}{2}}$	43.8%	0.661	0.284
2	A	$T = a+b(W/H)^{\frac{1}{2}}+cA$	48.3%	0.694	0.273

Example of Step 3 Regression Line:  $T = 0.9690 + 2.2249 (W/H)^{\frac{1}{2}} - 0.0077A$   
 Standard Error of Estimate ( $\sigma$ ) = 0.273 cm

Of the two ultrasonic methods described, the first was the more objective and easier to use. The method of measuring between the ribs to the lung interface is more difficult to use because of the lesser intensity echo from that interface which therefore could introduce significant error in accuracy.

Because of errors involved in predicting the chestwall thickness in the upper chest area, the precision from ultrasonic measurement was investigated on 45 subjects using the first method described previously. Over a one year period, all plutonium operating personnel that were scheduled for lung counting and found to have had two or more chestwall measurements were considered. Only the data from those subjects whose weight and thickness (front to back) had not varied more than 4% were used. The results of applying these constraints left 45 suitable subjects and two standard deviations were found to be 0.112 cm.

One error that is generally not considered in discussion of chestwall thickness measurements is that resulting from the ratio of fat to muscle. Although a small error is introduced from the direct ultrasonic measurement, the propagated error in a plutonium lung burden assessment can be of the order of 10%.

Even though several methods of predicting the chestwall tissue thickness were investigated, the results indicate that the most precise technique of determining this value is by ultrasonic measurement. It is, however, possible to conserve time used for routine counting by making only four to six routine chestwall measurements weekly to maintain the technique needed in actual lung burden assessment cases.

#### Acknowledgements

The authors would like to thank Ernest Arnett, M.D., for his invaluable work with the cadaver measurements and E. L. Saenger, M.D., and Dr. J. Kerieakes for the use of the medical facilities at the University of Cincinnati Medical College as well as their advice on techniques. Grateful thanks are also given to Mr. Anthony Grandillo for performing the regression analyses of the data.

#### References

1. Nuclear Associates, Inc., 35 Urban Avenue, Westbury, New York 11590, USA.
2. D. Ramsden, C. O. Peabody and R. G. Speight, United Kingdom Atomic Energy Authority Report AEEW-R, 14 (1967).
3. Hewlett-Packard Company, 1501 Page Mill Road, Palo Alto, California 94304, USA.
4. E. Arnett, unpublished results.
5. P. N. Dean, Health Physics, 24, 439 (1973).

## In vivo MEASUREMENT OF URANIUM CONTAMINATION IN THE LUNG

E. Riklis & N. Degani

Israel Atomic Energy Commission,  
Nuclear Research Center - Negev, Beer - Sheva, Israel.

**Abstract** - The commonly used method for detection of internal contamination of uranium, as well as of all other radionuclides - is urinalysis. In cases of nontransportable natural uranium contamination by inhalation, experience has shown that urinalysis gives erratic results which do not enable accurate calculation of the inhaled amount. A method for direct in vivo measurement of natural uranium, utilizing the 186 keV line of uranium-235, has been developed utilizing a Whole Body Counter by placing the detector directly over the lung area (1, 2). This procedure requires however corrections due to the presence of caesium-137 and potassium-40 in the body and the parallel measurement of a "double" as a control. A newly developed method gives direct in vivo determination of uranium contamination by counting the soft L X - rays (10 - 20 keV) contribution of uranium-238 and uranium-234. Counting is done inside the low background Whole Body Chamber with two proportional detectors (30 x 15 cm) flushed with argon and methane. Quantitative calibration was done with sponges saturated with  $UO_2$  and placed in a chest cavity of a male goat weighting 75 kg. Counts due to uranium are much higher than those contributed by the normally existing amounts of caesium-137 and potassium-40 thus enabling a detection threshold of at least 1/3 of the maximum permissible lung burden. Work is in progress for improvement of apparatus and methodology.

With the increased availability and use of radioactivity, knowledge of the behaviour in the human body of inhaled or ingested radioactive material has acquired a new significance.

Since most chemical elements which enter the body are eventually excreted, analysis of the urine became the most commonly used method for detection of radioactive internal contamination.

Any activity above zero of radioactivity in the urine is an indication of the presence of a contaminant, and rather elaborate methods of determination of the true amount of this radioactivity have been developed and are in practice in all bioassay laboratories in nuclear centers.

Accurate counting of radioactivity in urine is an easy task, at least for  $\beta$  and  $\gamma$  emitting radioisotopes. Calculating from the result the actual amount of radioactive material which exists in the body is another matter and is rather complicated and inaccurate. The rate of excretion of an element from the body depends on its physical state, chemical formula, particle size, the metabolic activity of the body, and other such factors. These become even more complicated when one deals with insoluble and  $\alpha$  emitting elements.

Our experience in one case of inhalation of insoluble natural uranium dust strengthened the notion that it is difficult to rely solely on urinalysis for quantitative determination for the amount of uranium inhaled and retained in the human body (1, 2). As has been found in this case - the urine of the contaminated subject showed great variations in uranium concentration from day to day (Fig.1), ranging from a high of about 2000  $\mu$ /liter to a low of about 100  $\mu$ /liter.

After analysis and reconstructing the case and concluding that we are dealing with a case of inhalation of insoluble natural uranium, it became clear that we have to develop a direct in vivo determination of the uranium in the lungs. The



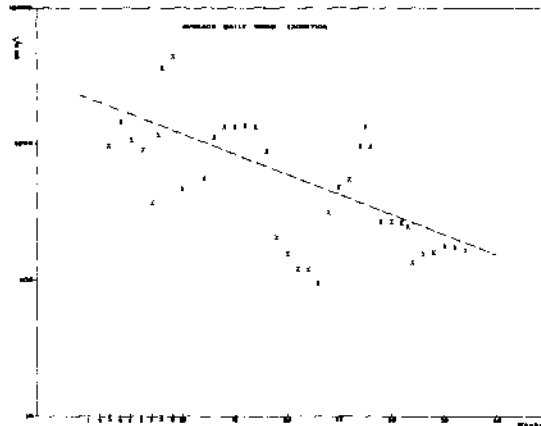


Fig. 1

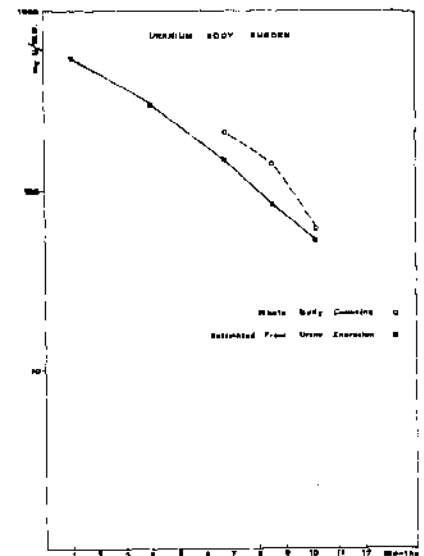


Fig. 2

availability of a Whole Body Counter was therefore useful in developing a direct counting method based on Coffield's method (3) which measures the counts at 186 K and 90 KeV, with the 5x9 inch crystal placed directly over the chest at a distance of 1 cm and with corrections made by counting of an unexposed "double", equal in weight and height to the exposed subject. Details of these calculations are given in Table I. Uranium standard was counted at a distance from the crystal of 22 and

Table I : Body Burden obtained by Whole Body Counting at 186 keV

months after exposure	cpm					$\mu\text{cm}^{-1}$	cpm		mg burd U/U
	U x=22cm	U x=11cm	URo	U*URo	UR1		N(UR1)	NoUR1	
7	68	130	174	179	260	0.13	86	350	207
9	74.5	144.9	180	187.8	245.7	0.113	65.7	266.4	141
11	78.6	149.9	147.3	155.5	175.5	0.113	20.2	114.5	61

U = Uranium-graphite standard containing 77 mg uranium. URo = control subject. UR1 = Subject under study. N(UR1) = counts obtained from subject. No(UR1) = Counts obtained from subject after correction for chest absorption.

Example of calculation:

$$N(\text{UR1}) = \text{No}(\text{UR1}) \cdot e^{-ux} \quad ; \quad \text{No}(\text{UR1}) = \frac{N(\text{UR1})}{e^{-ux}} = \frac{86}{e^{-1.4}} = 350 \text{ cpm} ;$$

$$\text{Body Burden} = \text{mg U/UR1} = \frac{\text{No}(\text{UR1}) \cdot \text{mgU}}{\text{cpm U}} = \frac{350 \times 77}{130} = 203 \text{ mg} ;$$

cm, and under the back of an unexposed person serving as a double. Thus the chest absorption  $\mu$  was determined.

Subtracting the counts obtained in the double (URO) from the counts obtained from subject (URI) gives the counts contributed at 186 KeV from the uranium in the lungs. When correction is made for chest absorption in accordance with the simplified formula  $N_0(\text{UR } 1) = \frac{N(\text{UR } 1)}{e^{-\mu x}}$ , the actual counts of uranium are obtained, and when the amount of uranium retained in the lung is determined.

The results of employing this method enabled us to assess the amount of natural uranium still present in the lungs of the exposed subject, to follow the rate of elimination from the body and (Fig. 2) calculate by extrapolation the amount which was inhaled at the time of exposure.

This method of direct counting of retained uranium in the lungs is certainly more accurate than urinalysis, as it counts retained rather than excreted uranium and is thus not dependant on physiological and other factors which influence the rate of excretion of radionuclides from the body. It is not, however, an easy method as it requires bringing both the subject and his "double" to the whole body counter at frequent intervals, measurements are time consuming and moreover, not all laboratories possess a Whole Body Counter which is rather expensive equipment.

Other methods for accurate direct determination of uranium contamination are therefore under investigation.

External counting of the low intensity uranium L X - rays (energies 13.6, 16.9, and 20.2 keV) seemed to us a promising technique for the estimation of insoluble natural uranium in the human body.

There are several publications on development of instruments and methods for the estimation of plutonium in the lungs by counting the L X-rays of uranium derived from it (4,5,6,7,8), but none about possibility of estimation of uranium as such.

Clearly, there are difficulties due to the fact that these low energy radiations are very easily absorbed in the tissues of the chest wall. The published half value thickness is about 0.7 cm of soft tissue and only 0.03 cm of bone. It seemed however possible that with this technique uranium in the lungs can be counted with a proportional counter without much interference of counts derived from caesium-137 (660 keV) which exists now in every human being.

We have used detector system comprised of two commercially made proportional detectors (30 cm x 15 cm) flushed with argon - methane gas and connected to a suitable single channel analyzer. The detectors were placed inside the low background

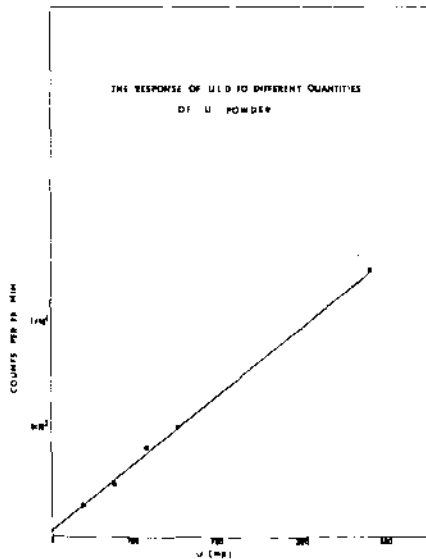


Fig. 3

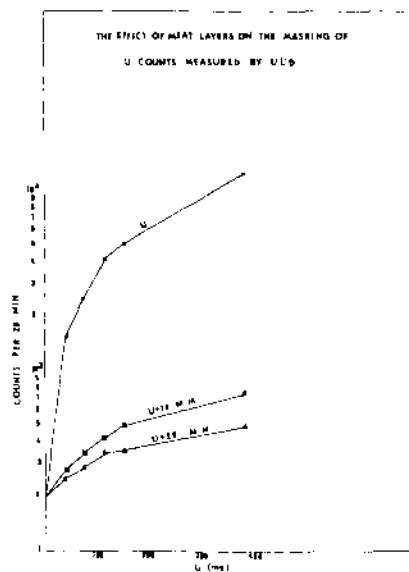


Table II : Counts Obtained in U L D

Isotopes and amount counted		Regions			
		I 3-16	II 2-6	III 10-24	
		Counts per 20 minutes			
$^{137}\text{Cs}$	$8 \times 10^{-9} \text{Ci}$	153	40	38	
	$100 \times 10^{-9} \text{Ci}$	908	152	230	
	$200 \times 10^{-9} \text{Ci}$	1883	290	431	
KCl	245 mg	450	112	148	
$\text{UF}_4$	50 mg	2013	325	1219	
	25 mg	1065	184	753	
	15 mg	697	132	419	
	7.5 mg	400	90	203	
$^{137}\text{Cs}$ + KCl + $\text{UF}_4$					
$8 \times 10^{-9} \text{Ci}$	245 mg	0 mg	571	107	141
"	"	50 mg	2410	423	1437
"	"	25 mg	1645	311	910
"	"	15 mg	1337	412	566
"	"	7.5 mg	885	278	280
Calculated net	50 mg	1839	316	1296	
$\text{UF}_4$ counts	25 mg	1074	204	769	
	15 mg	766	305	425	
	7.5 mg	314	171	139	
Calculated ratios	50 mg/8 nCi	13.1	8.2	32.1	
U/Cs	7.5 mg/8 nCi	2.6	2.2	5.3	
Background		100	18	24	

Work is now in progress with tissue equivalent phantom. It is hoped that when all measurements are repeated in the correct spectral region under controlled conditions, it will give us another method suitable for accurate estimation of the amount of uranium contamination in the human lung.

### Acknowledgements

Mr. E. Roma and Miss F. Gershuni are thanked for operating the WBC and ULD.  
Dr. M. Ronen is thanked for medical examinations.  
Mr. I. Ronen is thanked for performance of modifications in electronic equipment .

### References

1. Riklis E., Ronen M. and Gilbboa A. National Radiology Congress, Jerusalem (1968).
2. Ronen M. and Riklis E., IAEA Annual Report IA 1218, 1202 (1969).
3. Coffield R.E., Health Physics, 2, 269 (1960).
4. Taylor B.T. and Rundo J., Radioactivity in Man, p. 52 IAEA Vienna, (1964).
5. Rundo J., Taylor B.T., Booker D.V., Newton D. and Scargill D., Nature 217, 642 (1968).
6. Bukovitz A.G., Sayeg J.A., Spritzer A.A. and Brodsky A., Health Physics 17, 71 (1969).
7. Taylor B.T., Health Physics 17, 59 (1969).
8. Clemente G.F., These proceedings.

## EXPOSURE FROM NUCLEAR POWER

### "AS LOW AS PRACTICABLE" IN THEORY AND PRACTICE

G. Hoyt Whipple  
University of Michigan  
Ann Arbor, Michigan, U.S.A.

#### Abstract

The ICRP introduced the phrase "as low as practicable" into the language of radiation protection some years ago. Since that time, the phrase has served as a qualitative admonition to prudent judgment. The record of the nuclear power industry, in particular, bears witness to the care with which engineers have applied this concept to the design and operation of nuclear power plants.

Recently the U.S. Atomic Energy Commission has proposed to elevate "as low as practicable" to a regulatory standard with numerical limits. This proposal has the effect of reducing the ICRP limit for public exposure by a factor of 100 and essentially wipes out the opportunity for judgment, sound or otherwise.

Such a reduction in an environmental quality standard is probably unique in the field of public health. An action as dramatic as the proposed reduction is properly taken in circumstances of near-epidemic proportions. However, there is no evidence that radiation exposure at the ICRP limits has any demonstrable effects; there is in fact considerable evidence that it does not. Further, the nuclear industry has conducted its operations in such a way that public exposures are far below the ICRP limits.

The justification offered for setting a legal, numerical limit to "as low as practicable" is that if it can be done, it must be done. This is a philosophy that demands careful scrutiny. The costs in effort and money are considerable; the benefits, if any, are miniscule. Further, it is a philosophy without reason and without stopping place. Finally, it is an action which seriously undermines confidence in the ICRP standards.

#### Introduction

The ICRP introduced the phrase "as low as practicable" into the language of radiation protection in 1958.<sup>1</sup> This phrase, I believe, was intended as an admonition to prudent judgment in the face of the possibility that "any exposure may involve some degree of risk."<sup>2</sup>

The record of radiation exposure, both occupational and public, during the last several decades bears evidence to the care with which designers, engineers and operators have applied this concept. The public exposures which have resulted from the operation of nuclear power plants are particularly striking in this respect. In all but unusual instances, the public exposures from these plants have been less than a few percent of the limits recommended by the ICRP.

One can only believe that those who labored on the ICRP committee have been gratified with the result of their carefully worded recommendation.

U.S. A.E.C. Proposed Appendix I to 10 CFR Part 50

Comes now, however, the United States Atomic Energy Commission.

About two years ago this body issued a draft of a new proposed regulation.<sup>3</sup> The issuance of regulations is, of course, a responsibility of governmental agencies, and the US AEC has met this responsibility fully. The new proposed regulation of concern to us is known as "Appendix I to 10 CFR Part 50." For those of you who are not familiar with this proposed regulation, a terse summary may be useful.

The proposed Appendix I to 10 CFR Part 50 elevates "as low as practicable" to a regulatory standard with numerical limits. These limits are, for all practical purposes, 1/100 of the ICRP public limit; i.e., 5 mrem per year to any organ of the body. There are also, in this proposed regulation, annual release limits for certain isotopes, and some talk of the need for flexibility to allow nuclear power plants to produce electricity now and then. The essence of the regulation is, however, the public organ limit of 5 mrem per year.

The reduction of an environmental quality standard by a factor of 100 is probably unique in the field of public health. What circumstances, one wonders, have called for an action as dramatic as this?

Possible Need for the Proposed Regulation

Is it that the present limits of radiation exposure are producing unacceptably high rates of injury and death?

Is it that the nuclear power industry is abusing the present limits and that the proposed rule is needed to enforce them?

Is it that the margin of safety in the present limits has been found to be smaller than was intended?

Taking each of these possible explanations in turn, we find that (1) there is no evidence that radiation exposure at the present ICRP limits has any demonstrable effects, and considerable evidence that it does not, (2) the nuclear power industry has conducted its operations in such a way that maximum public exposures have seldom exceeded 1/100 of the ICRP limits, and (3) the U.S. National Council of Radiation Protection completed in 1971 an extensive review of the bases for radiation exposure limits and concluded that there is no need to reduce them.<sup>4</sup>

The recent BEIR Report<sup>5</sup> encourages consideration of quantifying the "as low as practicable" concept, but states that "there should not be attempted the reduction of small risks even further at the cost of large sums of money that spent otherwise, would clearly produce greater benefit."

As far as public health is concerned, there is no demonstrated need for the proposed regulation. Why, then, is this dramatic reduction of exposure limits necessary?

### Justification

If one searches through the voluminous material associated with the proposed Appendix I, he finds that the justification given by the AEC is this: since the nuclear industry has shown that it can operate at about 1/100 of the ICRP limits, the industry should be required to operate this way. In other words, if it can be done, it must be done. This is a new concept in public health and deserves thoughtful scrutiny. It is a concept which is beginning to appear in public health areas other than radiation and leads to the ultimate goal where all environmental contaminants are maintained forever at zero, whatever that may be.

The concept that if it can be done, it must be done has no stopping place. For example, suppose that the nuclear industry responds to the proposed Appendix I by designing nuclear plants which operate at 1/10,000 of the ICRP limit; it follows by the concept that they must be operated at this limit. The engineer who designs a safety factor into his plant is rewarded by having it eaten away by the next set of regulations. Such a drive to perfection (if zero is indeed perfection) has certain attractions to some people who do not count the cost, but the cost must be counted and it must be paid.

### The Costs

The additional costs which will be imposed on the electric energy produced in nuclear power stations by the proposed Appendix I may be examined in two ways: (1) in terms of the benefit-cost ratio, or (2) in terms of cost per person-rem avoided. (Women are being liberated by elimination of the unit man-rem from the U.S. vocabulary). Application of the benefit-cost ratio to the proposed Appendix I is a simple matter: the benefit to public health is zero; the cost will be appreciable; the value of the ratio is zero. So much for the benefit-cost analysis.

At the previous meeting of this Association, Hedgran and Lindell gave a charming paper which lead to an estimate of the value of a man-rem as about 1,000 Swedish crowns.<sup>6</sup> Currency fluctuations in the intervening years have introduced more uncertainty in this figure than the uncertainty of their estimate. Estimates of the costs of the person-rem to be saved by the enactment of Appendix I range from less than one hundred "early 1973 dollars" to four million dollars per person-rem.<sup>7</sup> Whatever the value of the dollar in terms of the crown, the Appendix I person-rem appear likely to be expensive.

There is another aspect to these Appendix I person-rem, quite apart from their cost. Hedgran, Lindell and the others who have speculated on the proper expenditure to spare a man-rem were considering actual exposures. Appendix I, on the other hand, is concerned largely with fictitious person-rem received by an imaginary child, drinking imaginary milk produced by an imaginary cow which is grazing on an imaginary pasture at the boundary of the plant site. Thus, real dollars are to be spent to spare make-believe doses. Estimation of the value of a make-believe person-rem may belong in the realm of fables, but surely not in the realm of radiation protection.

### The Critics of Nuclear Power

You will see that our search for some justification of the numerical interpretation of "as low as practicable" has so far been in vain. Perhaps looking at the matter from the point of view of the U.S. Atomic Energy Commission may suggest an answer.

Some years ago the late Andre Cipriani said that one should remind himself that government bureaucrats, in their inner hearts, are not purposely trying to make life impossible for the rest of us. (This, as friends of Dr. Cipriani will realize, is a sanitized version of his original statement).

Critics of nuclear power have predicted an assortment of disasters which make Dante's Inferno look like a summer afternoon in the park should the development of nuclear power continue. These critics have pleaded for reduction of radiation limits by factors of ten or more, reduction of these limits to zero (whatever that may be), a moratorium on nuclear power plants until it can be shown that they are absolutely safe, and the elimination of these plants altogether and forever. Still mindful of Dr. Cipriani's comment, I realize that to some extent a governmental agency should respond to the wishes of the public it serves. Perhaps it is here that we may find the explanation for Appendix I.

Nothing on this aspect of the case appears in the pages of reports, testimony and response to which Appendix I has given rise. There is no way of knowing how near to the truth is the suggestion that the purpose of this proposed regulation is to buy off the critics, but it is the only plausible explanation I have found. If this is in fact the explanation, it is ironic indeed: the critics of nuclear power have been trying for years to discredit the AEC; now, by proposing this rule, the AEC discredits itself.

There are circumstances in which one could forgive a frightened bureaucracy for proposing a regulation as unfortunate as Appendix I. Imagine, if you will, a world where radiation is the only cause of illness and death, where nuclear power is the only source of radiation, where alternate sources of power are in ample supply, and where the production of electricity by means other than nuclear power has no ill effects on public health and the environment. In such a world, Appendix I would make good sense. It is such a world as this that many of the critics of nuclear power envisage, but our world is nothing like this. The proposed regulation makes no sense whatever for today's world.

The public has become somewhat confused and suspicious by the controversy over radiation standards. The proposed Appendix I destroys confidence in the ICRP standards, which are surely the most firmly based and carefully conceived the world has ever known.

#### Summary and Conclusion

In theory, the principle of "as low as practicable" is an appeal to cautious judgment. The very low public doses and the complete lack of any ill effects on public health attest to the excellence of the theory and the scrupulous observation of it by the nuclear power industry.

In practice, as in the proposed Appendix I, the principle of "as low as practicable" becomes a stringent numerical standard, which is unnecessarily low and essentially unmeasurable. The proposal has already done considerable harm. It has increased the costs of designing, constructing and operating nuclear power plants. It has introduced bewildering confusion into the planning for electric power generation. The proposed Appendix I may increase cost and public opposition to the point where fission and even fusion are eliminated as sources of energy. This, in my opinion, would be a tragic development. I urge that the Appendix I interpretation of "as low as practicable" be buried as quickly and as quietly as possible.



### References

1. Recommendations of the International Commission on Radiological Protection (Adopted September 9, 1958), Pergamon Press, 1959, paragraph 45.
2. Recommendations of the International Commission on Radiological Protection (Adopted September 17, 1965), ICRP Publication 9, Pergamon Press, 1966, paragraph 52.
3. Atomic Energy Commission (10 CFR Part 50) Licensing of Production and Utilization Facilities, Light-Water-Cooled Nuclear Power Reactors, Proposed Rule Making. Federal Register Vol. 36, No. 111, ps. 11113-11117, June 9, 1971.
4. Basic Radiation Protection Criteria, Recommendations of the National Council on Radiation Protection and Measurements, NCRP Report No. 39, Washington, D.C., January 15, 1971.
5. The Effects on Populations of Exposure to Low Levels of Ionizing Radiation, Report of the Advisory Committee on the Biological Effects of Ionizing Radiations, Division of Medical Sciences, National Academy of Science, National Research Council, Washington, D.C. November 1972.
6. A. Hedgran and B. Lindell, PQR - A Special Way of Thinking, Health Physics 19: 121, 1970.
7. Final Environmental Statement Concerning Proposed Rule Making Action; Numerical Guides for Design Objectives and Limiting Conditions for Operation to Meet the Criterion "As Low As Practicable" for Radioactive Material in Light-Water-Cooled Nuclear Power Reactor Effluents, WASH-1258, (3 vol.) U.S. Atomic Energy Commission, July 1973.

## РАДИАЦИОННАЯ БЕЗОПАСНОСТЬ НАСЕЛЕНИЯ ПРИ ЭКСПЛУАТАЦИИ АЭС СССР

Н.Г.Гусев, В.А.Беляев, А.С.Зыкова, А.Н.Марей, Г.А.Кузнецова  
(Институт биофизики Министерства здравоохранения СССР),  
М.А.Баранов (Нововоронежская АЭС), Г.А.Гуляков (Гидропроект)

Союз Советских Социалистических Республик, Москва

### Abstract

The report considers some problems of radiation safety of a population due to airborne releases from the atomic power station. The basic national standards for radioactive releases (actual or predicted) from a PWR APS are given. The problem of the possible scope of the accident of APS which may cause an environmental emergency radiation dose and the problem of the population doses are discussed.

В настоящее время (к 1 января 1973 г.) в СССР эксплуатируются АЭС различного типа с общей электрической мощностью 2500 Мвт. В их числе /1-6/:

- четыре блока Нововоронежской АЭС (НВАЭС) мощностью около 1500 Мвт;
- два блока Белоярской АЭС (БАЭС) мощностью 300 Мвт;
- Сибирская АЭС мощностью 600 Мвт;
- Ульяновская АЭС с реактором кипящего типа мощностью 75 Мвт.

В июне 1973 г. состоялся энергопуск Кольской АЭС с серийным реактором ВВЭР-440, а в июле - реактора-размножителя на быстрых нейтронах с натриевым охлаждением БН-350 в г. Шевченко (реактор двухцелевого назначения). В ближайшее время предполагается пуск I блока нового серийного реактора канального типа РБМК-1000 на Ленинградской АЭС.

Планируется к 1975 г. довести мощность АЭС до 6000-8000 Мвт, а к 1980 г. - до 28000-30000 Мвт. Такая широкая программа строительства АЭС выдвигает на первый план проблему радиационной безопасности населения, проживающего в районе их размещения.

В докладе основное внимание будет уделено водо-водяным реакторам корпусного типа (ВВЭР), так как для этого типа реакторов в СССР накоплен наибольший опыт. Следует, однако, отметить, что многие положения рассматриваемых вопросов радиационной безопасности реакторов типа ВВЭР являются общими и для другого типа реакторов.

## 1. Основные регламенты по радиационной безопасности

Действующие в СССР официальные документы, регламентирующие допустимые уровни облучения для персонала АЭС и населения, в основном исходят из рекомендаций Международной комиссии по радиологической защите и Международного агентства по атомной энергии. Они отражены в национальных документах /7-10/, которые в нашей стране являются законодательными.

Так, согласно /7/ предельно допустимые выбросы (ПДВ) при нормальной эксплуатации АЭС и при любых погодных условиях не должны превосходить, кюри/сутки: I-131- 0,1, Sr-89, Sr-90- 0,001, суммы аэрозолей с периодом полураспада больше 1 суток (кроме I-131 и Sr-90-Sr-89)- 0,5 и смеси инертных радиоактивных газов (ИРГ) - 3500.

В настоящее время составляются программы для расчетов ПДВ конкретных АЭС. В целях прогнозирования радиационной обстановки на местности считается целесообразным на АЭС типа ВВЭР производить расчет ПДВ для изотопов H-3, C-14, Ar-41, Mn-54, Mn-56, Co-60, Kr-85, Sr-90,89, Zr-95, Nb-95, Ru-103,106, I-129,131, Xe-133, Cs-137, Ce-141,144, смеси изотопов Kr и Xe, а также I-131- I-135. Уместно заметить, что выброс таких социально значимых изотопов, как H-3, C-14, I-129, Sr-90, Cs-137 и некоторых других, нужно регламентировать не в национальных и даже не в региональных масштабах, а с учетом перспективы развития атомной энергетики (включая заводы по регенерации топлива) во всем мире.

На случай аварийных ситуаций на АЭС в СССР установлены следующие временные регламенты (см. табл. 1).

Таблица 1

Допустимые дозы аварийного облучения для населения D, бэр /9/

Меры защиты	Внешнее облучение	Облучение щитовидной железой	
		Дети	Взрослые
A. Дозы, которые не требуют специальных мер по защите	≤ 25	≤ 75	≤ 150
B. Дозы, требующие временных ограничений: укрытия в помещениях, ограничения потребления молока и других продуктов,	25-75	75-225	150-450
C. Дозы, требующие серьезных мер защиты вплоть до эвакуации	> 75	> 225	> 450

В настоящее время документы /7/ и /9/ пересматриваются.

## 2. Радиационная обстановка на АЭС типа ВВЭР

Многолетний опыт эксплуатации в СССР АЭС всех типов подтверждает, что за весь период их эксплуатации не было зарегистрировано ни единого случая, когда газоаэрозольные отходы достигли бы установленных среднесуточных величин (не говоря уже о годовых).

Рассмотрим в качестве примера одну из крупнейших в СССР Нововоронежскую АЭС (НВАЭС) с суммарной электрической мощностью ~ 1500 Мвт.

На НВАЭС работают 4 блока с реакторами типа ВВЭР: I блок (сентябрь 1964 г.) - 210 Мвт, II блок (декабрь 1969 г.) - 365 Мвт; III блок (декабрь 1971 г.) - 440 Мвт и IV блок (декабрь

1972 г.) - 440 Мвт. Реакторы ВВЭР-440 являются головными в этой серии. Сейчас на площадке НВАЭС проектируется У блок, который будет головным в серии ВВЭР-1000. НВАЭС расположена в густонаселенном районе европейской территории СССР. Именно по этой причине, а также учитывая масштабы и перспективу развития АЭС, на НВАЭС существует значительной по объему информации контроль внешней среды. Служба внешней дозиметрии производит контроль следующих сред: выброса из вентиляционных труб радиоактивных аэрозолей и газов; концентрации аэрозолей в атмосферном воздухе; скорости осаждения радиоактивных веществ на почву; воды и донных отложений реки Дон и окружающих озер; питьевой воды, грунтовых вод вблизи хранилищ твердых и жидких отходов; сбросной воды, охлаждающей конденсаторы турбин; воды с полей фильтрации, куда сбрасываются хозяйственно-фекальные отходы; фауны и флоры рек и озер; почвы, растительности, атмосферных осадков. Кроме того, на специально оборудованной автомашине измеряются уровни  $\beta$ - $\gamma$ -загрязненности почвы, а с помощью интегрирующих дозиметров - годовые дозы гамма-излучения. Контроль радиоактивности охватывает площадь с радиусом 50 км. Кроме суммарной  $\beta$ -активности, анализируются концентрации Co-60, Sr-89, Sr-90, Zr-95, Nb-95, Ru-103, Ru-106, I-131, Ce-141, Ce-144, Cs-137. Исследовательскими институтами производится также измерение H-3 и C-14. Достаточно подробные данные об объеме внешнего дозиметрического контроля, величинах выбросов и радиационной обстановке на местности до 1970 г. приведены в работах /II-12/. Поэтому приведенные ниже данные относятся к 1971, 1972 и первой половине 1973 г., когда работали все блоки (см. табл. 2 и 3).

Таблица 2  
Фактические выбросы аэрозолей на НВАЭС

Годы	1971 г.,	1972 г.,			1973г. (за 5 месяцев)		
	МКЮРИ/ГОД	МКЮРИ/ГОД			МКЮРИ		
Блоки	(I) <sup>х</sup> + II	I	II	III	I	II	III + IV
I-131	16,6	162,8	82,9	1,65	17,0	0,83	7,3
Cs-137	7,9	36,4	3,4	0,16	5,8	0,57	1,8
Sr-90	0,94	12,7	0,64	0,022	2,1	0,14	0,06
Ce-141, 144	5,4	27,5	2,45	0,13	5,1	0,46	0,26
Co-60	6,4	17,5	3,4	0,41	4,9	1,5	-

<sup>х</sup>/ В 1971 г. I блок находился на модернизации.

Таблица 3  
Фактические выбросы ИРГ ( Кг , Хе) на НВАЭС

Годы	1971 г.,	1972 г.,		1973г. (за 6 месяцев),	
	КЮРИ/ГОД	КЮРИ/ГОД		КЮРИ	
Блоки	I + II	I + II	III	I + II	III + IV
Выброс	380	20000	400	16000	750
% от ПДВ	0,03	1,6		2,6	

Из табл. 2 и 3 видно, что газозеролевые выбросы по всем четырем блокам НВАЭС очень низки. Так, выбросы по аэрозолям составляют менее 1% от ПДВ, а от двух серийных блоков ВВЭР-440 они еще меньше. По ИРГ суммарные выбросы в 1971-1973 гг. составляли от 0,03 до 2,6%, а для III и IV блоков - от 0,03 до 0,12% от ПДВ. Необходимо отметить, что основной вклад в выбросы дает I (несе-

рийный) блок. Удельная активность воды теплоносителя III и IV блоков примерно на 2 порядка меньше, чем для I блока, и определяется в основном изотопами наведенной активности F-18, Na-24, Ar-41 и K-42.

В табл. 4 и 5 приведены некоторые данные о концентрациях радиоактивных веществ в атмосферном воздухе и скорости осаждения на почву.

Таблица 4

Концентрации радиоактивных аэрозолей в атмосферном воздухе  $\gamma$ , 10<sup>-17</sup> кюри/л

R, км	1971 г.			1972 г.			1973 г.		
	Sr-90	Cs-137	Ce-141,144	Sr-90	Cs-137	Ce-141,144	Sr-90	Cs-137	Ce-141,144
0,5	0,53	0,89	13,5	0,27	0,57	3,8	0,12	0,30	1,40
2-3	0,43	0,65	9,2	0,22	0,51	2,8	0,06	0,11	2,74
4-5	0,58	0,92	15,4	0,25	0,23	3,5	0,10	0,16	1,52
8	0,40	0,73	11,2	0,17	0,27	2,7	0,056	0,32	1,06
15	0,46	0,72	10,8	0,17	0,25	2,4	0,048	0,10	1,56
50X	0,41	0,63	11,4	0,23	0,32	3,0	0,026	0,44	1,59

X/ г. Воронеж.

Таблица 5

Выпадение радиоактивных веществ из атмосферного воздуха  $\gamma$ , мкюри/км<sup>2</sup>год (в 1973 г. мкюри за I квартал)

R, км	1971 г.			1972 г.			1973 г. (I кв.)		
	Sr-90	Cs-137	Ce-141,144	Sr-90	Cs-137	Ce-141,144	Sr-90	Cs-137	Ce-141,144
1,5-2	1,43	2,28	38,4	0,70	1,13	10,6	0,15	0,21	0,51
4-6	1,32	2,16	26,5	0,54	0,99	7,8	0,078	0,21	0,68
9-12	1,58	2,22	27,6	0,59	1,33	7,6	0,072	0,38	0,45
50X	1,37	2,32	35,5	0,69	0,95	12,3	0,189	0,42	1,17

X/ г. Воронеж.

Анализ табл. 4 и 5 показывает, что как  $\gamma$ , кюри/л, так и F, мкюри/км<sup>2</sup>год практически не изменяются с расстоянием от АЭС, а по абсолютной величине не отличаются от соответствующих величин для изотопов глобального происхождения. Заметим, что короткоживущих изотопов (в том числе I-131) ни в атмосферном воздухе, ни на почве не наблюдалось.

Анализ радиоактивности других сред (воды, рыбы и т. д.) подтверждает, что выбросы НВАЭС совершенно не влияют на радиационную обстановку, и существующие уровни загрязнения внешней среды определяются изотопами глобального происхождения, которые характерны для этого периода для центральных районов СССР.

Экспериментальные данные, приведенные в табл. 4 и 5, а также аналогичные данные за 1965-1970 гг. дают возможность определить среднегодовые скорости осаждения изотопов на почву  $V_g$ . Она оказывается равной  $V_g \approx 1,0$  см/сек.

Интегрирующие  $\gamma$ -дозиметры, расставляемые ежегодно в радиусе до 50 км, не показывают различия внешних полей гамма-излучения от уровней естественного фона. Расчеты показывают, что в 1972 г., когда работали три блока, максимальная доза от внешнего  $\gamma$ -излучения, обусловленная выбросом ИРГ, была меньше 0,2 мбэр/год, а от I-131 на щитовидную железу детей (с учетом пищевой цепочки в мае-октябре) менее 1,5 мбэр/год при среднем естественном фоне

90±4 мбэр/год. Следовательно, расчетные величины дозовых нагрузок на население от АЭС меньше колебаний естественного фона и в тысячи раз меньше ПДД для отдельных лиц из населения.

Анализ газоаerosольных выбросов на НВАЭС за весь период ее эксплуатации, а также анализ выбросов других АЭС СССР дает возможность сделать заключение, что расчетные пределы доз на отдельные лица из населения, обусловленные этими выбросами, не достигали 5 мбэр/год, а усредненные дозы по большим группам не достигали 1 мбэр/год. Жидкие и твердые отходы НВАЭС (которые в данном докладе не рассматриваются) практически не изменяют радиационной обстановки на местности.

### 3. Проектируемые газоаerosольные отходы при нормальной эксплуатации АЭС

При нормальной эксплуатации радиационную обстановку как на АЭС, так и во внешней среде определяет степень герметичности твэлов и оборудования теплоносителя. Вследствие процессов коррозионно-усталостного типа оболочка тепловыделяющих элементов (твэлов) могут иметь микротрещины, а затем и более крупные дефекты. При решении балансных уравнений для расчета активности теплоносителя в проектируемых АЭС допускаются предельные условия, а именно: наличие 1% оболочек твэлов с газовой негерметичностью (микротрещины) и 0,1% более крупных дефектов в оболочках, допускающих прямой контакт топлива с водой теплоносителя /13/. Постоянная скорости утечки летучих изотопов из-под оболочек (через микротрещины) в I контур принимается равной  $1 \cdot 10^{-5}$  сек<sup>-1</sup>. Тогда в теплоносителе реактора ВВЭР-440 (полный объем которого равен 200 м<sup>3</sup>) удельная активность изотопов будет достигать (мкюри/л): по сумме изотопов - 139, по I-131 - 1,9, по сумме изотопов йода - 12,2, по сумме изотопов Кг и Хе - 112. Загрязненность воздуха в помещениях рассчитывается из условия допущения неорганизованных протечек теплоносителя 0,2 т/час. При этом предполагается, что из воды в паровоздушную фазу изотопы переходят в соотношениях: ИРГ - 100%, изотопы йода - 1%. Загрязненный воздух из помещений и технологические сдувки из оборудования поступают на системы подавления активности. Они включают в себя (в различных вариантах): газгольдеры выдержки (для снижения активности технологических сдувок); фильтры ФПП из тонковолокнистой ткани, дающие коэффициент улавливания по aerosолям до 99,9%; фильтры с угольной насадкой для улавливания газообразной фазы йода. В последнее время на некоторых АЭС для задержки Кг и Хе проектируются также установки подавления активности на фильтрах с активированным углем. В табл. 6 в качестве примера приведены расчетные выбросы одной из проектируемых АЭС типа ВВЭР-440. Сравнимые (или несколько меньшие) выбросы принимаются и для других типов проектируемых АЭС. Необходимо отметить, что эти величины получены при наиболее пессимистических предположениях, о которых сказано выше. В действительности, как видно из предыдущего раздела, выбросы на реакторах ВВЭР-440 значительно меньше проектных величин.

При расчете ПДВ I-131 в атмосферу учитывается пищевой путь поступления по цепочке воздух-трава-молоко. По многолетним экспериментальным данным авторами получены следующие соотношения для I-131, которые используются при нормировании выбросов I-131:

$\frac{\text{мкюри/л молока}}{\text{мкюри/м}^3 \text{ воздуха}} = 700;$	$\frac{\text{мкюри/кг травы}}{\text{мкюри/м}^3 \text{ воздуха}} = 5000;$	$\frac{\text{мкюри/л молока}}{\text{мкюри/кг травы}} = 0,12$
---	--	--

Рассеяние газоаerosольных отходов после их очистки производится через трубы высотой 120-150 м. Важным фактором защиты населения

является установление вокруг АЭС санитарно-защитной зоны. В каждом конкретном случае размеры этой зоны определяются органами Министерства здравоохранения СССР.

Таблица 6

Расчетные выбросы изотопов проектируемой АЭС типа ВВЭР-440,  
кюри/сутки

Изотоп	Выброс, кюри/сутки	Изотоп	Выброс, кюри/сутки	Изотоп	Выброс, кюри/сутки
Sr-89	$1,5 \cdot 10^{-5}$	Ba-140	$2,6 \cdot 10^{-4}$	Fe-59	$1,6 \cdot 10^{-5}$
Sr-90	$2,4 \cdot 10^{-6}$	La-140	$1,4 \cdot 10^{-4}$	Co-60	$2,6 \cdot 10^{-4}$
I-131	$2,2 \cdot 10^{-2}$	Na-24	$6,0 \cdot 10^{-4}$	Xe-133	250
$\Sigma I$	$5,6 \cdot 10^{-2}$	Cr-51	$1,2 \cdot 10^{-3}$	$\Sigma ИРГ$	350
Cs-137	$2,2 \cdot 10^{-4}$	Mn-56	$1,3 \cdot 10^{-3}$	$\Sigma аэрозоли$	0,18

4. О масштабе повреждения активной зоны, при котором создаются дозы аварийного облучения на местности

В работах /13, 14/ советскими специалистами высказаны соображения по поводу характера развития крупной аварии на реакторах типа ВВЭР, масштабов возможного аварийного выброса во внешнюю среду и технических средств подавления активности. Подчеркивается, что вероятность возникновения аварии может быть сведена к минимуму за счет высокого качества оборудования, периодического и непрерывного контроля за его состоянием; своевременным срабатыванием систем защиты ядерной установки, предотвращающих возникновение и развитие аварии; разработки системы локализации распространения радиоактивных веществ за пределы АЭС и надежных систем аварийного охлаждения. Наиболее опасной считается крупная невосполнимая течь теплоносителя I контура с последующим нарушением теплового режима активной зоны реактора, разгерметизацией оболочек твэлов и далее частичным оплавлением ядерного топлива. Согласно этой модели предполагается, что развитие крупной аварии на реакторах типа ВВЭР может происходить в три последовательные этапа:

I этап - разрыв I контура и невосполнимая штатными средствами течь теплоносителя в герметичное помещение, где расположено оборудование;

II этап - частичное или полное разрушение оболочек твэлов;

III этап - частичное оплавление ядерного горючего.

Основная тактика обеспечения безопасности по модели работы /14/ сводится к тому, чтобы на I этапе аварии беспрепятственно с помощью взрывных клапанов выбросить в атмосферный воздух паровоздушную смесь теплоносителя и максимально ограничить развитие II и III этапов аварии путем использования специальной системы аварийного охлаждения и конденсации пара и локализации активности в пределах герметичного помещения.

Назовем условно масштабом аварии ( $f_2$ ) долю оплавленного ядерного топлива.

Используя экспериментальные данные работы /13/, ниже рассчитан масштаб аварий, при котором создаются на местности дозы аварийного облучения, приведенные в табл. I для группы А.

Расчет сделан для двух групп критических изотопов: смеси ИРГ (т. е. изотопов Kr и Xe) и смеси изотопов йода. Критическими группами населения выбраны дети в возрасте до I года. Сбозначим  $A_n$  и  $A_c$  активность изотопа в активной зоне и в теплоноси-

теле I контура соответственно, кюри. Тогда полная активность аварийного выброса  $Q$  будет равна

$$Q = A_c f_3 f_4 f_5 f_6 \quad \text{I этап аварии} \quad (1)$$

$$Q = A_n f_1 f_5 f_6 \quad \text{II этап аварии} \quad (2)$$

$$Q = A_n f_1 f_2 f_3 f_6 \quad \text{III этап аварии} \quad (3)$$

Коэффициенты  $f_i$  приведены в табл. 7.

Таблица 7

Коэффициенты для расчета аварийных выбросов

Название коэффициента	ИРГ	Дод- IZI	Этап аварии
$f_1$ - доля активности, выходящей из твэла	$(0,12-2)10^{-2}$ 1,0	$(0,12-2)10^{-2}$ 0,8	II III
$f_2$ - доля оплавленного топлива-масштаб аварии	Подлежит	определению	III
$f_3$ - доля активности, вышедшая из технологического оборудования (с учетом адсорбции на внутренних поверхностях оборудования и трубопроводов)	1,0	0,5-1,0	I
$f_4$ - доля от содержащегося в теплоносителе газа, вышедшего при разрыве I контура	1,0	0,4	I
$f_5$ - доля активности теплоносителя, переходящая в парогазовую форму	1,0	$1 \cdot 10^{-2}$	I
$f_6$ - доля активности, вышедшая из технологического помещения (уменьшение обусловлено адсорбцией на стенах помещений и оборудования, а также радиоактивным распадом)	0,25	0,1	I-III

Поскольку в нашей работе ставится задача определения масштаба аварии, т. е. коэффициента  $f_2$  на III стадии аварии, мы не приводим здесь результатов расчета выбросов для первого этапа аварии. Упомянем лишь, что по расчетам /13/ они не создают чрезвычайно опасных доз на местности. Методически масштаб III этапа определяется следующим образом: вначале рассчитывается аварийный выброс ( $Q$ ), при котором создаются дозы аварийного облучения, приведенные в табл. I. Затем из соотношения (3) определяется соответствующая этому выбросу процентная доля поврежденной активной зоны  $f_2$ , условно названная масштабом аварии, т. е.

$$f_2 = (Q \cdot 100\%) / (A_n f_1 f_3 f_6). \quad (4)$$

Соотношение между индивидуальной дозой  $D$  и кратковременным выбросом  $Q$  имеет следующий вид:

$$D = P \frac{\text{Бэр.м}}{\text{сек.кюри}} C_1 \frac{\text{сек}}{\text{м}^3} Q \text{ кюри}. \quad (5)$$

Здесь  $C_1$  - так называемый метеорологический фактор разбавления



(или фактор "вытяжки"), определяемый известной формулой Пасквилле-Гиффорда, для короткого выброса. Графическая зависимость  $C_1$  от расстояния, высоты выброса, скорости осаждения и категорий погоды по Пасквиллу даны, например, в работах /15, 16/.  $\bar{P}$  - нормализованный множитель, численно равный мощности дозы бэр/сек при концентрации изотопа в воздухе  $\gamma$ , кюри/м<sup>3</sup>.

Для ряда задач более удобно соотношение

$$D \text{ бэр} = \bar{P} \frac{\text{бэр} \cdot \text{м}^3}{\text{сек} \cdot \text{кюри}} \left( \gamma \frac{\text{кюри}}{\text{м}^3} t \text{ сек} \right) \quad (6)$$

Здесь  $t$  - продолжительность экспозиции. Величину  $(\gamma t)$  при постоянной во времени концентрации и  $\int \gamma(t) dt$  при зависящей от времени концентрации называют интегралом концентрации.

Таким образом, из формулы (6) может быть определен интеграл концентрации  $(\gamma t)$  или  $\int \gamma(t) dt$  соответствующий заданной дозе  $D$ . Рассмотрим более подробно значения нормализованного множителя  $\bar{P}$ . Для расчета поля  $\gamma$ -излучения от факела радиоактивных газов ИРГ обычно допускается предположение о полубесконечном размере облака. Как известно, в этом случае

$$\bar{P} = 0,25 E (\text{бэр} \cdot \text{м}^3) / (\text{кюри} \cdot \text{сек}), \quad (7)$$

где  $E$  - энергия  $\gamma$ -квантов, Мэв/распад.

Для этих условий  $\gamma$ -излучение на местности (с учетом многократного рассеяния в воздухе) может быть рассчитано также по формуле:

$$D, \text{ рад} = 2\pi 0,87 (\gamma t) \sum_i \frac{K_{\gamma, i}(E_i)}{\gamma_i(E_i)}, \quad (8)$$

где  $K_{\gamma, i}(E_i)$  - гамма-постоянная  $i$ -ой энергии,  $\frac{(\text{р} \cdot \text{м}^2)}{(\text{сек} \cdot \text{кюри})}$ ;  $\gamma_i(E_i)$  - коэффициент истинного поглощения  $\gamma$ -квантов в атмосферном воздухе, 1/м; 0,87 - переводной множитель от единиц рентген к рад.

В табл. 8 приведены уточненные радиационные характеристики для смеси ИРГ и изотопов йода, относящиеся к реактору на тепловых нейтронах типа ВВЭР-440 с тепловой мощностью 1400 Мвт, продолжительность кампании  $T = 3$  года и выдержка - от 0 до 6 часов.

Для расчета дозы  $\gamma$ -излучения использовались нормализованные множители  $\bar{P}$ , приведенные в формуле (7) или табл. 8. Для расчета дозы на щитовидную железу детей нормализованные множители получены, исходя из стандартов МКРЗ и модели, предложенной в работе /17/. При ингаляционном пути поступления I-131  $\bar{P} = 800$ , I-131 в сопровождении I-132-I-135  $\bar{P} = 1440$  и при поступлении I-131 через молоко  $\bar{P} = 1,5 \cdot 10^5$  (бэр·м<sup>3</sup>)/(кюри·сек). Заметим, что интегралы концентрации  $(\gamma t)$ , соответствующие заданной дозе  $D$ , могут быть получены с помощью формулы (6) из соотношения

$$(\gamma t) (\text{кюри} \cdot \text{сек}) / \text{м}^3 = D \text{ бэр} / \bar{P} (\text{бэр} \cdot \text{м}^3) / (\text{кюри} \cdot \text{сек}) \quad (9)$$

При разовом загрязнении пастбищ используются такие соотношения:  $\bar{P} = 10,7$  (бэр·л молока)/(мккюри·сутки). Для детей до 1 года доза  $D$  на щитовидную железу равна: 11,5 бэр/мккюри I-131, 20,5 бэр/мккюри I-131 в присутствии I-132 - I-135 при ингаляции и 15,5 бэр/мккюри I-131 при поступлении с молоком. Расчет масштаба аварии произведен для дозы аварийного облучения 25 бэр от внешнего  $\gamma$ -излучения и 75 бэр на щитовидную железу детей (т. е. доз, не требующие принятия специальных защитных мероприятий). За основу принят серийный реактор типа ВВЭР-440. Расстояния, где расположены населенные пункты - рецепторы, выбраны равными:

Таблица 8

Радиационные характеристики для смесей ИРТ  
и изотопов йода реактора ВВЭР-440

Величина	Время выдержки, мин						
	0	1	10	30	60	180	360
Полное содержание в активной зоне, $10^6$ кюри							
Кг, Хе	590	490	341	286	256	218	188
I-131 - I-135	380	360	330	320	300	240	200
I-131	34,8	34,8	34,8	34,8	34,8	34,6	34,4
Энергия E, Мэв/распад	0,74	0,57	0,52	0,51	0,47	0,35	0,25
$K_u$ , р.м <sup>2</sup> /сек.кюри, $10^{-5}$	7,90	6,4	5,7	6,1	6,0	4,9	3,8
$P$ , бэр.м <sup>3</sup> /сек.кюри	0,18	0,14	0,13	0,13	0,12	0,081	0,061

$R_1 = 50$  км и  $R_2 = 4$  км. Скорость осаждения на почву  $V_g$  для ИРТ принята равной нулю, а для изотопов йода  $V_g = 1$  см/сек. Категория погоды принята по классификации Пасквилла. За  $f_2 = 100\%$  принято полное оплавление ядерного горючего активной зоны. При этом сделано допущение, что специальных мер по подавлению и локализации активности не сделано. Результаты расчета масштаба аварии приведены в табл. 9.

Таблица 9

Масштаб аварии  $f_2\%$ , при котором может достигнуть доза внешнего облучения на все тело 25 бэр и доза на щитовидную железу у детей 75 бэр,  $h_{эфф} = 30$  м (для реактора ВВЭР-440)

ПДД, источник	Категории погоды по Пасквиллу					
	A	B	C	D	E	F
Расстояние $R_1 = 50$ км						
D = 25 бэр, ИРТ	>100	>100	>100	>100	>100	>100
D = 75 бэр, I-131, вдыхание	>100	>100	>100	>100	23	62
D = 75 бэр, I-131-I-135, вдыхание	>100	>100	>100	78	13	35
D = 75 бэр, I-131, молоко	>100	5,6	1,8	0,7	0,12	0,32
Расстояние $R_2 = 4$ км						
D = 25 бэр, ИРТ	>100	>100	>100	>100	32	14
D = 75 бэр, I-131, вдыхание	65	5,6	3,7	1,5	0,34	0,15
D = 75 бэр, I-131-I-135, вдыхание	36	3,2	2,1	0,81	0,19	0,08
D = 75 бэр, I-131, молоко	0,35	0,03	0,02	0,008	0,0019	0,0008

Из табл. 9 видно, что для населенного пункта, находящегося на  $R_1 = 50$  км, допустимые дозы аварийного облучения 25 бэр внешнего  $\gamma$ -излучения и 75 бэр на щитовидную железу детей при вдыхании изотопов йода или совсем не достигаются даже при стопроцентном оплавлении активной зоны, или достигаются при очень большом (и маловероятном) повреждении активной зоны. Исключение составляет пищевой путь поступления йода: если загрязняются пастбища, то достаточно сравнительно небольшого масштаба аварии ( $f_2 = 0,12-1,8\%$  для категорий C - F), чтобы появилась необходимость принятия решения - прежде всего ограничения потребления молока.

На расстоянии 4 км радиационная обстановка ухудшается и при меньших масштабах аварии. Тем не менее поле  $\gamma$ -излучения от облака ИРТ (кроме двух последних категорий E и F) все же оказывается сравнительно небольшим даже при крупных авариях.

Однако опасность облучения изотопами йода может быть значительной даже при небольших масштабах аварии, когда оплавляются лишь доли процента ядерного горючего. Естественно, что с помощью рассмотренного метода может быть решена и другая задача - расчет дозы на население при заданном масштабе аварии.

Подобные расчеты необходимы для разработки инженерных мероприятий по подавлению активности при авариях.

Заметим, что изложенная методика определения масштаба аварии ( $f_2$ ) с помощью соотношения (4) не зависит от выбранной модели подавления активности. (Здесь мы использовали одну из возможных моделей /14/.) Коэффициенты "естественной фильтрации" активности  $f_1$  (оболочками твэлов),  $f_2$  (стенками трубопроводов и оборудования) и  $f_6$  (стенами помещений АЭС) характерны для любого типа аварии.

### 5. О методике расчета популяционной дозы

В настоящее время при оценке степени риска для населения от применения атомной энергии рассчитывают популяционную дозу. При этом обычно используется единица размерности "человек.бэр". Если  $N_j$  - число лиц, получивших дозу от определенного вида радиации  $D_j$  бэр, то популяционная доза может быть определена из соотношения

$$D_p = \sum_j N_j D_j. \quad (10)$$

Расчет популяционной дозы по формуле (10) может быть сравнительно легко осуществим от непосредственных прямых путей воздействия радиации на человека, например: на профессиональных работников атомных производств, на население от внешнего потока  $\gamma$ -квантов радиоактивного облака или осадков на почве; от вдыхания радиоактивных аэрозолей и т. д.

Но применение формулы (10) становится затруднительным, а иногда даже невозможным, когда нужно определить вклад в популяционную дозу от загрязненных продуктов питания, которые проходят длительный путь от места их производства (и радиоактивного загрязнения) до потребителя. Это в первую очередь относится к таким долгоживущим изотопам как  $H-3$ ,  $C-14$ ,  $Str-89, 90$ ,  $I-129$ ,  $Cs-137$  и ряду других. При определенных условиях это относится и к  $I-131$ .

Хотя степень загрязнения пищевых продуктов вблизи атомного объекта может быть известна, но, в какой местности, кем и когда они потребляются, часто является неопределенным. Для такого пути воздействия нами предлагается упрощенная схема, которая, однако, требует дискуссионного обсуждения.

Определим вклад в популяционную дозу от определенного изотопа в форме

$$D_p = \sum_j D \left( \frac{\text{бэр}}{\text{кюри}} \right) N_j (\text{чел}) \cdot I_{m,j} \left( \frac{\text{кюри}}{\text{чел}} \right) = D I_m. \quad (11)$$

Здесь  $D$ , бэр/кюри, - нормированная доза при поступлении в организм единичной активности  $I_m = \sum_j N_j I_{m,j}$  - кюри, суммарное содержание изотопа в продуктах питания в момент заглатывания пищи,  $I_{m,j}$  - поступление в организм данного изотопа с пищей количеству лиц  $N_j$  (индекс  $j$  относится к уровню загрязненности пищи). Дозовый коэффициент  $D$  зависит от возраста человека, пути поступления в организм и радиобиологических констант и с помощью стандартов МКРЗ может быть рассчитан. Обратим внимание прежде всего на то, что в формуле (11) популяционная доза выражается в единицах "бэр", а не "человек.бэр". Собственно эта размерность для  $D_p$  вытекает и из формулы (10), если в ней доза на индивидуум  $D$  выражается в "бэр/человек". Рассмотрим теперь более подробно множитель  $I_m$ .

Если исходить из концепции беспороговости действия радиации,

то при расчете популяционной дозы  $D_p$ , а следовательно, степени риска, нет необходимости знать, как и где распределились загрязненные продукты в данной популяции. Достаточно знать, сколько радиоактивного вещества находилось в данных продуктах питания в момент их потребления населением. В простейшем случае (если пренебречь радиоактивным распадом, изменением концентрации при переработке и т. д.) суммарное поступление  $I_m$  в организм людей с продуктами питания можно заменить суммарным содержанием активности данного изотопа в продуктах питания  $A_m$ , т. е.

$$I_m, \text{ кюри в организм людей} \approx A_m \text{ кюри в продуктах питания} \quad (12)$$

Суммарную величину активности в продуктах питания, употребляемых человеком или животными (например, коровами) определить значительно легче, чем индивидуальную дозу на отдельное лицо. Эту характеристику можно получить путем непосредственных измерений уровней загрязнения продуктов питания или путем расчета, применяя, например, методику Пасквилла-Гиффорда /18-20/ с заданной скоростью осаждения на почву. В последнем случае для оценки популяционной дозы требуется знание нормализованной дозы  $\bar{d}$ , бэр/кюри, и выброса в вентиляционную трубу  $Q$ , кюри.

Формула (5) дает возможность рассчитать дозу от выбросов АЭС на отдельное лицо из населения, т. е. индивидуальную дозу, и входящий в эту формулу множитель  $C_i$  представляет собой "индивидуальный" коэффициент разбавления. Если пользоваться моделью Гиффорда, то при разовом выбросе  $C_i$  определяется в виде

$$C_i = \frac{F(x)}{\sigma_y \sigma_z u} \exp \left\{ -\frac{1}{2} \left( \frac{y^2}{\sigma_y^2} + \frac{z^2}{\sigma_z^2} \right) \right\}, \quad (13)$$

где  $F(x)$  - безразмерная функция, учитывающая истощение радиоактивного факела за счет радиоактивного распада и осаждения изотопов на почву. Остальные величины общеизвестны /19, 20/.

Расчет популяционной дозы может быть сделан по формуле, аналогичной (5), с той лишь разницей, что в нее входит популяционный фактор разбавления  $C_p$ :

$$C_p = \int \rho C_i dS = \bar{\rho} \int C_i dS = \bar{\rho} \bar{C}_p, \quad (14)$$

где  $\rho$  - распределение по площади населения, животных или урожайности культур на  $1 \text{ м}^2$ , а  $\bar{C}_p$  - нормированный популяционный фактор разбавления, сек/м.

Поскольку угол расширения струи выбросов обычно меньше  $90^\circ$ , то удовлетворительным приближением можно сделать замену  $y = \theta x$  и  $\sigma_y = \sigma_\theta x$ , где  $\theta$  - азимутальный угол между направлением на ре-цептор и осью струи,  $\sigma_\theta$  - его дисперсия. Подставляя в (14) значение  $C_i$  из формулы (13) и производя интегрирование по окружности, получим

$$\bar{C}_p = \sqrt{\frac{2}{\pi}} \frac{F(x)}{u} \int_0^x \frac{1}{\sigma_z} \exp \left( -\frac{z^2}{\sigma_z^2} \right) d\theta. \quad (15)$$

Тогда по аналогии с формулой (5) при прямом пути воздействия радиации на человека популяционная доза  $D_p$  будет равна

$$D_p = \bar{P} \left( \frac{\text{бэр}}{\text{человек}} \right) \left( \frac{\text{м}^3}{\text{кюри сек}} \right) \bar{\rho} \left( \frac{\text{человек}}{\text{м}^2} \right) \bar{C}_p \left( \frac{\text{сек}}{\text{м}} \right) Q \text{ (кюри)}. \quad (16)$$

При учете более сложного пути воздействия, когда загрязненные ра-

диактивными веществами продукты питания поступают в торгово-заготовительную сеть, по аналогии с формулой (II) имеем

$$D_p = \bar{D} \left( \frac{\text{бэр}}{\text{кюри}} \right) I_{\text{ш}} (\text{кюри}) = \bar{D} \left( \frac{\text{бэр}}{\text{кюри}} \right) k \left( \frac{\text{м}}{\text{сек}} \right) \bar{C}_p \left( \frac{\text{сек}}{\text{м}} \right) Q (\text{кюри}). \quad (I7)$$

В формулах (I6, I7)  $\bar{C}_p$  определяется выражением (I5). Коэффициент  $k$  зависит от типа пищевой цепочки для данного изотопа. Например, при поступлении изотопа в организм через цепочку воздух-пастбище-молоко-человек коэффициент  $k$  будет равен

$$k = k_1 V_E \left( \frac{\text{м}}{\text{сек}} \right) \bar{\rho} \left( \frac{\text{коров}}{\text{м}^2} \right) \zeta S \frac{\text{м}^2}{\text{корова.сутки}} \frac{T_{\text{эфф}} \text{сутки}}{0,693} \quad (I8)$$

Здесь  $k_1$  - доля от поедаемого коровой с кормом изотопа, перешедшая в молоко;  $V_E$  - скорость оседания изотопа на почву;  $\bar{\rho}$  - средняя плотность коров на загрязненной территории;  $\zeta$  - доля изотопа, задерживаемая на траве;  $S$  - площадь, с которой корова поедает траву в течение одних суток;  $T_{\text{эфф}}$  - эффективный период полувыведения изотопа из съедобной части травы.

Для других пищевых цепочек, например, воздух-зерновые культуры-человек коэффициент будет равен

$$k = k_2 \left( \frac{\text{кюри/кг}}{\text{кюри/м}^2} \right) V_E \frac{\text{м}}{\text{сек}} \bar{\rho} \frac{\text{кг}}{\text{м}^2} \quad (I9)$$

Здесь  $k_2$  - активность изотопа в I кг зерна, полученного с территории, загрязненной интегральным выпадением I кюри/м<sup>2</sup>;  $\bar{\rho}$  - урожайность, кг/м<sup>2</sup>,  $V_E$  - скорость оседания, м/сек.

Фактор  $\bar{C}_p$  - весьма удобная для оценочных расчетов величина, зависящая только от условий истечения и разбавления примеси. Как видно из формулы (I5), он не зависит от горизонтальной дисперсии  $\sigma_y$ . Рост вертикальной дисперсии ( $\sigma_z$ ) с расстоянием ограничен толщиной слоя перемешивания. Поэтому единственным фактором, ограничивающим величину  $\bar{C}_p$ , является истощение струи за счет радиоактивного распада во время движения и выпадения на почву. Для долгоживущих изотопов расстояние, для которого необходимо рассчитывать  $\bar{C}_p$ , достигает сотен и даже тысяч километров. Время движения выбросов со средним ветром на столь большие расстояния превосходит сутки. Поэтому при расчете  $\bar{C}_p$  существенным оказывается суточный ход категории стабильности. Фактор  $\bar{C}_p$  является численной величиной, не зависящей от расстояния в отличие от индивидуального фактора  $C_p$ , который является функцией расстояния.

В заключение отметим, что термин "бэр", в свете вышесказанного, является более правильным критерием оценки популяционной дозы, чем широко используемый термин "человек.бэр".

#### Литература

1. А. М. Петросьянц. От научного поиска к атомной промышленности. Изд. 2. М., Атомиздат, 1972.
2. А. М. Петросьянц и др. Атомная энергия, 31, вып.4, 315 (1971).
3. В. П. Денисов и др. Атомная энергия, 31, вып.4, 323 (1971).
4. А. М. Петросьянц и др. Атомная энергия, 31, вып.4, 333 (1971).
5. А. И. Лейпунский и др. Атомная энергия, 31, вып.4, 344 (1971).
6. Ф. Я. Овчинников и др. Эксплуатация реакторных установок Нововоронежской АЭС. М., Атомиздат, 1972.
7. Санитарные правила проектирования и эксплуатации АЭС, СП-38/3-68. М., 1968.
8. Нормы радиационной безопасности, НРБ-69. М., Атомиздат, 1972.
9. Временные методические указания для разработки мероприятий по защите населения в случае аварии ядерных реакторов, № 872/1-70. М., 1970.

10. Основные санитарные правила работы с радиоактивными веществами и другими источниками ионизирующих излучений. М., Атомиздат, 1973.
11. Ф. Я. Овчинников и др. Объем дозиметрического контроля в районе расположения Нововоронежской АЭС. Доклад на симпозиуме по вопросам безопасности АЭС. Болгария, сентябрь 1971 г.
12. Ф. Я. Овчинников и др. Газоаэрозольные отходы НВАЭС и методы их определения (см. библиографию /II/).
13. Л. М. Лузенов и др. Безопасность атомных электростанций с точки зрения последствий крупных аварий. Советско-шведский симпозиум по безопасности АЭС. Швеция, Стутсвик, март, 1973 г.
14. А. М. Букринский и др. Система обеспечения безопасности при авариях с разуплотнением первого контура (см. библиографию /I3/).
15. A. J. Brook. The Effect of deposition on the concentration of winborne material. UKAEA, AHSB(S)R 157, Risley, 1968.
16. Pamela B. Bryant. Methods of estimation of the dispersion of windborne material and data to assist in their application. UKAEA, AHSB(RP)R 42, Harwell, 1964.
17. Pamela M. Bryant. Health Physics, 17, No 1, 51 (1969).
18. F. Pasquill. The Meteorology Magazine, 90, 33 (1961).
19. F. A. Gifford. J. Appl. Meteorology, 6, 644 (1967).
20. Application of meteorology to safety at nuclear plants, IAEA, Safety series, No 29, Vienna, 1968.

AVERAGE EFFLUENT RELEASES FROM U.S. NUCLEAR POWER REACTORS,  
 COMPARED WITH THOSE FROM FOSSIL-FUELED PLANTS,  
 IN TERMS OF CURRENTLY APPLICABLE ENVIRONMENTAL STANDARDS\*

Andrew P. Hull  
 Health Physics and Safety Division  
 Brookhaven National Laboratory  
 Upton, New York 11973

Abstract<sup>†</sup>

Between 1967 and 1972, eighteen "second generation" light-water-cooled nuclear power plants, with capacities in the range of 500-800 MW(e) have been put into operation in the United States. These were in addition to ten smaller demonstration plants and one high-temperature gas-cooled nuclear power plant in operation at the start of this period. The reported yearly air effluent releases of radioactive gases, halogens and particulates, and liquid effluent fission and activation products and of tritium from these plants are evaluated on a Ci/10<sup>3</sup> MW(e) basis, and the overall yearly averages for the various types of reactors [boiling water (BWR), pressurized water (PWR) and high temperature gas-cooled (HTGR)] are compared.

The complete first and second generation data are used to project the effluent releases for a "reference" 1,000 MW(e) BWR, PWR and HTGR. The yearly effluent releases for such reference reactors, at an 80% availability, would be:

	<u>AIRBORNE</u>		<u>LIQUID</u>	
	<u>Gaseous</u>	<u>Halogens &amp; Particulates</u>	<u>Fission &amp; Activation Products</u>	<u>Tritium</u>
BWR	1.66x10 <sup>6</sup> Ci	5.31 Ci	49.6 Ci	104 Ci
PWR	9,650 Ci	0.17 Ci	30.2 Ci	5,750 Ci
HTGR	2,760 Ci	< 0.02 Ci	0.27 Ci	835 Ci

These and the amounts of effluents released from reference 1,000 MW(e) fossil-fueled plants are compared in terms of relative environmental concentrations and their relationship to the applicable U.S. environmental standards for the principal constituents in their respective plant air-effluent streams. The largest required annual dilution volume for the most restrictive constituent is as follows:

<u>Plant Type</u>	<u>Pollutant</u>	<u>EPA &amp; AEC Standard</u>	<u>Annual Discharge Quantity**</u>	<u>Air Dilution Volume (m<sup>3</sup>)</u>
Coal (3.5% S)	SO <sub>2</sub>	0.03 ppm	3.66x10 <sup>8</sup> lb.	2.14x10 <sup>15</sup>
Oil (1.5% S)	SO <sub>2</sub>	0.03 ppm	1.23x10 <sup>8</sup> lb.	6.95x10 <sup>14</sup>
Gas	NO <sub>2</sub>	0.05 ppm	2.71x10 <sup>7</sup> lb.	1.23x10 <sup>14</sup>
BWR	Short-lived radio-gases	3x10 <sup>-8</sup> Ci/m <sup>3</sup>	1.66x10 <sup>6</sup> Ci	5.54x10 <sup>13</sup>
PWR	<sup>85</sup> Kr & <sup>133</sup> Xe	3x10 <sup>-7</sup> Ci/m <sup>3</sup>	9,650 Ci	3.32x10 <sup>10</sup>
HTGR	Short-lived radio-gases	3x10 <sup>-8</sup> Ci/m <sup>3</sup>	2,760 Ci	9.22x10 <sup>10</sup>

\*\*Based on 80% availability

\* Research carried out at Brookhaven National Laboratory under contract with the U. S. Atomic Energy Commission.

† Revised August, 1973 to incorporate 1972 data.

## Introduction

During the late 1960's a widespread concern for the overall quality of the environment evolved in the United States. This led to the enactment of a variety of legislative and administrative measures intended to decrease the quantities of pollutants released to the air and to waters.

This concern also extended to the radioactivity released to the environment in nuclear power plant effluent streams. It was heightened by the widespread dissemination by the media of claims by Sternglass<sup>1</sup> of a causal connection between power reactor effluents and infant mortality, as well as on the arguments made by Gofman and Tamplin<sup>2</sup> in the context of power reactors, that a U.S. population exposure of 170 millirems per year would lead to large increases in prevailing U.S. cancer mortality rates.

In what seemed to at least some observers<sup>3</sup> a response to the popular concern so evoked, rather than to the scientific evidence, the U. S. Atomic Energy Commission (AEC) formally adopted "as low as practicable" into its regulations governing radioactive effluents, and proposed related numerical guidance for light-water-cooled nuclear power reactors.

Up to recently, much of this concern about power reactor effluents has been narrowly focused on their absolute amounts, and on the degree of risk occasioned thereby. Among the first reviews of past experience to establish patterns and trends of effluent releases was one by Blomeke and Harrington<sup>4</sup> covering the years up to 1967, and one covering the same period by Brinck and Kahn<sup>5</sup>. Starting in 1969, the Division of Compliance of the AEC (now the Division of Regulatory Operations) published yearly summaries of power reactor effluents, itemized by the categories of noble and activation gases, halogens and particulates (with a half-life greater than 8 days), mixed fission and corrosion products (MFP) and tritium in liquid effluents. It was qualitatively apparent from these data that the overall amounts of power reactor effluents were increasing; that the gaseous and halogen releases from the boiling water reactor (BWR) type were larger than those from the pressurized water reactor (PWR) type; that their fission and corrosion product releases were rather comparable, and that tritium in liquid effluents from PWR's exceeded that from BWR's.

Since it is related to the meeting of projected electrical energy demand, a reasonable debate about the risks occasioned by nuclear power plant effluents should also include a comparative consideration of that occasioned by conventional fossil-fueled plants. One of the first such comparisons was made in 1967 by Terrill et al.<sup>6</sup>. They set forth discharge quantities per year - MW(e) of SO<sub>2</sub>, NO<sub>2</sub>, radioactive <sup>226</sup>Ra and <sup>228</sup>Ra in fly ash from coal, oil and gas-fueled plants, and also of radioactive noble gases and <sup>131</sup>I from nuclear plants. Using these data with AEC concentration standards for radioactivity or recommended concentration standards for conventional agents, they calculated a yearly volume of air required for dilution [m<sup>3</sup>/MW(e)]. They utilized only a limited amount of PWR effluent release data available to them at that time, as well as concentration standards for conventional pollutants that have since been superseded by much lower ones promulgated<sup>7</sup> by the U. S. Environmental Protection Agency (EPA). However, they showed that the amounts of air required for dilution of the yearly amounts of radioactivity, as well as the conventional pollutants emitted from fossil-fueled power plants, were much greater than those required to dilute the radioactivity emitted from PWR's.

That these conclusions might not apply to the same degree to BWR effluents was suggested in 1969 by Fish<sup>8</sup>, who also suggested that the air quality criteria for SO<sub>2</sub> utilized by Terrill et al. were higher than more recently



adopted ones. In a further consideration along these same lines, Hull<sup>9</sup> utilized the average of all releases from nuclear power plants for 1969, as well as more conservative air quality criteria for non-radioactive agents. His conclusions were in essential agreement with the earlier comparisons, but also showed that both coal and oil plants required larger dilution volumes than BWR's. A similar, but somewhat more sophisticated comparison, which considered the residence time of airborne pollutants, was made in 1970 for coal and nuclear-fueled plants in West Germany by Jansen et al.<sup>10</sup>, who arrived at a similar conclusion.

In the absence of a much needed biological effect related unit for conventional pollutants analogous to the man-rem, these comparisons to concentration standards appear to offer the most objective basis available for weighing the relative risks of the presently available choices for producing electrical power. However, in a recent comparison along this same line, Starr et al.<sup>11</sup> observed that the air quality standards for conventional pollutants are much closer to concentrations at which prompt medical effects are perceivable, than are those for radioactivity. In the absence of well controlled studies of the possible effects of long-term exposures to low levels of conventional pollutants, if a linear dose-effect relationship is applied their effects would be greater than those suggested from considerations based on air quality standards per se.

In what follows, the previous comparisons<sup>9</sup> are updated to include a consideration of how the trends in effluent release rates have been affected by the larger "second generation" light-water-cooled nuclear power plants, with capacities in the range of 500-800 MW(e), some eighteen of which have been put into operation from 1967 on, in addition to those operating at that time. It also incorporates recently adopted EPA Air Quality Standards as the basis for comparison.

#### Reactor Effluent Releases

In order to provide as consistent as possible a basis for evaluating the trend of releases from nuclear power reactors, it is desirable that they be separated by type, and that these releases be normalized to the integrated quantity of electricity generated, rather than on plant capacity. Since the reactor effluent release data summaries by the AEC have not until 1971 included the latter data, it is not clear that previous comparisons have been made on this basis. For 1967 and 1968 the AEC reactor effluent data utilized herein was published in 1969 by the Joint Committee on Atomic Energy<sup>12</sup>. For the years previous to 1971, the amounts of electricity generated by nuclear reactors was obtained from a 1971 AEC report<sup>13</sup> on the operating history of U.S. nuclear power reactors. Since it was available, the data for one relatively small high temperature gas-cooled reactor was also included.

The effluent release data reported by the AEC have been set forth in their four major categories, as indicated above. Starting in 1972, supplementary information on individual nuclides within these categories was also included in the AEC report. Although such data would permit a more precise evaluation of the radiological significance of reactor effluent releases, since it has not been available over the period of interest, it has not been included in this study.

Yearly overall average amounts of gaseous, halogen and particulate, liquid fission and corrosion products, and tritium activity from 1967 to 1972 have been calculated in curies per 10<sup>3</sup> megawatt hours(e). These have been obtained by dividing the total of each reported reactor effluent category by the total electrical power generated by the various reactor types.

The nuclear power reactors operated between 1967 and 1972, with their net electrical power capacities, are shown in Table I. Although omitted from the detailed presentations, the releases from the two reactors which ceased operation during the study period have been included. Shippingport was not included since release data from it was not included in the AEC reports.

Table I  
U.S. Nuclear Power Reactors Operated Between 1967 and 1972<sup>(14)</sup>

<u>Name</u>	<u>Type</u>	Power	<u>Start Up</u>	<u>Shut Down</u>
		<u>Net MW(c)</u>		
Shippingport	PWR	90.0	1957	-
Dresden I	BWR	200	1959	-
Yankee	PWR	175	1960	-
Big Rock	BWR	70.3	1962	-
Elk River	BWR	22.0	1962	1968
Indian Point I	PWR	265	1962	-
Saxton	PWR	3.0	1962	1972
Humboldt	BWR	68.5	1963	-
Peach Bottom	HTGR	40.0	1966	-
San Onofre	PWR	430	1967	-
La Crosse	BWR	53.2	1967	-
Connecticut Yankee	PWR	575	1967	-
Oyster Creek	BWR	640	1969	-
Nine Mile	BWR	625	1969	-
R. E. Ginna	PWR	420	1969	-
Dresden II	BWR	800	1970	-
Millstone I	BWR	652	1970	-
H. B. Robinson	PWR	700	1970	-
Monticello	BWR	545	1970	-
Point Beach	PWR	497	1970	-
Palisades	PWR	700	1971	-
Dresden III	BWR	800	1971	-
Quad Cities I	BWR	800	1971	-
Vermont Yankee	BWR	514	1972	-
Quad Cities II	BWR	800	1972	-
Point Beach	PWR	497	1972	-
Surry I	PWR	788	1972	-

The trend of the gaseous effluent release rate is shown in Figure 1. It is apparent that the average release rate from BWR's has continued to exceed that of PWR's, but that factor has decreased from about 1,000 to closer to 100 between 1967 and 1972. An examination of the individual data, shown in Figures 1A for BWR's and 1B for PWR's, discloses that this is attributable both to the lower "second generation" BWR release rates, which have averaged about 1/10 of the pre-1967 BWR's, and to an increasing trend with time from several of the older PWR's.

A similar pattern for halogens and particulates with a half-life greater than 8 days is evident from Figure 2, except that the average release rates from BWR's have also increased over the period from 1967 to 1972. The release data for individual reactors, shown in Figures 2A and 2B, indicate that although the spread is not large, this is principally due to increases over this period from the older BWR's and that most of the "second generation" BWR release rates have been somewhat less than the overall average. The pattern for individual PWR's is less obvious, but most of the increase in the overall average is again attributable to older plants.

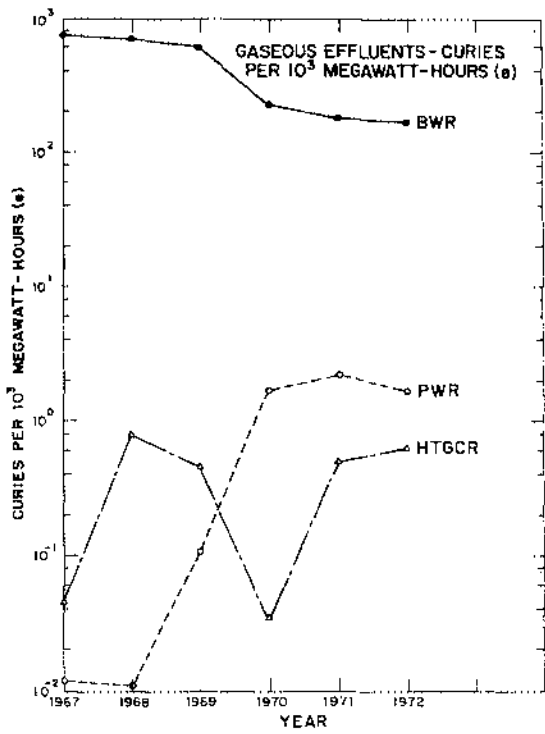


Fig. 1

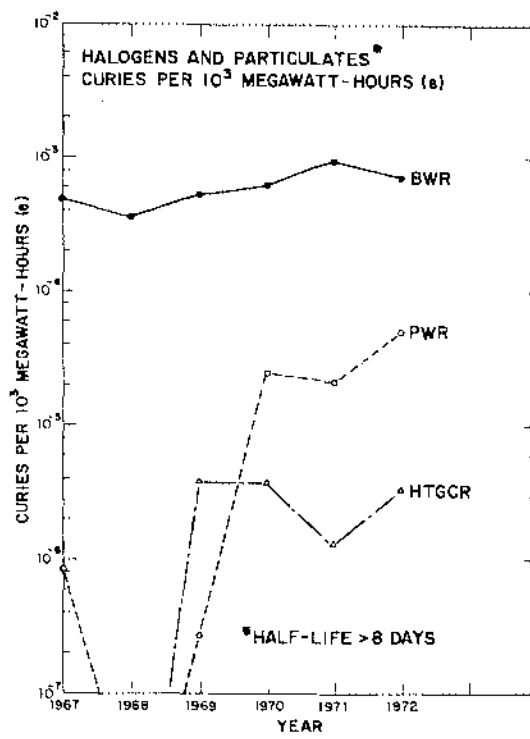


Fig. 2

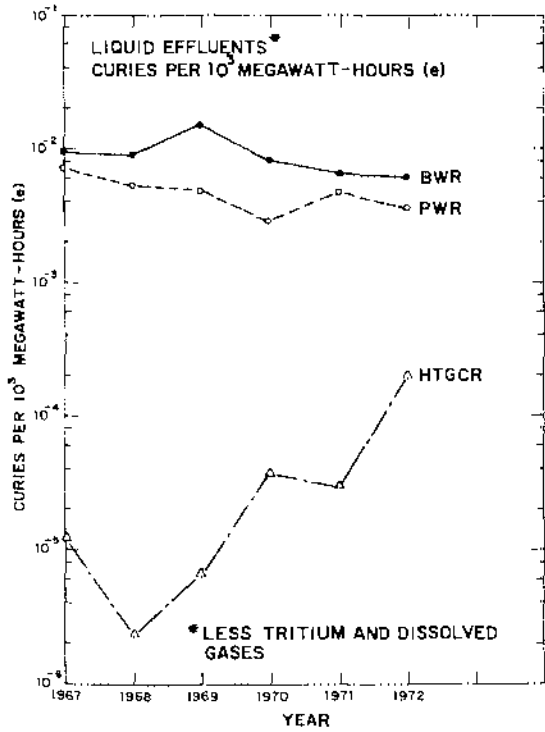


Fig. 3

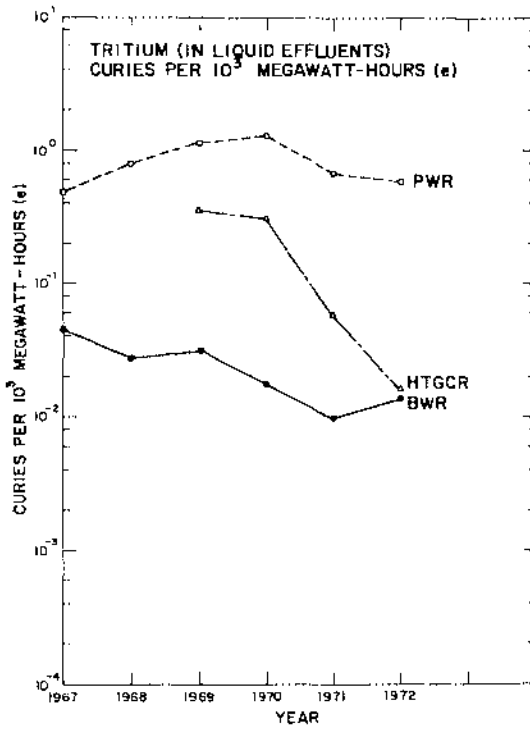


Fig. 4

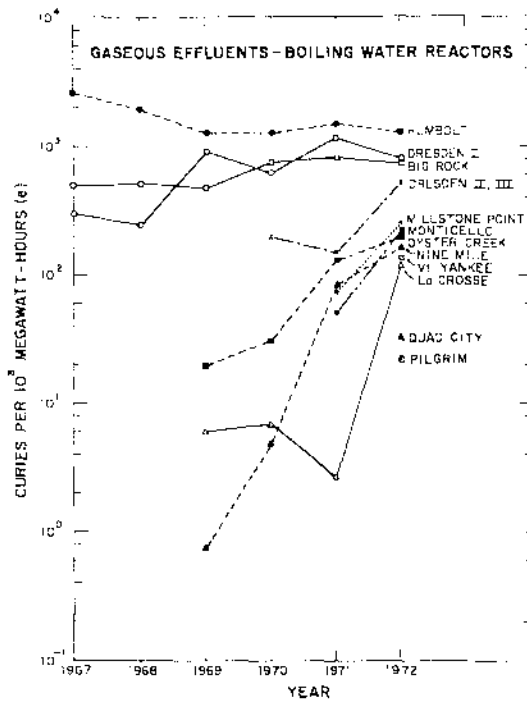


Fig. 1a

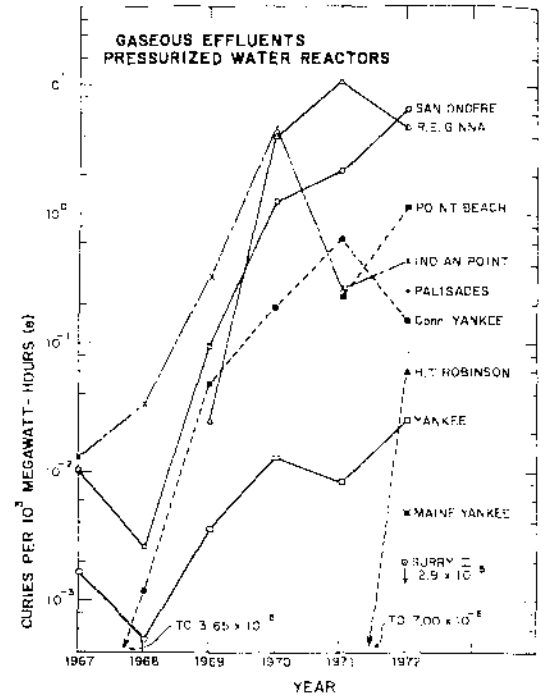


Fig. 1b

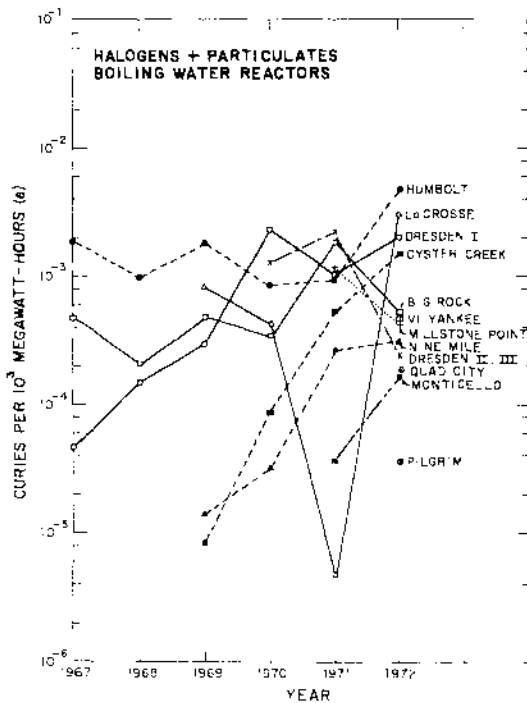


Fig. 2a

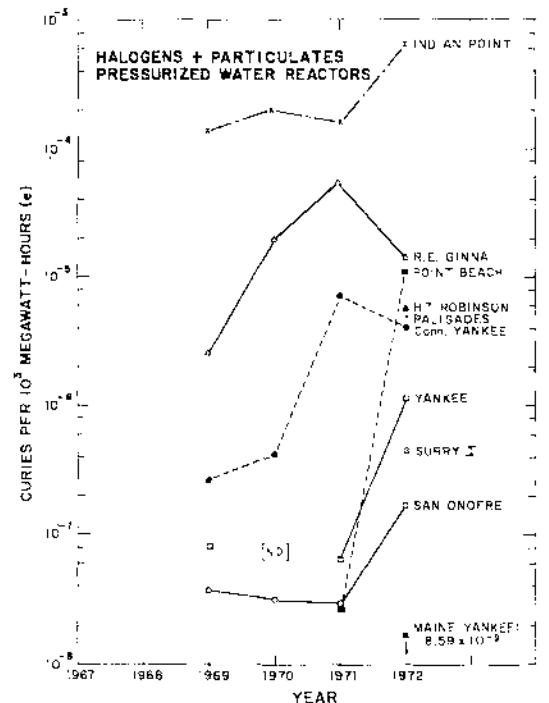


Fig. 2b

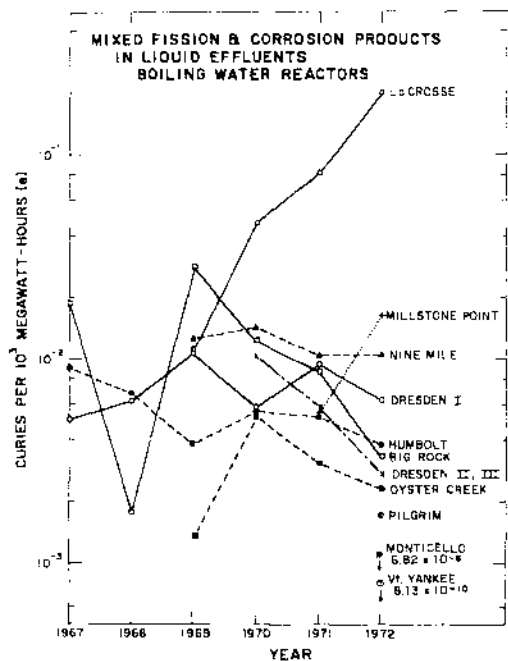


Fig. 3a

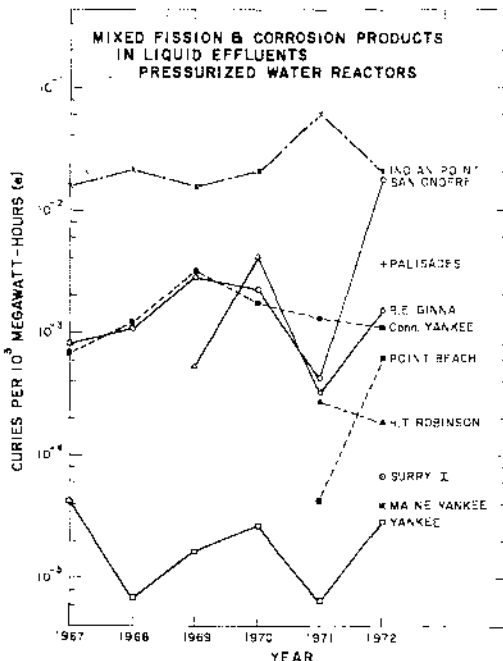


Fig. 3b

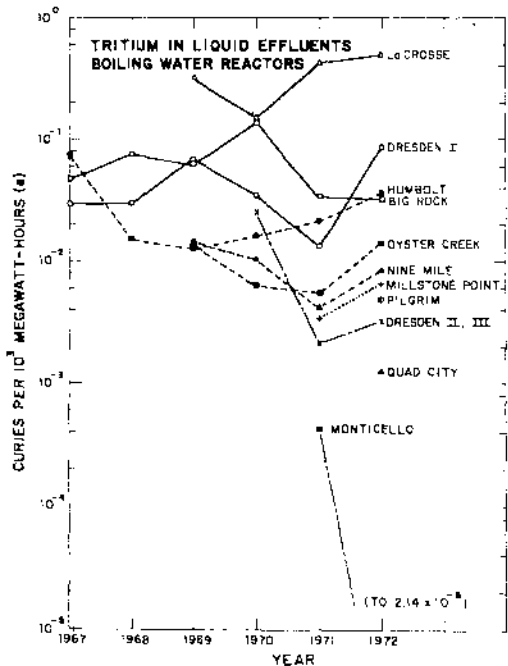


Fig. 4a

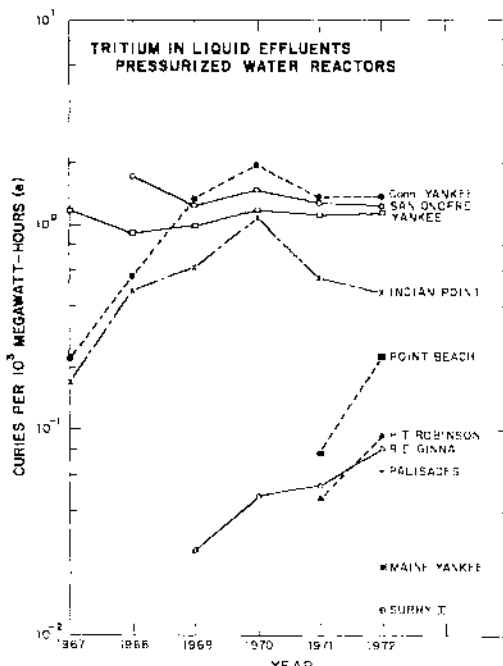


Fig. 4b

As shown in Figure 3, the average mixed fission and corrosion product activity release rates from BWR's have consistently been somewhat larger than those from PWR's. Both have declined somewhat over the period under consideration. While the relative influences of older and "second generation" reactors are not altogether consistent, the individual reactor release rate data shown in Figures 3A and 3B suggest that for both the BWR's and PWR's the release rates from the "second generation" reactors have generally been somewhat less than from the older plants.

From Figure 4, it is obvious that the relative overall release rates of tritium in liquid effluents from BWR's and PWR's has been the reverse of the situation for gaseous effluents. Although rates for both reactor types have declined in recent years, those from PWR's have been about one hundred times the rates from BWR's. In both cases the individual release data shown in Figures 4A and 4B indicate that most of the reduction in the overall averages is attributable to the lower release rates from "second generation" plants.

With appropriate caution that, as suggested by the increasing trend of many of the release rates from older plants, the performance of the newer "second generation" plants might be expected to deteriorate somewhat in this regard over future years, these data appear useful as a means of projecting the anticipated yearly effluent releases from a "reference" 1,000 MW(e) capacity nuclear power reactor, available for 80% of the time as a base load plant. These projections are:

Table II

Projected Effluent Releases of Radioactivity in the Effluents of "Reference" 1,000 MW(e) Power Reactors

	AIRBORNE EFFLUENTS (Curies/year)		LIQUID EFFLUENTS (Curies/year)	
	Gaseous	Halogens & Particulates	Fission and Corrosion Products	Tritium
BWR	1.66x10 <sup>6</sup>	5.31	49.6	104
PWR	9,650	0.17	30.2	5,750
HTGR	2,760	< 0.02	0.27	835

The concern of this paper is with the relationship of effluents from nuclear power reactors and fossil-fueled plants to environmental standards in their immediate vicinities. However, it should be noted that the operation of a nuclear power reactor also results in the release of radioactivity to the environment when its fuel is reprocessed. From data given in Ref. 2, Vol. I, pp. 1711-31, for effluents released between 1966 and 1971 from the one commercial fuel reprocessing plant in the United States, the amounts so released from the reprocessed fuel of the above indicated 1,000 MW(e) capacity power reactor would be as follows:

Table III

Projected Effluent Releases of Radioactivity in the Effluents of a Plant Reprocessing Fuel with a Total Exposure of 8.72 x 10<sup>5</sup> Mwd

	AIRBORNE EFFLUENTS (Ci/yr)		LIQUID EFFLUENTS (Ci/yr)	
	Gaseous	Halogens & Particulates	Fission and Corrosion Products	Tritium
	2.68x10 <sup>5</sup> *	0.40**	53.5***	4,830

\* <sup>85</sup>Kr - 100%; \*\* <sup>131</sup>I - < 0.07 Ci; \*\*\* <sup>90</sup>Sr - 8.3 Ci.

It is also possible to project the release rates from a nuclear power reactor from theoretical considerations, starting with assumed fuel leakage rates, transfer coefficients from the primary to other systems, and the eventual release from the reactor to the environment. Such an approach has been utilized by the AEC in its Environmental Statement<sup>15</sup> concerning the rulemaking action in connection with its proposed numerical guidance for light-water-cooled nuclear power reactors. For 3,500 MW(t) plants with effluent control equipment similar to that employed in currently operating power reactors, their "base case" projections are as follows:

Table IV

AEC "Source Term" Projections of Radioactivity in the Effluents from Base Case 3,500 MW(t) Light-Water-Cooled Nuclear Power Reactors

	AIRBORNE EFFLUENTS (Ci/yr)		LIQUID EFFLUENTS (Ci/yr)	
	<u>Gaseous</u>	<u><sup>131</sup>I</u>	<u>Fission and Corrosion Products</u>	<u>Tritium</u>
BWR	2.9x10 <sup>6</sup>	15	1,800	20
PWR	4.6x10 <sup>4</sup>	1.2	210	350

It appears that for all categories but tritium, the AEC assumptions and calculational method lead to projected releases in excess of those predicated on experience to date. Other aspects of the overall reactor-environmental model utilized by the AEC in framing its proposals to limit radioactivity in light-water-cooled reactor effluents have been discussed elsewhere<sup>3</sup>. The influence of these proposals, whether or not they are finally adopted in their present form, seems likely to considerably diminish the release rates of most categories of nuclear power reactor effluents in the United States, thus making projections based on experience to date upper limits of what may be anticipated.

Comparison With Fossil-Fueled Plant Effluents

When the relationship of conventional pollutants from fossil-fueled power plants to air quality standards is compared to that between airborne radioactive effluents from nuclear power stations and radiation concentration guides, the much greater public concern which the latter has evoked in recent years seems difficult to comprehend. The nature of these relationships is suggested by a straightforward method of calculating the volume of air required to dilute the yearly amounts of various kinds of pollutants or radioactivity emitted from a "reference" 1,000 MW(e) capacity plant to the currently applicable EPA air quality standards or AEC radiation concentration guides. If it is further assumed that the same meteorological considerations, whatever these may be, are applicable, then these need not be specified in making such a comparison.

In the current calculation, the following heat values of fuels have been utilized:

- Bituminous Coal - 26,200 BTU/ton
- Crude Oil - 5,800,000 BTU/barrel
- Natural Gas - 1,035 BTU/ft<sup>3</sup>

Emission factors for various fossil fuels have been obtained from a U.S. Office of Science and Technology report<sup>16</sup>. The amounts of radioactivity in coal and oil originally indicated by Eisenbud and Petrow<sup>17</sup> have been slightly modified, considering more recent data reported<sup>18</sup> in 1968. The results are as follows:

Table V

Volume of Air Required to Meet Concentration Standards  
and Average Site Boundary Concentrations for Yearly Emission  
from a 1,000 Megawatt(e) Power Station  
(Operated as a Base-Load Plant, with an 80% Availability)

Type Plant	Pollutant	Standard <sup>(A)</sup>	Discharge Quantity <sup>(B)</sup>	Dilution Volume (10 <sup>9</sup> m <sup>3</sup> )	Site Bndry <sup>(C)</sup> Concentration
COAL	SO <sub>2</sub> (3.5% S)	0.03 ppm	3.66x10 <sup>8</sup> lb.	2.14x10 <sup>6</sup>	0.20 ppm
	NO <sub>2</sub>	0.05 ppm	5.50x10 <sup>7</sup> lb.	2.49x10 <sup>5</sup>	0.04 ppm
	CO	9.0 ppm*	1.38x10 <sup>6</sup> lb.	63.5	0.02 ppm
	Hydrocarbons	0.24 ppm**	5.50x10 <sup>5</sup> lb.	156	0.001 ppm
	Particulates (97.5% Removal)	75 µg/m <sup>3</sup>	1.25x10 <sup>7</sup> lb.	75,500	18 µg/m <sup>3</sup>
	<sup>226</sup> Ra	2x10 <sup>-12</sup> Ci/m <sup>3</sup>	0.0170 Ci	8.5	4.2x10 <sup>-16</sup> Ci/m <sup>3</sup>
	<sup>228</sup> Th	2x10 <sup>-13</sup> Ci/m <sup>3</sup>	0.108 Ci	708	2.6x10 <sup>-16</sup> Ci/m <sup>3</sup>
OIL	SO <sub>2</sub> (1.5% S)	0.03 ppm	1.23x10 <sup>8</sup> lb.	6.95x10 <sup>5</sup>	0.07 ppm
	NO <sub>2</sub>	0.05 ppm	5.42x10 <sup>7</sup> lb.	2.45x10 <sup>5</sup>	0.04 ppm
	CO	9.0 ppm*	2.08x10 <sup>4</sup> lb.	0.95	2.61x10 <sup>-4</sup> ppm
	Hydrocarbons	0.24 ppm**	1.17x10 <sup>6</sup> lb.	4,720	0.004 ppm
	Particulates (97.5% Removal)	75 µg/m <sup>3</sup>	5.88x10 <sup>6</sup> lb.	35,400	8.4 µg/m <sup>3</sup>
	<sup>226</sup> Ra	2x10 <sup>-12</sup> Ci/m <sup>3</sup>	6.0x10 <sup>-4</sup> Ci	0.3	1.5x10 <sup>-18</sup> pCi/m <sup>3</sup>
	<sup>228</sup> Th	2x10 <sup>-13</sup> Ci/m <sup>3</sup>	1.3x10 <sup>-3</sup> Ci	6.7	3.2x10 <sup>-18</sup> pCi/m <sup>3</sup>
GAS	SO <sub>2</sub>	0.03 ppm	2.78x10 <sup>4</sup> lb.	157	1.5x10 <sup>-5</sup> ppm
	NO <sub>2</sub>	0.05 ppm	2.71x10 <sup>7</sup> lb.	1.23x10 <sup>5</sup>	0.02 ppm
	Particulates (97.5% Removal)	75 µg/m <sup>3</sup>	1.04x10 <sup>6</sup> lb.	6,290	1.5 µg/m <sup>3</sup>
NUCLEAR	<sup>85</sup> Kr & <sup>133</sup> Xe	3x10 <sup>-7</sup> Ci/m <sup>3</sup>	9,650 Ci	33.2 PWR	2.3x10 <sup>-11</sup> Ci/m <sup>3</sup>
	Short-lived radioactive gases	3x10 <sup>-8</sup> Ci/m <sup>3</sup>	1.66x10 <sup>6</sup> Ci	5.54x10 <sup>4</sup> BWR	4.0x10 <sup>-9</sup> Ci/m <sup>3</sup>
	<sup>131</sup> I (Inhalation)	1.0x10 <sup>-10</sup> "	{ 0.2 Ci 5.3 Ci	{ 2.0 PWR 53.0 BWR	{ 4.2x10 <sup>-16</sup> Ci/m <sup>3</sup> 1.3x10 <sup>-14</sup> Ci/m <sup>3</sup>
	<sup>131</sup> I (Air-Grass-Milk)	1.4x10 <sup>-13</sup> " ***	{ 0.2 Ci 5.3 Ci	{ 1,430 PWR 37,800 BWR	{ 4.2x10 <sup>-16</sup> Ci/m <sup>3</sup> 1.3x10 <sup>-14</sup> Ci/m <sup>3</sup>

\* Maximum 8-hour concentration, once per year. Yearly average not specified.

\*\* Maximum 3-hour concentration (6-9 A.M.) once per year.

\*\*\* "Concentration factor" of 700 applied to inhalation standard for <sup>131</sup>I.

(A) EPA "National Primary and Secondary Air Standards", (Federal Register, Vol. 36 No. 84, Part II, pp. 8186-87, 4/30/71), and AEC "Standards for Protection Against Radiation", 10CFR20.

(B) Discharges from PWR and BWR are derived from weighted average 1967-1972 release data as summarized by the Directorate of Regulatory Operations, USAEC.

(C) Based on average X/Q at 500 m (for release height of 100 m) of 6.2x10<sup>-8</sup> sec/m<sup>3</sup>, for 25 operational or proposed nuclear power stations (Table 7.10, Ref. 15).



If the EPA's Air Quality Standards are given the same weights that the AEC's radiation protection standards have been, then the significant contribution of nuclear fueled plants to limiting overall air pollution (or at least minimizing its increase) is obvious. This is especially so relative to coal and oil-fired power plants.

It is of interest to examine the consequences, were the air quality standards applied to a hypothetical individual at the "fence post" at the boundary of conventional fueled plants, as is the practice for nuclear power reactors. The results of such a calculation of average concentrations of conventional pollutants and of radioactivity at 500 meters distance from a 100 meter stack of the "reference" 1,000 MW(e) base load plants, are also indicated in Table V. Unless limited to low sulfur fuels, the reference coal and oil fueled plants would exceed the SO<sub>2</sub> concentration standard. All conventionally fueled plants would approach the NO<sub>2</sub> concentration standard. The reference BWR would approach the gaseous effluent and <sup>131</sup>I radiation concentration standards. However, it should be noted in this connection that the current design provision of catalytic recombiners, which reduce the volume of the BWR off-gas, thereby permitting longer holdup and/or charcoal filtration prior to stack discharge, should materially reduce both of these components of their airborne effluent releases.

The foregoing comparisons admittedly omit the airborne effluent releases from fuel reprocessing plants. However, it appears that these will continue to be located at a relatively few sites more remote from surrounding populations than most future nuclear power reactor sites. If so, the "local" effects of their effluent releases appear less important on an integrated population exposure basis than those from nuclear power plants.

Data for similar comparisons of conventional radioactivity and pollutants in liquid effluents is not available. However, studies(19,20,21) to date in the vicinity of several nuclear power reactors suggest that their radioactive liquid effluents have been dosimetrically insignificant. Comparable studies of the degree of pollution of streams and ground water by releases peculiar to fossil-fueled plants have not appeared in the literature, so it also appears to be minimal.

#### References

1. A.P. Hull and F.J. Shore, "Sternglass: A Case History", BNL-16613 (1972). A review of most of the Sternglass claims.
2. -----, "Environmental Effects of Producing Electric Power", JACE Hearings, Part 2, Vol. II (1970). Contains most of the Gofman-Tamplin papers, as well as related commentary.
3. A.P. Hull, "Reactor Effluents: As Low as Practicable or as Low as Reasonable", Nuclear News (11/72).
4. J.O. Blomeke and F.E. Harrington, "Management of Radioactive Wastes at Nuclear Power Stations", ORNL-4070 (1968)
5. W.L. Brinck and B. Kahn, "Radionuclide Release at Nuclear Power Stations", published in Environmental Surveillance in the Vicinity of Nuclear Facilities, W. Reinig, ed., C.C. Thomas (1970).
6. J.G. Terrill, E.D. Harwood and I. Paul Leggett, "Environmental Aspects of Nuclear and Conventional Power Plants", Journal of Industrial Medicine and Surgery, 36:6 (1967).

7. -----, "National Primary and Secondary Ambient Air Quality Standards", Environmental Protection Agency, Federal Register, 36:84 Part II (1971).
8. B.R. Fish, "The Role of Nuclear Energy in the Control of Air Pollution", Nuclear Safety, 8:4 (1969).
9. A.P. Hull, "Some Comparisons of the Environmental Risks from Nuclear and Fossil-Fueled Power Plants", Nuclear Safety, 12:3 (1971).
10. P. Jansen, S. Jordan and W. Schikerski, "An Approach to Compare Air Pollution of Fossil and Nuclear Power Plants", Environmental Aspects of Nuclear Power Stations, IAEA (1971).
11. C. Starr, M.A. Greenfield and D.F. Hausknecht, "A Comparison of Public Health Risks: Nuclear vs. Oil-fired Power Plants", Nuclear News (10/72).
12. -----, Selected Materials on Environmental Effects of Producing Electric Power, JCAE Print, USGPO 32-466 (1969).
13. -----, "Operating History, U.S. Nuclear Power Reactors", WASH-1203-71 (1971).
14. -----, "Nuclear Reactors Built, Being Built, or Planned in the United States as of Dec. 31, 1972", TID-8200-R27 (1973).
15. -----, "Final Environmental Statement Concerning Proposed Rule Making Action: Numerical Guides for Design Objectives and Limiting Conditions for Operation to Meet the Criterion 'As Low as Practicable' for Radioactive Material in Light-Water-Cooled Nuclear Power Reactor Effluents", WASH-1258 (7/73).
16. -----, "Considerations Affecting Steam Power Plant Site Selection", a report sponsored by the Energy Policy Staff, Office of Science and Technology (1968).
17. M. Eisenbud and H.G. Petrow, "Radioactivity in the Atmospheric Effluents of Power Plants that Use Fossil Fuels", Science, 144 (4/17/64).
18. -----, "Environmental Effects of Fossil-Fuel and Nuclear Power Plants", Progress Report #1, Bureau of Radiological Health, Environmental Control Administration, Public Health Service (10/68).
19. J.W. Lentch, M.E. Wrenn and T.J. Kneip, "Manmade Nuclides in the Hudson River Estuary", presented at the Fifth Annual Health Physics Society Mid-Year Topical Symposium, Idaho Falls (11/70).
20. B. Kahn et al., "Radiological Surveillance Studies at a Boiling Water Nuclear Power Station", BRH/DER 70-1 (1970).
21. B. Kahn et al., "Radiological Surveillance Studies at a Pressurized Water Nuclear Power Station", USEPA Report RD 71-1 (1971).

## ENVIRONMENTAL TRITIUM AND THE DOSE TO MAN

Burton G. Bennett

Health and Safety Laboratory  
U. S. Atomic Energy Commission  
New York, New York 10014

### Abstract

The dose to man from acute or chronic intake of tritiated water has been determined using a three compartment model to describe the retention of tritium in loose water and in bound organic form in the body. The retention times determined from occupational exposure cases are reviewed. The total tissue dose, using representative retention half-times for the three components of 9, 30, and 450 days, is 84 mrad per 1 mCi intake by standard man, 84% of which is due to tritium in body water and 16% to bound tritium in tissue. The record of environmental tritium concentrations in surface waters has been compiled. Maximum levels caused by weapons testing occurred in 1963-64. Subsequent decrease is exponential with a half-time of 3.2 years. By relating environmental tritium levels to daily intake by man and applying the dose model, the dose commitment of 1.5 mrad from fallout tritium was determined.

### Introduction

Tritium is produced naturally by cosmic ray interactions in the atmosphere. Superimposed on the natural tritium background are varying amounts of man-made tritium. Nuclear weapons testing introduced substantial amounts of tritium into the environment, although the levels of fallout tritium are currently declining. The expanding nuclear power industry is expected to cause increasing amounts of tritium to be released. These continual additions of man-made tritium to the environment are cause for repeated monitoring of environmental levels of tritium and for re-evaluation of the dose consequences.

In this study a detailed tritium dose model is presented which allows one to determine accurately the contribution to tissue dose from tritium in loose water in tissue and from tritium combined in tissue following acute and chronic intakes. The experience from occupational exposure cases is reviewed to indicate the average half-times of the retention components. A record of tritium concentrations in environmental surface water is compiled. By relating these levels to the daily tritium intake by man and applying the dose model, the dose commitment from weapons-produced tritium has been computed.

### Tritium Retention in Man

The experience from observations of human cases of accidental tritium exposure is summarized in Table 1. The initial rate of elimination of tritium from the body is exponential with a half-time ranging from 4 to 18 days.<sup>1-8</sup> Identification of additional retention components is limited to the few cases reported with intakes large enough to allow relatively long-term monitoring of tritium excretion. In the 415 day observation of a case, Sanders and Reinig<sup>9</sup> identified three retention components of half-times 6.1, 23, and 344 days. The first component can be associated with retention of free water in the body and the other two components with bound tritium in tissue. It should be noted, that a diuretic was administered in this case from the 3rd to 35th day following the exposure, which undoubtedly increased the turnover rate of body water.

TABLE 1  
TRITIUM RETENTION HALF-TIMES IN MAN

Year	Investigator	Cases Studied	Notes	T <sub>1</sub>	T <sub>2</sub>	T <sub>3</sub>
1951	Finson, Anderson <sup>1,2</sup>	9	range 9.3-13	11.3		
1957	Fallot <i>et al.</i> <sup>3</sup>	20	range 5-11	8.5		
1960	Foy, Schnieden <sup>4</sup>	10	high ambient temp.	7.5		
1962	Richmond <i>et al.</i> <sup>5</sup>	5		9.5		
1963	Wylie <i>et al.</i> <sup>6</sup>	7	range 6-12	8.5		
1965	Butler, Leroy <sup>7</sup>	310	range 4-18	9.5		
1966	Osborne <sup>8</sup>	30	range 6.4-14.4	10.5		
1968	Snyder <i>et al.</i> <sup>10</sup>	1		8.7	34	
1968	Sanders, Reinig <sup>9</sup>	1	diuretic used	6.1	23	344
1969	Minder <sup>13</sup>	1			10-30	139-230
1971	Lambert <i>et al.</i> <sup>12</sup>	1		9.1	36	
1972	Moghissi <sup>14</sup>	3			21-26	280-550
Reasonable Range:				8.5-11	20-36	200-550
Assumed Average:				9	30	450

Snyder *et al.*<sup>10</sup> studied another case for 255 days and identified two retention components of half-times 8.7 and 34 days. No particular treatment procedure was applied to this individual following exposure. Further analysis of this data has shown that the data are not inconsistent with a fit by three components of half-times 8.7, 30, and 550 days.<sup>11</sup>

Another case was recently reported by Lambert, Sharpe, and Dawson.<sup>12</sup> Retention half-times of 9.1 and 36 days are very similar to the results reported by Snyder *et al.* The observation period, 161 days, is not quite long enough to allow a definitive fit with three retention components.

Approximate long-term retention components have been inferred from a few additional studies of occupational exposures.<sup>13,14</sup> The differences in retention times in the cases reported allude to the variability which can be anticipated due to differences in metabolism, age, water intake, ambient temperature and treatment procedures. Reasonable ranges of the retention components have been indicated in Table 1. Approximate average retention half-times of 9, 30, and 450 days have been assumed for the model calculations.

#### Tritium Dose Model

##### Acute Intake

The dose to tissue following an acute intake of tritium arises from tritium in body water and from bound tritium in tissue. In computing the dose to tissue, the combined tritium in tissue is often neglected, it being regarded as an insignificant contributor to the total dose. Alternatively, the dose to body water (or a tissue with 100% water) is computed, this dose being a conservative estimate of the total tissue dose.<sup>15</sup> Since the tissue dose is more a measure of the hazard than the water dose, it is useful to determine accurately the dose to tissue.

A three compartment model is necessary to reflect the three retention components that have been identified. Sanders and Reinig<sup>9</sup> suggested the model represented by the diagram in Figure 1. A is the body water compartment, and B and C are bound hydrogen (tritium) compartments. The transfer coefficients represents constant fractional exchange rates of the compartment hydrogen (tritium) contents.

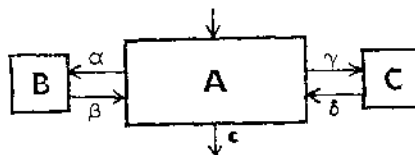


Figure 1. Three compartment model of tritium in the body.

A Markov chain calculation is used to determine the tritium concentrations in the three separate compartments at various times after intake. The fraction of assimilated

tritium in each compartment,  $f_A$ ,  $f_B$ ,  $f_C$ , are computed for as many iterations as may be required (each iteration representing one day). In analyzing the occupational exposure cases, combinations of exchange and elimination rates were selected so that close agreement was obtained with the measured fractional assimilated tritium in the body water.<sup>9,11</sup> For other calculations the transfer coefficients correspond to assumed retention half-times and compartment sizes.

The cumulative dose to tissue from an acute intake of HTO is computed from this model as follows:

$$\text{Dose (wet tissue)} = .75 \int_0^{\infty} q_A f_A dt + .25 \int_0^{\infty} (q_B f_B + q_C f_C) dt$$

The first term is the dose to tissue due to tritium in loose water within tissue. The second term is the contribution to tissue dose from tritium combined in tissue. Wet tissue is assumed to consist of 75% water and 25% tissue solids. A 70 kg man contains an average 42 kg water and an estimated 10 kg active tissue solids (70 kg - 42 kg water - 13 kg fat - 5 kg mineral bone).<sup>16</sup> The specific activities of tritium in the free water and bound compartments following an intake,  $I$  (mCi), are  $q_A = I/42$  mCi/kg and  $q_{B,C} = I/10$  mCi/kg times the fractional amount of the intake in these compartments,  $f_A$ ,  $f_B$  and  $f_C$ , respectively. The relationship .29 mrad per mCi day/kg is used to convert the units.

A calculation based on average retention half-times of 9, 30, and 450 days results in an estimated total tissue dose of 84 mrad per mCi intake.<sup>11</sup> This dose is due 84% to tritium in water within the tissue and 16% to combined tritium in tissue. Fifty percent of the total tissue dose is delivered within 11 days, and 90% of the dose is delivered within 200 days. The bound hydrogen compartment sizes were inferred to be 120 g for B and 600 g for C, consistent with the total amount of tissue solids (10 kg) consisting of ~7% hydrogen. A maximum of 0.7% of the tritium intake becomes combined, this maximum being reached in about 20 days. The rapidly declining concentration of tritium in the body water allows only relatively small amounts of the intake to become combined. The bound tritium in tissue is, thus, not the major contributor to the total dose to tissue following a single intake of HTO, but its contribution (16%) is not insignificant.

In additional model calculations with the first component half-time  $T_1$  varying from 6 to 12 days with 30 and 450 day combined components, an empirical relationship is obtained.

$$\text{Tissue Dose} = 9 T_1 + 3 \text{ mrad per mCi intake}$$

The relationship reflects a direct dependence of the amounts of bound tritium on the amount of tritium in the source reservoir, the body water compartment. For the assumed compartment sizes and within reasonable variation of transfer rates from the bound compartments, the second and third component half-times are not required in this relationship. The formula should be generally useful in providing an approximate estimate of the tissue dose following observation of the initial removal rate.

Intake of tritium other than in the form HTO may require special consideration. Bound tritium compounds in food may be more directly assimilated into bound compounds in tissue. Further study will be required to give the significance of the form of the tritium intake.

#### Chronic Intake

The model can be used for chronic intake situations by computing the compartment contents on a daily basis. For chronic intake of  $1 \mu\text{Ci}/\ell$ , the equilibrium dose rate to active wet tissue is 95 mrad/yr. The dose to body water is

$$.001 \frac{\text{mCi}}{\text{kg}} \times .29 \frac{\text{rad/d}}{\text{mCi/kg}} \times 365 \frac{\text{d}}{\text{yr}} = 106 \frac{\text{mrad}}{\text{yr}}$$

The dose to tissue containing 75% water is  $106 \times .75 = 80$  mrad/yr, neglecting the dose due to tritium combined in tissue solids. The dose to active tissue is thus 1.2 times the dose due to HTO in tissue.

The dose model assumes that the tritium becomes uniformly combined in actively

metabolizing tissue and that all of the hydrogen of active tissue solids is exchangeable. These assumptions lead to conservative estimates of the dose.

In the model calculations, a water balance of 3.08 l/d has been assumed, based on the 42 kg body water compartment size and transfer coefficients corresponding to the 9, 30, and 450 day retention half-times. For an HTO intake of 1 mCi/d, the equilibrium specific activity is

$$1 \text{ mCi/d} \div \left[ 3.08 \frac{\text{kg H}_2\text{O}}{\text{d}} \times \frac{1 \text{ kg H}}{9 \text{ kg H}_2\text{O}} \right] = 2.92 \text{ mCi/kg H}$$

The tritium content of each compartment at equilibrium (the hydrogen content times the specific activity) is 13.6, .35, and 1.75 mCi for A, B, and C, respectively. Radioactive decay allows the long half-time compartment, C, to reach only about 90% of the intake specific activity. The equilibrium dose rate to wet tissue is

$$\begin{aligned} \text{A} \quad & \frac{13.6 \text{ mCi}}{42 \text{ kg H}_2\text{O}} \times \frac{.75 \text{ kg H}_2\text{O}}{\text{kg wet tissue}} \times \frac{.29 \text{ mrad/d}}{\text{mCi/kg}} = 70 \frac{\text{mrad}}{\text{d}} \\ \text{B} \quad & \frac{.35 \text{ mCi}}{10 \text{ kg solids}} \times \frac{.25 \text{ kg solids}}{\text{kg wet tissue}} \times \frac{.29 \text{ mrad/d}}{\text{mCi/kg}} = 2.5 \frac{\text{mrad}}{\text{d}} \\ \text{C} \quad & \frac{1.75 \text{ mCi} \times .90}{10 \text{ kg solids}} \times \frac{.25 \text{ kg solids}}{\text{kg wet tissue}} \times \frac{.29 \text{ mrad/d}}{\text{mCi/kg}} = 11.4 \frac{\text{mrad}}{\text{d}} \\ & \text{Total: } 84 \frac{\text{mrad}}{\text{d}} \end{aligned}$$

As expected, the dose commitment following a single intake (84 mrad per mCi intake) becomes the equilibrium dose rate for chronic intake (84 mrad/d per 1 mCi/d intake).

#### Fallout Tritium

##### Levels in Surface Waters

Tritium is produced by fusion bombs and also by neutrons released during fission bomb explosions and in small amounts by the fission process itself. The amount of bomb-produced tritium is uncertain. Eriksson estimated that 1900 MCi were produced from 1952 through 1962.<sup>17</sup> This compares with natural tritium production of around 2 to 6 MCi/yr. Additional amounts of tritium have been released into the atmosphere by French and Chinese weapons testing conducted since 1964, but the total is a small fraction of that released during the earlier testing period.

The effect of weapons-produced tritium on concentrations of tritium in the environment, in drinking water, food and in man has not been monitored in great detail. Data from the U. S. Geological Survey's program of analyzing river water for tritium content provides the most useful data for ascertaining the tritium levels in environmental waters in the U. S. The data for 1961-68 for 20 streams throughout the U. S. have been published.<sup>18</sup> Preliminary data for 15 rivers for 1969-70 have also been obtained.<sup>19</sup> Tritium data from the earlier testing period, 1952-61, are not nearly as complete. Measurements of Mississippi River water for 1954-57<sup>20,21</sup> and of Rio Grande River water for 1957-58<sup>22</sup> have been included, and comparisons were made with Ottawa River data<sup>23</sup> in arriving at representative average tritium levels.<sup>24</sup>

Figure 2 shows the average U. S. data. The concentration of tritium in environmental waters reflects the weapons testing activity. Sharp increases are indicated following the testing series in 1954, 1956 and 1958. The very active test period in late 1961 and 1962 caused the peak concentrations in U. S. rivers in 1963-64. The declines in concentration during the 1959-60 moratorium and after the 1963 Test Ban Treaty are evident. Recent high yield tests in the Northern Hemisphere by the Chinese may be responsible for the relatively higher tritium levels measured in 1969-70 as compared to the previously declining values.

The average U. S. river tritium concentrations declined with a half-time of 3.2 years from 1963 until 1969 and 5.0 years during 1969 and 1970. Tap water analyzed at the Health and Safety Laboratory in New York City reflecting the lower Hudson Valley watershed area, showed somewhat lower tritium concentrations than the U. S. average in 1970. More recent tap water samples would indicate that the U. S. average beyond 1970 resumes the 3.2 year half-time decline. The absence of high yield atmospheric tests in the Northern Hemisphere in the past two years would also support this assumption.

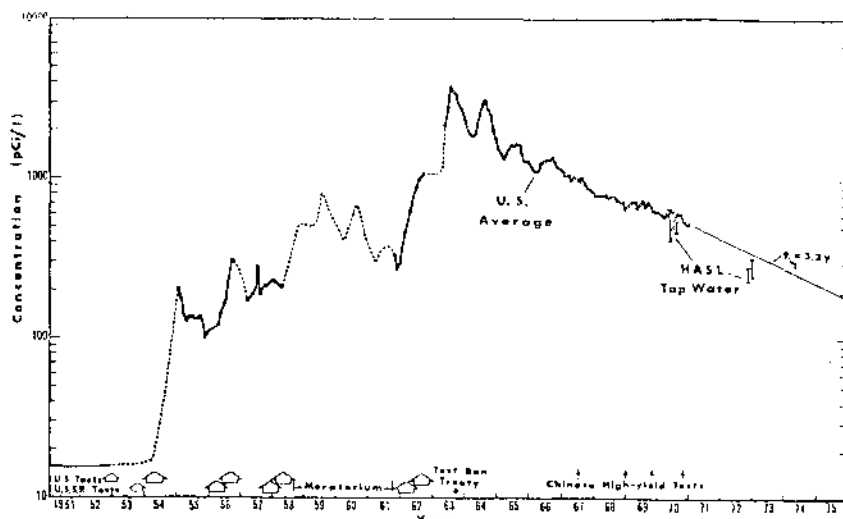


Figure 2. Environmental tritium in surface waters.

### Tritium Intake

If a relationship can be found between the variations in tritium levels in environmental waters and the variations in the amounts of tritium intake by man or the levels of tritium in the body, the dose to man can be determined. The data of Bogen<sup>25</sup> show the relationship for New York in 1970 between the tritium levels of drinking water (530 pCi/l), loose water in food (860 pCi/l), oxidation water of food (2050 pCi/l), and water vapor in air (930 pCi/l). The drinking water intake by man can be assumed to be 1.4 l/d. Intake by inhalation and transpiration of water vapor is computed to be .13 l/d, based on average air temperature and humidity. The food water intake, based on analysis of food items of a standard consumption diet, was found to be 1.26 l/d loose water and .29 l/d oxidation water.<sup>25</sup> The effective concentration of the total tritium intake is thus:

$$(1.4 \times 530 + .13 \times 930 + 1.26 \times 860 + .29 \times 2050) \div 3.08 = 825 \text{ pCi/l}$$

These data indicate that, currently, one should assume somewhat higher concentrations of tritium intake than indicated by the tritium concentration in drinking water. During the earlier fallout period, when environmental levels of tritium were increasing, the tritium concentrations in water were probably higher than in food. Since specific data are lacking, it is probably most reasonable to assume that the tritium levels in surface waters reflect directly the tritium intake by man.

### The Dose Commitment

The average natural concentration of tritium in environmental waters (16 pCi/l) results in a dose to man of 1.5 mrad/yr, based on the three compartment dose model. The dose commitment to man from weapons-produced tritium is the dose due to the increases in tritium intake concentrations above the natural level. Extrapolation of the increased levels beyond 1972 is necessary to account for the exposure which is yet to be experienced. Assuming a 3.2 year half-time for weapons-produced tritium in the environment beyond 1970, a return to the natural background tritium level occurs in 1987.

The tritium intake is assumed to be 3.08 l/d times the effective tritium intake concentration (the average tritium concentration in surface waters). The dose model is used to determine the bound and loose tritium compartment contents and the dose for the entire period that the tritium levels are above the natural background. The dose commitment thus determined is 1.5 mrad. Details of the calculation are shown in Table 2. The highest annual dose due to weapons-produced tritium was .21 mrad in 1963 and 1964. The calculation assumes no further atmospheric weapons testing and takes no account of future tritium releases from nuclear facilities.

The ratios of specific activities (pCi <sup>3</sup>H per kg hydrogen) of the organic bound and

RELATIONSHIP BETWEEN TISSUE AND BOUND-LOOSE TRITIUM IN THE BODY

Year	Tissue Dose (rad)		Bound-Loose Ratio	Year	Tissue Dose (rad)		Bound-Loose Ratio	Year	Bound-Loose Ratio		Bound-Loose Ratio
	Fallout	Total			Fallout	Total			Fallout	Total	
1952	3.6	1.5	.92	1965	156.8	126.3	1.21	1978	8.5	11.0	1.32
1953	0.1	1.6	.90	1966	122.0	121.5	1.17	1979	7.6	8.9	1.32
1954	3.8	5.1	.79	1967	97.0	98.5	1.15	1980	5.7	7.2	1.32
1955	10.5	12.0	.73	1968	76.4	77.9	1.28	1981	4.3	5.8	1.32
1956	76.4	17.0	.63	1969	68.4	69.9	1.21	1982	3.2	4.7	1.32
1957	18.6	9.8	.75	1970	58.7	59.6	1.21	1983	2.3	3.8	1.32
1958	10.3	31.8	.57	1971	47.4	48.9	1.25	1984	1.6	3.1	1.32
1959	53.1	56.6	.45	1972	38.0	39.3	1.28	1985	1.0	2.5	1.42
1960	47.2	58.7	.41	1973	30.4	31.9	1.40	1986	0.5	2.0	1.32
1961	12.1	33.8	.95	1974	24.1	25.0	1.41	1987	0.2	1.7	1.21
1962	36.6	28.1	.81	1975	18.1	20.8	1.52	1988	0.1	1.6	1.08
1963	210.3	232.0	.73	1976	15.1	16.8	1.32	1989	0.1	1.6	1.01
1964	210.9	232.6	1.10	1977	12.1	13.6	1.32	1990	0.0	1.5	.97

Dose Commitment: 1.5 mrad

loose water tritium in the body, determined from this computation, are included in Table 2. The ratio would be 1.0 under natural conditions, except that radioactive decay does not allow complete equilibrium in the bound compartment with the slowest turnover time. As the concentrations of tritium intake increase, labeling of the bound components lag with respect to the specific activity of the loose water compartment. The bound-loose tritium label is retained longer than the activity in body water, and the bound-loose tritium ratio becomes greater than one. The ratio is currently about 1.3 in man, according to this computation, which is not unlike the ratios actually being measured in human tissue.<sup>26</sup>

#### Other Estimates of the Dose Commitment

Indirect estimates of the dose commitment can be made by comparing the total amount of weapons-produced tritium with the natural production rate, as is done by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR).<sup>27</sup> The uncertainties involved, however, affect the accuracy of these estimates.

Most of the weapons-produced tritium, an estimated 1900 MCi,<sup>17</sup> was introduced into the Northern Hemisphere. The stratospheric residence time of tritium (~1 yr) does not allow inter-hemispheric mixing to be an important factor. Ten percent may be a reasonable approximation for stratospheric tritium carried to the Southern Hemisphere.<sup>28</sup> The dose commitment for the Northern Hemisphere is consistent with the above direct computation if the natural production rate is taken as 3.4 MCi/yr (1.7 MCi/yr in each hemisphere). The natural background tritium dose to man is .0015 mrad/yr.

$$.0015 \text{ mrad/yr} \times \frac{1900 \text{ MCi} \times .90}{1.7 \text{ MCi/yr}} = 1.5 \text{ mrad}$$

UNSCEAR computed a range of possible dose commitments for the Northern Hemisphere, based on various assumptions of the natural tritium background and inter-hemispheric mixing of stratospheric weapons tritium. A conservative estimate for the Northern Hemisphere fallout tritium dose commitment of 4 mrad was obtained.<sup>27</sup>

Tritium released from nuclear power facilities are as yet insignificant, compared to the amounts of weapons-produced tritium. It will be desirable, however, to continue to measure the environmental tritium levels and to ascertain the contribution from nuclear power activities. With use of the dose model presented here, the dose commitment from future activities involving release of tritium to the environment can be determined.

#### References

1. Pinson, E. A., E. G. Anderson, Los Alamos Report LA-1218 (1951).
2. Pinson, E. A., W. H. Langham, J. Appl. Physiol. 10, 108 (1957).
3. Fallot, P., A. Aeberhardt, J. Masson, Int. J. Appl. Rad. Iso. 1, 237 (1957).
4. Foy, J. M., H. Schnieden, J. Physiol. 154, 169 (1960).
5. Richmond, C. R., W. H. Langham, T. T. Trujillo, J. Cell Comp. Physiol. 59, 45 (1962).
6. Wylie, K. F., W. A. Bigler, G. R. Grove, Health Phys. 9, 911 (1963).
7. Butler, H. L., J. H. Leroy, Health Phys. 11, 283 (1965).
8. Osborne, R. V., Health Phys. 12, 1527 (1966).
9. Sanders, S. M., W. C. Reinig, Diagnosis and Treatment of Deposited Radio-nuclides, p. 534, Excerpta Medica Foundation (1968).



10. Snyder, W. S., B. R. Fish, S. R. Bernard, M. R. Ford, J. R. Meir, *Physics Med. Biol.* 13, 547 (1968).
11. Bennett, B. G., USAEC Report HASL-253 (1972).
12. Lambert, B. E., H. B. A. Sharpe, K. B. Dawson, *Am. Ind. Hyg. Ass. J.* 32, 682 (1971).
13. Minder, W. *Strahlentherapie* 137, 700 (1969).
14. Moghissi, A. A., M. W. Carter, E. W. Bretthauer, *Health Phys.* 23, 805 (1972).
15. Osborne, R. V., *Rad. Res.* 50, 197 (1972).
16. Woodard, H. Q., USAEC Report HASL-229 (1970).
17. Eriksson, E., *Tellus XVII*, 1 (1965).
18. Wyerman, T. A., R. K. Farnsworth, G. L. Stewart, *Rad. Health Data and Rep.* 11, 421 (1970).
19. Wyerman, T. A., private communication (1972).
20. Begemann, F., W. F. Libby, *Geochimica et Cosmochimica Acta* 12, 277 (1957).
21. Begemann, F., A. Turkevich, AFOSR Report TR-5841 (1957).
22. von Buttlar, W., *J. Geophys. Res.* 64, 1031 (1959).
23. Brown, R. M., *Geochimica et Cosmochimica Acta* 21, 199 (1961).
24. Bennett, B. G., USAEC Report HASL-268, p. I-50 (1973).
25. Bogen, D. C., C. A. Henkel, C. G. C. White, G. A. Welford, *Proceedings Tritium Symposium, Las Vegas (1971)*.
26. Bogen, D. C., private communication (1972).
27. UNSCEAR, *Ionizing Radiation: Levels and Effects*, vol. 1, p. 41, United Nations (1972).
28. Krey, P. W., B. Krajewski, USAEC Report HASL-215 (1969).

## RISQUE INDIVIDUEL, DOMMAGE COLLECTIF, ET CENTRALES NUCLEAIRES

M. DELPLA

Comité de radioprotection, EDF  
Paris, France

Abstract - The author specifies the way that leads from an individual risk to the theoretical collective damage in the matter of irradiation by nuclear power plants; in so doing he endeavours to show people's reactions towards nuclear power plants can be altered and on the other hand set free from any emotional loads. Finally he states the necessity of defining our concepts precisely and of giving them denominations that cause no anxiety.

### 1 - Introduction

Les termes "risque" et "dommage" foisonnent dans la littérature qui traite de radioprotection. De nombreux auteurs les emploient indifféremment; certains vont même jusqu'à désigner par risque, ou par dommage, les effets différés de l'irradiation, .. ou l'irradiation elle-même.

La personne (physique ou morale) qui crée un risque doit — sans contexte — réparation du dommage, s'il s'en produit. En ce qui concerne les centrales nucléaires le risque proviendrait, si l'on n'y prenait garde, des rayonnements ionisants. Le dommage s'exprimerait, par exemple, en cancers, en "morts génétiques". En matière de réparation, on rechercherait un lien de causalité entre le risque créé et le dommage incriminé, cas par cas, et a posteriori.

En 1955, à Genève, au cours de la conférence organisée par l'ONU sur les utilisations pacifiques de l'énergie nucléaire, le monde industriel et scientifique a cru — sans doute un peu prématurément — que cette forme d'énergie allait vite prendre le pas sur les autres formes, grâce aux centrales nucléaires.

L'hygiéniste, jusque là, observait l'évolution de l'état sanitaire de la population soumise à une pollution industrielle croissante; il suivait les résultats des recherches accomplies en vue de limiter cette pollution; au besoin, il engageait le législateur à prescrire des mesures de précaution. L'un et l'autre se résignaient: l'amélioration du niveau de vie et, partant, du bien-être et de l'état sanitaire, due au développement industriel, permettait de tolérer des inconvénients jugés raisonnablement supportables. Connaissant les dangers des rayonnements ionisants et, partant, d'un développement anarchique de l'industrie nucléaire, sous le couvert et sous l'autorité de la commission internationale de radioprotection (CIRP), il a émis des recommandations<sup>1</sup> suivant lesquelles le législateur a, préventivement, limité l'irradiation des personnes à un niveau jugé très prudent. Par la suite, n'ignorant pas l'importance des décisions prises, il a voulu prévoir quelles pourraient être les conséquences lointaines de cette irradiation. Ses prévisions, qu'il juge admissibles, alarment le profane.

Dans ce texte, de longueur limitée, destiné à souligner la nécessité de définitions, je me bornerai à reprendre des notions récemment exposées ailleurs<sup>2-4</sup>. Je vais seulement rappeler comment, pour ascoir ses prévisions, l'hygiéniste est passé de la notion de risque individuel à celle de dommage collectif; ce faisant, je mettrai en relief la nature toute théorique du risque qui sert de base au calcul de ce dommage, ainsi que la nécessité d'éviter les dénominations qui, sans raison, donnent prise à l'angoisse.

## 2 - Risque individuel

Préalablement, je proposerai une définition du risque individuel. La recherche de son expression quantitative se heurte à d'innombrables difficultés : l'hygiéniste a adopté une relation simplifiée, d'où découle un risque bien particulier. J'ai proposé de l'appeler : "risque individuel théorique", pour bien éviter de la confondre avec le précédent. Diverses dénominations utilisées, dont je donnerai quelques exemples, ne sont pas sans ambiguïté.

### 2.1 - Définition

D'une façon générale, on peut définir un risque individuel par la probabilité, pour une personne placée dans des circonstances particulières, de subir un préjudice du fait de ces circonstances.

Ici, les "circonstances particulières" sont dues à l'irradiation ajoutée par l'industrie nucléaire. Le préjudice se traduirait par des dommages corporels observables sur certaines des personnes irradiées (effets somatiques différés) ou seulement sur leur descendance (effets génétiques).

### 2.2 - Recherche d'une relation quantitative

Les affections susceptibles de résulter de l'irradiation ne lui étant pas spécifiques, un rapport de causalité entre risque individuel et irradiation ne peut être établi que statistiquement : on compare des groupes d'individus identiques, sauf pour ce qui concerne celle-ci. Dans chacun de ces groupes on détermine la proportion des personnes atteintes d'une affection déterminée; soit  $p$  sa valeur dans un groupe qui a été irradié, et  $p_0$  sa valeur dans le groupe témoin, non irradié. La différence  $(p - p_0)$  est fonction : du débit d'équivalent de dose ( $d$ ), de l'équivalent de dose ( $D$ ), du champ d'irradiation ( $f$ ). La forme de la fonction dépend en outre de l'affection considérée (cancer, cause de mort prématurée, par exemple) de la population (âge, sexe, mode de vie, etc...); elle dépend aussi des conditions d'observation, laquelle débute au temps  $\tau$  après le début de l'irradiation (suivant la valeur de  $\tau$ , elle peut porter sur la population elle-même, sur sa progéniture, ou sur sa descendance lointaine) et se prolonge un temps  $t$ .

A condition d'être connus avant l'irradiation, ayant été déterminés sur des groupes semblables d'effectif suffisant,  $p$  et  $p_0$  - et aussi leur différence - peuvent être assimilés à des probabilités. Pour en simplifier la détermination, le chercheur fixe les caractéristiques de l'irradiation ( $d$ ,  $D$  et  $f$ ), il arrête les conditions d'observation ( $\tau$  et  $t$ ), il précise les caractéristiques de la population observée, enfin, il choisit l'affection dont il recherchera le diagnostic. En irradiant plusieurs groupes, donnant à  $D$  autant de valeurs qu'il le juge utile, il peut, du moins en principe, pour un individu semblable à ceux qui constituent les groupes et pour l'affection considérée, traduire la variation du rapport de causalité graphiquement, et même mathématiquement, par une approche statistique, avec une relation de la forme :

$$(p - p_0) = \psi(D). \quad (1)$$

Généralement trouvée positive pour des valeurs de  $D$  très élevées, la probabilité  $(p - p_0)$  peut, lorsque  $D$  diminue : au mieux, s'annuler en changeant de signe, au pire, ne plus différer statistiquement de zéro. Cette dernière éventualité (due, soit à ce que l'irradiation n'a produit aucun effet traduit par l'affection recherchée, soit à ce que les effectifs des groupes étaient insuffisants) laisse le chercheur, et, partant, l'hygiéniste, dans l'incertitude.

Comme de fortes irradiations peuvent provoquer des affections nombreuses et comme, pour chaque affection,  $(p - p_0)$ , fonction de D, dépend de nombreux paramètres, pragmatique, la CIRP<sup>1</sup> a adopté la forme de relation quantitative la plus simple.

### 2.3 - La relation simplifiée adoptée

La CIRP a fait l'hypothèse que, pour toute affection que peut provoquer l'irradiation, la probabilité d'induction, indépendante de d, est proportionnelle à D. Si k représente la constante de proportionnalité, on peut écrire, pour l'affection considérée :

$$(p - p_0) = k D . \quad (2)$$

La valeur numérique de  $(p - p_0)$  étant obtenue à partir de valeurs élevées de d et de D, il me paraît important, dans le domaine des faibles doses, d'écrire :

$$(\pi - \pi_0) = \chi D . \quad (3)$$

pour bien marquer, par des lettres différentes, l'hypothèse de la validité de la substitution de  $\chi$  à k. Comme  $(p - p_0)$ , la valeur calculée  $(\pi - \pi_0)$  est aussi dénommée "probabilité". Voyons de plus près.

### 2.4 - Dénomination proposée : risque individuel théorique

Dans l'expression (2) ci-dessus, la différence  $(p - p_0)$  ne peut qu'être positive : cette variation de probabilité est donc un risque, un risque supplémentaire. On assimile  $(\pi - \pi_0)$  au risque individuel ajouté par toute irradiation de D rem. Pour bien marquer que, dans le domaine des valeurs faibles de D,  $(\pi - \pi_0)$  ne correspond à aucune observation ou expérimentation, je l'ai appelé : risque individuel théorique (RIT).

En pratique, l'observation porte sur des groupes d'individus qui diffèrent, mais que l'on recrute au hasard dans la population : la valeur  $(p - p_0)$  qui en découle est une probabilité individuelle moyenne, donc  $(\pi - \pi_0)$  est un RIT moyen.

Les prévisions ne valent que pour une population semblable à la population d'origine des groupes, que pour l'affection considérée, que pour les mêmes conditions d'observation. Le RIT est évalué, le plus souvent, soit pour la vie entière, soit en moyenne annuelle.

La constante  $\chi$  est donc le RIT moyen supplémentaire de contracter l'affection considérée, par rem, soit pour la vie entière, soit ramené à l'année. Sa valeur est déterminée pour chacune des affections susceptibles d'être mises en cause (par effet somatique, par effet génétique).

Jusqu'ici, chacun pour ses besoins forge des dénominations particulières, cela ne va pas sans inconvénients pour interpréter les valeurs obtenues.

### 2.5 - Quelques dénominations usitées

On trouve des définitions éparses dans le rapport de 1964 du comité scientifique de l'ONU<sup>5</sup> (UNSCEAR) et dans la publication 8 de la CIRP<sup>6</sup>. Le numérateur de la constante  $\chi$ , non explicitée, est appelée le "risque estimé", par rem (ou par rad), par million de personnes, soit pour la vie entière, soit par an. En 1972 l'UNSCEAR<sup>7</sup> l'appelle "coefficient de régression", ce qui a l'avantage d'en montrer la nature.

Dolphin et Marley<sup>8</sup> donnent une expression que je peux traduire, avec mes notations :

$$v = \chi N D , \quad (4)$$

où  $(\pi - \pi_0) = \nu/N$ . Ils appellent  $\chi N$  le "coefficient de risque" qui, disent-ils, est exprimé habituellement en nombre de cas (pour l'affection considérée) par million d'homme-rads ( $ND = 10^6$ ), pour la vie entière.

Enfin, alors que la CIRP<sup>6</sup> définit le "risque relatif" par le quotient :  $(\pi - \pi_0)/p_0$ , l'UNSCEAR<sup>7</sup> le définit par :  $(p - p_0)/p_0$ .

A titre d'exemple, pour souligner, au besoin, la nécessité d'une codification de notre langue dans notre spécialité, je vais passer en revue les expressions usitées par une même école, celle d'Alice Stewart. Elle définit d'abord un "risque relatif" (1958)<sup>9</sup>, différent des précédents, puis un "risque absolu" (1970)<sup>10</sup>; enfin Kneale (1971)<sup>11</sup> a introduit un "risque relatif ajouté". Entre temps, la CIRP<sup>12</sup> avait rapporté des valeurs de la "proportion de cas attribuables à l'irradiation" (selon Stewart, 1968).

### 3 - Domage collectif

On doit, logiquement, appeler le dommage calculé à partir du RIT<sup>2</sup> : dommage collectif théorique (DCT). Il se déduit simplement de l'expression (3) :

$$(DCT) = \chi ND, \quad (5)$$

où N représente l'effectif de la population irradiée par l'industrie nucléaire; ou encore, si les membres de cette population sont différemment irradiés :

$$(DCT) = \chi (\sum D_i), \quad (6)$$

où  $\sum D_i$  est la somme des équivalents de dose individuels.

Cette dernière formule, tout à fait générale, s'applique aussi bien à la prévision d'effets somatiques différés qu'à celle d'effets génétiques à la première génération; des valeurs de  $\chi$  ont été proposées<sup>6</sup>. Elles dépendent de l'effet somatique considéré. Une seule suffit pour l'ensemble des effets génétiques traduits, suivant H. J. Muller<sup>13</sup>, en "morts génétiques". Une formule semblable peut aussi s'appliquer aux générations lointaines : sous irradiation constante (D conserve la même valeur à chaque génération) la théorie montre qu'un nouvel équilibre finit par être atteint; les hypothèses de calcul supposent, soit des effets antagonistes de "pressions" de mutation ( $\chi$  a même valeur à chaque génération) et de sélection, soit l'existence d'une "dose doublante". Quelques publications donnent des résultats de calcul de DCT à l'équilibre<sup>14</sup>, ou  $(DCT)_\infty$ ; elles inquiètent.

### 4 - Discussion

Quel qu'il soit, un risque doit d'abord être bien défini, puis évalué par l'hygiéniste qui étudie, s'il y a lieu, à combien le réduire pour le rendre acceptable. C'est au législateur de prescrire les mesures nécessaires, compte tenu des considérations objectives de l'hygiéniste, et aussi, certainement, des réactions subjectives des intéressés.

En ce qui concerne l'énergie nucléaire, l'hygiéniste a innové. On sait que les recommandations de la CIRP<sup>1</sup> indiquent, pour les différents groupes considérés dans la population, des limites d'irradiation d'autant plus basses que leurs effectifs sont plus élevés (c'est logique: voir formule 5 ci-dessus). Vouloir prévoir ce que donneraient de telles limites, l'hygiéniste a calculé les principaux DCT, y compris le  $(DCT)_\infty$ . Les résultats ont permis à la CIRP de juger

que les valeurs trouvées étaient bien "admissibles", ou encore "l'enveloppe du risque, acceptable". Logique, conformément aux conséquences prévisibles suivant son hypothèse de proportionnalité, elle a aussi jugé prudent de prescrire que l'irradiation des personnes soit aussi réduite que possible. Cependant elle n'ignore pas que, pour obtenir un effet déterminé par l'irradiation il faut donner, d'abord à d, puis à D, une valeur supérieure au seuil correspondant à l'effet considéré. Les valeurs de ces seuils sont inconnues pour la plupart des effets différés, à moins qu'ils ne soient très élevés, comme, par exemple, pour la cancérogénèse de la peau par radiothérapie<sup>15,16</sup>, ou du squelette contaminé par le radium 226<sup>17</sup>; ou encore pour l'obtention de mutants, dès la première génération, par irradiation de souris femelles<sup>18,19</sup>. Sur des générations successives de rongeurs, malgré de fortes irradiations, le  $(DCT)_{\infty}$  demeure nul<sup>20</sup>.

Il est même possible d'obtenir l'inversion de certains effets par diminution progressive de d (à D constant) ou de D (à d constant); par exemple, de façon spectaculaire pour la longévité des rats mâles<sup>21</sup> (augmentée d'un tiers par 1000 rems en un an); et même, sans doute, pour une affection maligne : le lymphome de la souris<sup>22,23</sup>.

On comprend ainsi que des membres autorisés de la CIRP déclarent nettement qu'elle n'a aucune raison de publier des recommandations plus restrictives que les dernières<sup>1</sup>.

Cependant, dans l'esprit du public, l'irradiation, traduite en leucémies et en malformations congénitales, frappe essentiellement les enfants. Et personne n'admet d'ajouter un tel risque à son compte, aussi faible soit-il. Et des personnes souvent bien intentionnées, des personnalités même, croyant savoir et censées savoir, mais n'ayant pas su interpréter les textes de la CIRP, y puisent des arguments pour alimenter leur propre inquiétude et pour la propager.

Dans les pays intéressés le législateur a calqué sa réglementation sur les recommandations de la CIRP. Voyant que la perspective de la multiplication rapide de centrales nucléaires provoque des réactions de défense, lorsque ce n'est déjà fait<sup>24</sup>, il se sent disposé à prescrire des mesures restrictives. L'industriel est prêt à le suivre, voire à le précéder. L'inquiétude du public les rend perplexes, c'est le moins que l'on puisse dire : ils ne sauraient négliger les facteurs subjectifs.

Demain, appelé à rechercher a posteriori une relation de cause à effet, le juriste aura bien des difficultés pour disculper la centrale nucléaire tant que subsistera l'hypothèse d'une relation de proportionnalité. Il faudrait lui en donner les moyens; je l'avais proposé il y a une dizaine d'années<sup>25</sup>.

## 5 - Conclusion

La CIRP est sereine, mais l'interprétation de ses textes, difficile. C'est à nous, spécialistes de radioprotection, de rassurer en informant. Commençons par réviser notre nomenclature spécialisée dans un sens qui facilite cette tâche. J'ai proposé de présenter cette communication dans cette intention.

Dans les textes de cette commission on trouve les DCT sous la dénomination "enveloppe de risque". Une telle dénomination, a fortiori, si on lui associe des valeurs numériques, provoque des réactions subjectives qui rendent bien malaisée toute information objective. Aussi, en terminant, me permettrai-je de souhaiter vivement que la CIRP désavoue son hypothèse de proportionnalité.

## Bibliographie

- 1 - ICRP, Recom. adopted Sept. 9, 1958, Pergamon, Paris (1959).
- 2 - Delpla M. et Vignes S., Tendance en radioprotection, SFRP, Bordeaux (1972) 19-43.
- 3 - Delpla M., XVII<sup>e</sup> Congrès int. Médecine Travail, Buenos Aires (1972) à paraître.
- 4 - Delpla M., III<sup>e</sup> Congrès régional radioprotection, Jérusalem (1973) à paraître.
- 5 - UNSCEAR, Supplement n° 14 (A/5814), Nations Unies, New York (1964).
- 6 - ICRP, Publication 8, Pergamon, Paris (1966).
- 7 - UNSCEAR, Supplement n° 25 (A/8725) United Nations, New York (1972).
- 8 - Dolphin G.W. and Marley W.G., AHSB(RP)R 93, Harwell, UKAEA (1969).
- 9 - Stewart A., Webb J. and Hewitt D., Brit. Med. J. (1958) 1495-1508.
- 10 - Stewart A. and Kneale G.W., Lancet, 6 (1970) 1185-1188.
- 11 - Kneale G.W., Biometrics (1971) 563-590.
- 12 - ICRP, Publication 14, Pergamon, Paris (1969).
- 13 - Muller H.J., Am. J. Public Health 54 (1964) 42-50.
- 14 - Morgan K.Z., Hearing US Senate, Gov. Print. Of. (1968) 31-64.
- 15 - Maisin J., Symposium Euratom, Bruxelles (1961) 285-296.
- 16 - Rowell N.R., Brit. J. Radiol. 45 (1972) 610-620.
- 17 - Mays C.W., Dougherty Th. F., Taylor G.N., Stover B.J., Jee W.S., Christensen W.R., Dugherty J.H., Stevens W., and Nabors Ch. J., COO-119-242, University of Utah (1970) 385-409.
- 18 - Russell W.L., Nucleonics 23 (1965) 53 - 56, 62.
- 19 - Russell W.L., A/CONF.49/P/677, UN, New York, 13 (1972) 487-500.
- 20 - Green E.L., An. Rev. Genetics 2 (1968) 87-120.
- 21 - Carlson L.D. and Jackson B.H., Rad. Res. 11 (1959) 509-519.
- 22 - Kaplan H.S., J. Nat. Cancer Inst. 10 (1949) 267-279.
- 23 - Mewissen D.J., CONF-691212; US Dep. Com., Springfield (1971) 413-424.
- 24 - Dir. Regul. Stand., Proposed rule making action, USAEC (1973).
- 25 - Delpla M., lettre Direction Protection sanitaire, Euratom (26/12/63).

AN EVALUATION OF NUCLEAR GAS STIMULATION IN TERMS  
OF POTENTIAL RADIATION EXPOSURE TO THE PUBLIC<sup>a,b</sup>

P. S. Rohwer, C. J. Barton, R. E. Moore, and S. V. Kaye  
Environmental Sciences Division  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830

Abstract

Experience gained from Projects Gasbuggy and Rulison and their follow-up studies indicates that natural gas produced from a nuclearly stimulated well field will contain small amounts of man-made radioactivity as it leaves the gas processing plant and enters commercial distribution channels. Individual and population doses have been estimated for hypothetical uses of such gas. For example, it is estimated that residential use of nuclearly stimulated gas in unvented cook stoves would result in an average total-body dose to the house occupants of approximately 0.2 millirem/year. Radon concentrations measured in natural gas at various locations in the United States average approximately 20 pCi per liter. Assuming this concentration of radon in the unvented cooking case mentioned, the lung dose is estimated to be 1.5 millirems per year. All of the dose estimates discussed are used to give perspective to the additional radiation exposure of the public which could occur due to use of gas from nuclearly stimulated wells. Both somatic risk and genetic risk are considered in the assessment of relative hazard. Comparisons are made with other risks encountered in the normal activities of life in the United States. The studies summarized show that the radiological impact of either domestic or industrial use of the gas can be small.

Introduction

The U.S. Atomic Energy Commission's Plowshare Program is almost exclusively devoted at present to the development of the nuclear gas stimulation concept. Two experiments involving detonation of single nuclear explosives in low permeability rock formations, Gasbuggy and Rulison, have been conducted to date. Results of these experiments are considered very encouraging by the AEC and the industrial sponsors. Rio Blanco, the third experiment, involved the use of three nuclear devices in one well hole to stimulate gas production in thicker rock formations than would be possible with only one explosive. The explosives were detonated simultaneously on May 17, 1973.

The Health Physics and Environmental Sciences Divisions of the Oak Ridge National Laboratory have been investigating the radiological impact of potential uses of natural gas from wells stimulated with nuclear explosives. This paper summarizes the more important results of these studies.

Radionuclide Inventory

Radionuclides found in gas produced from the completed experiments, Gasbuggy and Rulison, were  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{37}\text{Ar}$ ,  $^{39}\text{Ar}$ ,  $^{85}\text{Kr}$ , and  $^{203}\text{Hg}$ . of the radionuclides

<sup>a</sup>For presentation at the Third International Radiological Protection Association Meeting, Washington, D.C., September 1973.

<sup>b</sup>Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.



found,  $^3\text{H}$ ,  $^{14}\text{C}$ , and  $^{85}\text{Kr}$  have been studied in greatest detail because they contribute over 99% of the potential dose equivalent\* at the radionuclide concentrations observed, and at those projected for future wells (see Table 1).

Table 1. Relative Percentage of Total Estimated Dose from Man-made Radionuclides in Nuclearly Stimulated Natural Gas

Radionuclide	Projected Average First Year Concentration (pCi/cm <sup>3</sup> )	Projected Average Lifetime Concentration (pCi/cm <sup>3</sup> )	Percentage of Estimated Total Somatic Dose <sup>a</sup>	Percentage of Estimated Total Genetic Dose <sup>b</sup>
$^3\text{H}$	<20	1.0	60	93
$^{14}\text{C}$	<1	0.02	0.3	0.3
$^{85}\text{Kr}$	65	3.3	39	6
All others			<<1	<<1

<sup>a</sup>Dose to total body.

<sup>b</sup>Dose to gonads.

The tritium concentration used in this paper, 1 pCi/cm<sup>3</sup>, is the value projected for the average concentration during the lifetime production of future wells.<sup>1</sup> The concentrations for  $^{14}\text{C}$  and  $^{85}\text{Kr}$  are scaled to the tritium value on the basis of literature values or fission and activation yield.<sup>2-5</sup> The tritium inventory is distributed in the H<sub>2</sub>O-H<sub>2</sub>-hydrocarbon system with less than 25% of the total production in the form of hydrocarbons or hydrogen. The remaining 25% appears as tritiated water (HTO) which would normally be removed at the wellhead. Carbon dioxide is also normally removed from natural gas before it enters commercial distribution channels. The tritium and  $^{14}\text{C}$  present as hydrocarbons will be released as HTO and  $^{14}\text{CO}_2$ , respectively, when the gas is burned.

#### Estimation of Dose to Man

There are numerous pathways through which radionuclides present in gas from nuclearly stimulated wells may cause radiation exposure to man. Our studies have indicated that  $^3\text{H}$  and  $^{85}\text{Kr}$  are the critical radionuclides and that exposure to combustion products from unvented home usage of natural gas containing radionuclides is the critical exposure pathway.

We can calculate the dose to an individual resulting from combustion of gas in unvented home appliances and heaters based on the projected radionuclide concentrations given in Table 1. For a residence (93 m<sup>2</sup> floor space, 227 m<sup>3</sup> volume) of normal construction with one air change per hour ventilation rate, we estimate the following potential total-body doses (mrem/year) for various unvented domestic uses: cooking, 0.16; water heater, 0.38; refrigerator, 0.20; and heating (5000-degree days), 2.8. The maximum doses estimated for an individual at the projected radionuclide concentrations and assumed exposure conditions with no venting of appliances or heaters is less than 20 mrem/year to the total body. Most cities and states in the United States now require venting of all heaters and appliances except those used for cooking. If this requirement is implemented, the estimated average dose to an individual in the exposed population would be slightly less than 0.2 mrem/year for gas containing the average

\*Dose equivalent (rem) = Absorbed Dose (rads) x modifying factors. For the sake of convenience, "dose" will be used hereafter instead of "dose equivalent."

lifetime radionuclide concentrations listed in Table 1. This estimated dose includes a contribution from unvented cooking (0.76 m<sup>3</sup> of gas per day, United States average) plus an average atmospheric contribution from all gas used in the area. This latter contribution was shown in a study of the Los Angeles Basin to be less than 10% of the calculated individual dose from unvented cooking.<sup>6</sup> Even this small calculated average individual dose (0.2 mrem/year) would deliver a potential 200 man-rems per year to each million people exposed. The man-rem dose estimate is obtained by summation of all individual doses within the exposed population.

Use of nuclearly stimulated natural gas in power stations has been suggested as an alternative to residential use. Power station use of nuclearly stimulated gas was given consideration in the Rulison study<sup>7,8</sup> by assuming that the Cherokee electricity generating plant located in the Denver, Colorado, metropolitan area burned 2.66 x 10<sup>6</sup> m<sup>3</sup> of gas per day (9.72 x 10<sup>6</sup> m<sup>3</sup> per year) contaminated with the projected average lifetime radionuclide concentrations listed in Table 1. The estimated dose to the population (1,500,000) in the Denver area due to power station use of that quantity of gas is 0.32 man-rem. The maximum individual dose estimate for the entire area is 0.006 millirem/year.

The same 9.72 x 10<sup>6</sup> m<sup>3</sup> of gas would supply 174,000 households for 1 year under the following assumptions: unvented cooking (0.76 m<sup>3</sup>/day), vented water heater (1.8 m<sup>3</sup>/day), and vented heating for 5000-degree days per year (13.4 m<sup>3</sup>/day). If each household is assumed to have 3.5 residents, the total number of persons exposed is 610,000, approximately one-third of the total population in the Denver metropolitan area. Then the comparable estimated population dose due to residential gas use is 110 man-rems for 9.72 x 10<sup>6</sup> m<sup>3</sup> of gas having the projected lifetime radionuclide concentrations. Thus, under the conditions specified, the population dose estimate for household use of the nuclearly stimulated gas is nearly 350 times that for power station use.

#### Assessment of the Estimated Dose to Man

Assessment of the dose estimate projected for use of gas from nuclearly stimulated fields can vary in form and complexity. We believe that the assessment should begin with the recognition that natural gas contains natural radioactivity and that one result of nuclear stimulation is an incremental change in the total radioactivity concentration to which gas users are exposed.

It has been known for nearly 70 years that natural gas contains a radioactive species, radon. Samples supplied by gas transmission companies were analyzed in 1972 and 1973 by scientists in four institutions to provide data on radon concentration in gas being supplied to several metropolitan areas in the United States.<sup>9</sup> The average value (20 pCi/liter) for all sample locations is used in our dose estimations.

One exposure situation that we consider for radon daughters produced by decay of radon in natural gas is the same as that assumed in the previously described studies.<sup>7</sup> An unvented kitchen range using 0.76 m<sup>3</sup> of gas per day was assumed to be located in a house having a volume of 227 m<sup>3</sup>. We calculated the concentration of radon daughters in the home for air change rates varying from 0.25 to 2.0 changes per hour. We then estimated doses to the bronchial epithelium from radon daughters resulting from decay of radon introduced with the natural gas and compared these doses with those from an assumed concentration of 0.13 pCi/liter of radon (the average concentration from a number of radon measurements in the United States) and each of its daughters in ventilation air. The estimated dose rate to the bronchial epithelium due to radon and its daughters in the ventilation air was 1300 to 1400 millirem per year. Additional estimated

dose to the bronchial epithelium due to the radon (20 pCi/liter) present in natural gas ranges from 90 mrem/year, for 0.25 air change per hour, to 5 mrem/year, for two air changes per hour. At most, the estimated dose increase due to radon present in natural gas is less than 7%, considering only the two sources of radon. The relative importance of this natural activity in the gas is reduced still further if one considers the daughter activity due to the decay of radon and thoron emanating from home building materials which in some situations exceeds our assumed concentration in ventilation air by a factor of 10. It appears likely, therefore, that the dose which can be attributed to the radon in natural gas is small (<1%) compared to the total dose received in the home from all sources of airborne radioactivity. These localized radon-radon daughter dose estimates are for a limited tissue volume: the basal cells of the bronchial epithelium, which are assumed to be the critical tissue. It has been estimated that the corresponding dose to the total lung mass (1000g) is an order of magnitude lower.<sup>10</sup> The dose to the total lung due to radon in the gas (1.5 millirem/year, assuming one air change per hour) is more suitable for comparison with the total-body estimates obtained for the man-made radioactivity in nuclearly stimulated gas. The comparison indicates that the projected concentrations of man-made radioactivity will contribute a dose which is approximately 12% of the dose due to radon in the gas. Local conditions may, however, alter this percentage significantly.

Another possible assessment, and one which will be required, is comparison of the dose estimate with applicable radiation safety standards. At this time, however, there are no standards which are specific to the use of nuclearly stimulated gas. The Federal Radiation Council (FRC) has established 170 millirem/year as the upper limit for the average total-body dose to a suitable sample of an exposed population group for radiation from all sources exclusive of natural background and medical exposures.<sup>11</sup> However, this single source of exposure must not be permitted to take up a disproportionate share of the 170 millirem/year total. The estimated average total-body dose for the population group expected to be exposed via home use of nuclearly stimulated gas is a small fraction (0.1%) of the dose limit of 170 millirem/year.

Our estimate of dose to the total body from nuclearly stimulated gas (0.2 millirem/year) may also be put in perspective by comparing it with dose estimates (millirem/year to the total body) for other sources of radiation received by members of the public: natural background radiation, 130; medical diagnostic X-rays, 110; nuclear weapons fallout, 2; consumer devices, 2; industrial uses of radiation, <1.<sup>12</sup>

A hypothetical assessment of the projected dose may be obtained by estimating the risks which the exposure represents in terms of additional deaths, additional death equivalents due to radiation-induced life span shortening, and additional genetic deaths. A total risk estimate was obtained by summing all three types in spite of the recognized inherent difficulties in combining somatic and genetic insults whose manifestations may differ so greatly. The factors used to convert estimates of radiation dose into estimates of risk are those suggested by the ICRP.<sup>13,14</sup> Those factors are based on the conservative assumption that there is a linear relationship between dose and effect. The estimates of additional deaths calculated here are believed to be upper limits of risk for the low dose levels considered. The actual risk in fact may be zero, for at such low doses, there is no practical method to reliably determine the actual risk involved. The risk estimated for the projected gas usage is compared in Table 2 with similar estimates of risk for other sources of radiation exposure of the public.

The population dose (man-rem) estimates for the hypothesized gas uses may also be assessed, but to a lesser extent, as there have been no official numerical limits established with which the population dose estimates can be compared. We

Table 2. Comparison of Estimated Deaths Due to Man-made Radioactivity in Nuclearly Stimulated Natural Gas with Similar Estimates for Other Sources of Radiation Exposure of the Public in the United States

Sources of Exposure	Estimated Deaths per Million Individuals Exposed <sup>a</sup>
Natural background radiation <sup>b</sup>	17
Radioactivity in natural gas <sup>c</sup>	
Natural (radon + daughters)	0.3
Man-made	0.03
Other man-made sources of radiation	
Medical diagnostic x-rays	20
Fallout from nuclear weapons	0.4
Consumer devices	0.4
Industrial uses of radiation	<0.2
Power reactors	<0.2

<sup>a</sup>Obtained by summing estimated somatic and genetic effects; therefore, some of these estimated deaths will occur among the exposed individuals or the first generation of their offspring, but a large majority (over 80%) will occur in succeeding generations.

<sup>b</sup>0.1 rem per year.

<sup>c</sup>Based on projected radionuclide concentrations used in this study.

have shown that the man-rem dose to the local population is sensitive to the manner of gas usage. The population dose in the Denver area due to background radiation (~200 mrem per person) is nearly  $3.0 \times 10^5$  man-rems per year, while that estimated for residential gas use (110 man-rems) is 0.037% of the background dose. Dose to the global population is another point to be considered for comparison. Based on dose conversion factors presented in a recent report of the United Nations Committee on the Effects of Atomic Radiation (UNSCEAR),<sup>15</sup> the estimated infinite dose (integrated over infinite time) to the population of the northern hemisphere due to the release of the man-made radioactivity in that volume of gas is approximately 840 man-rems. Nearly all (99%) of that infinite dose is contributed by <sup>14</sup>C due to its long radioactive half-life (5730 years). This estimated dose must be added to the estimated local population dose in assessing the total population dose incurred as a result of the release.

#### Summary and Conclusions

The radiological impacts of hypothetical uses of nuclearly stimulated gas for domestic and industrial purposes were studied. The critical exposure pathway was determined to be the release of combustion products from unvented appliances in the home. The estimated average total body dose from man-made radioactivity for that pathway is 0.2 millirem per year of gas use. The estimated lung dose due to natural radioactivity (radon) in the gas is 1.5 millirem/year. The critical man-made radionuclides are <sup>3</sup>H, <sup>14</sup>C, and <sup>85</sup>Kr. The largest fraction of the local population dose received via the critical pathway is due to <sup>3</sup>H, followed by <sup>85</sup>Kr. Carbon-14 is of importance as the major contributor to the infinite population dose estimated for the northern hemisphere, with the total dose to the local population being very dependent on the manner of gas use. The radiological impact of the hypothesized gas use was assessed in terms of dose and in terms of risk, in the interest of incorporating radiological impact of gas use as an integral part of the cost-benefit analysis for the development of nuclear gas stimulation technology. The results of the assessment indicate that the radiological impact would be very small. Although the dose and risk estimates obtained in this study are small, the possible exposures still must be scrutinized to achieve the lowest practicable local and global doses.

### References

1. Barton, C. J., D. G. Jacobs, M. J. Kelly, and E. G. Struxness, "Radiological Considerations in the Use of Natural Gas from Nuclearly Stimulated Wells," Nuclear Tech. 11, 335 (1971).
2. Kirk, W. R., Krypton-85, A Review of the Literature and an Analysis of Radiation Hazards, U.S. Environmental Protection Agency, Office of Research and Monitoring, Washington, D. C. 20460 (January 1972).
3. Teller, E., et al., The Constructive Use of Nuclear Explosives, p. 90, McGraw-Hill Book Company, New York, 1965.
4. Dudey, N. D., Review of Low-Mass Atom Production in Fast Reactors, USAEC Report ANL-7434 (1968).
5. Green, J. B., Jr., and R. M. Lessler, Reduction of Tritium from Underground Nuclear Explosives, USAEC Report UCRL-73258 (September 1971).
6. Jacobs, D. G., et al., Theoretical Evaluation of Consumer Products from Project Gasbuggy, Final Report, Phase II: Hypothetical Population Exposures Outside San Juan Basin, USAEC Report ORNL-4748 (February 1972).
7. Barton, C. J., R. E. Moore, and S. A. Hanna, Quarterly Progress Report on Radiological Safety of Peaceful Uses of Nuclear Explosives: Hypothetical Exposures to Rulison Gas, USAEC Report ORNL-TM-3601 (October 1971).
8. Moore, R. E., and C. J. Barton, Progress Report on Radiological Safety of Peaceful Uses of Nuclear Explosives: Dose Estimations for the Hypothetical Uses of Nuclearly Stimulated Natural Gas in the Cherokee Electricity Generating Plant, USAEC Report ORNL-TM-4026 (in press).
9. Barton, C. J., R. E. Moore, and P. S. Rohwer, Contribution of Radon in Natural Gas to the Natural Radioactivity in Homes, USAEC Report ORNL-TM-4154 (April 1973).
10. Holleman, D. F., Radiation Dosimetry for the Respiratory Tract of Uranium Miners, Colorado State University Report COO-1500-12 (December 1968).
11. Federal Radiation Council Report No. 1, Background Material for the Development of Radiation Protection Standards (May 1960).
12. Kelly, M. J., P. S. Rohwer, C. J. Barton, and E. G. Struxness, "Relative Risks from Radionuclides Found in Nuclearly Stimulated Natural Gas," IAEA PNE Panel, November 27-30, 1972, Vienna, Austria.
13. International Commission on Radiological Protection, The Evaluation of Risks from Radiation, ICRP Publication 8, Pergamon Press, New York (1966).
14. International Commission on Radiological Protection, Radiosensitivity and Spatial Distribution of Dose, ICRP Publication 14, Pergamon Press, New York (1969).
15. United Nations, General Assembly, A Report of the United Nations Scientific Committee on the Effects of Atomic Radiation, Vol. 1, Levels, New York (1972).

GENETIC DOSE LIMIT FOR GENERAL POPULATION, DOSE LIMIT AND DERIVED CONCENTRATION GUIDES FOR MEMBERS OF THE PUBLIC COMPULSIVE ACTION GUIDES FOR EMERGENCY SITUATIONS. A proposal for México, based on data available on Mexican people.

R.M. de Nulman.  
 Environmental Radiactivity Section.  
 Instituto Nacional de Energía Nuclear  
 México, D. F., México.

C. Archundia de la Rosa.  
 Centro de Estudios Nucleares  
 Universidad Nacional Autónoma de México  
 México, D.F. México.

Using methods recommended by ICRP & UNSCEAR, the mean age of childbearing and genetic dose limit was calculated for mexican population. Data for mexican population when available, complemented with internationally recommended data are used to derive concentration guides for several radionuclides in various environmental media. Derived concentration guides in sea water are calculated applying the specific activity concept. Emergency projected doses demanding action to be taken are proposed. Data lacking is emphasized in order to encourage further studies on habits, critical pathways and transfer factors through food chain for radionuclides in Mexico.

Introduction

Mexico is a developing country with great energetic needs and is now on its way to start using nuclear energy for power production. On this behalf, Uranium ore mining and milling and fuel fabrication is being considered on an industrial scale.

For radiation protection purposes the recommendations of the International Commission on Radiological Protection are now being applied, and regulations are to be issued in a near future, as needed, taking into account habits and characteristics of mexican population.

Genetic Dose Limit

ICRP<sup>1</sup> recommends a genetic dose limit of 5 rems. In the UNSCEAR Report to the General Assembly<sup>2</sup> there is a detailed discussion of the genetically significant dose, and applying the method outlined with data for Mexican population<sup>3,4,5</sup>, a mean age of childbearing of 35 years was obtained, this gives an annual genetically significant dose limit of 0.144 man rem per 10<sup>6</sup> inhabitants. Apportionment of this dose, on applying the recommendations of K.Z. Morgan<sup>6</sup> and rounding off numbers is stated in Table I.

T A B L E 1			
GENETIC DOSE LIMIT FOR GENERAL POPULATION: 5 rems in 35 years			
ANNUAL GENETICALLY SIGNIFICANT DOSE: 0.14 x 10 <sup>6</sup> man rem per 10 <sup>6</sup> inhabitants			
APPORTIONMENT IN MAN REMS PER YEAR 10 <sup>6</sup> INHABITANTS			
Diagnosis	5 x 10 <sup>4</sup>	Other environmental sources	5 x 10 <sup>3</sup>
Therapy	10 <sup>4</sup>	Future Applications and	
Nuclear energy		emergency situation	10 <sup>5</sup>
production	10 <sup>4</sup>	Occupational exposure	10 <sup>4</sup>

It may be noticed that a substantial portion of the genetic dose available is allowed for medical exposure. The genetic dose due to this source tends to increase the benefits being for the present generation whilst the risk will

burden future generations, and so it has to be recorded and included in what would be an acceptable genetic risk from all uses of radiation.

Dose Limits and Concentration Guides

Radionuclides, sources and environmental pathways considered as most important for the transfer to man of radionuclides introduced in the environment, were obtained from the general guidelines for the growth of nuclear industry, as set forward by the Nuclear Energy Institute in its development program<sup>7</sup> and on the nuclear power reactor of Laguna Verde, Veracruz to start operating in 5 or 6 years. By now, there is no experience on pathways for radionuclides from source to man in Mexico, and this approach is a purely theoretical one.

Some studies have been made in Mexico on characteristics and habits of Mexican population<sup>8,9,10,11,12</sup>, this studies were performed for medical or nutritional reasons. They show that a wide difference exists specially on nutritional habits, in different zones of Mexico and even between habits of people living in the same area, but engaged in different economical activities, this does not permit to find the critical pathway for Mexico as a whole, although it is feasible with a well designed survey for a small area, as the one affected by effluents from a nuclear reactor.

Federal Radiation Council<sup>13</sup> has been issuing concentration guides for average population applying a 1/3 safety factor to dose limits set forth by ICRP for individuals in the critical population. Since in Mexico there is also an incomplete knowledge of some data needed, an arbitrary safety factor of 1/4 is used instead. Morphological and physiological parameters obtained from studies on Mexican population<sup>8,11</sup> are summarized in Table II.

PARAMETER	MEN
Body Weight	65 Kg
Daily water intake	1300 ml
Body fluids	39 Kg
Mineral bone	4.6 Kg
Calcium content	975 g
Thyroid	35 g

For other data needed ICRP and other sources were consulted<sup>14,15,16,17,18</sup>,  
<sup>9</sup>Data on food consumption were obtained from Zubiran et. al<sup>20</sup>, Table III.

FOOD	GROSS WEIGHT IN GRAMS CONSUMED BY	
	RURAL POPULATION	URBAN POPULATION
Corn	399	202
Bread and Pasta	25	129
Rice	5	10
Beans	45	45
Meat	47	76
Milk	62	241
Cheese	3	3
Eggs	5	13
Vegetables	81	114
Edible roots	11	20
Fruit	36	72
Sugar	39	77
Fats	13	26
Cacao	3	0
Other	0	12

Table IV shows concentration guides or working limits for average population.

T A B L E IV		
WORKING LIMITS OR CONCENTRATION GUIDES FOR AVERAGE POPULATION IN MEXICO		
RADIONUCLIDE	WORKING LIMIT OR CONCENTRATION GUIDE	IN
$^{226}\text{Ra}$	3 pCi/l	Drinking water
$\text{U}_{\text{nat}}$	3 $\mu\text{g}/\text{m}^3$	air
$^3\text{H}$ (HTO)	$30 \times 10^{-3}$ $\mu\text{Ci}/\text{m}^3$	air
$^4\text{A}$	$3 \times 10^{-2}$ $\mu\text{Ci}/\text{m}^3$	air*
$^{85\text{m}}\text{Kr}$	0.1 $\mu\text{Ci}/\text{m}^3$	air*
$^{85}\text{Kr}$	0.2 $\mu\text{Ci}/\text{m}^3$	air*
$^{87}\text{Kr}$	$2 \times 10^{-2}$ $\mu\text{Ci}/\text{m}^3$	air*
$^{88}\text{Kr} - ^{88}\text{Rb}$	$10^{-2}$ $\mu\text{Ci}/\text{m}^3$	air*
$^{131\text{m}}\text{Xe}$	0.3 $\mu\text{Ci}/\text{m}^3$	air*
$^{133}\text{Xe}$	0.3 $\mu\text{Ci}/\text{m}^3$	air*
$^{135\text{m}}\text{Xe} - ^{135\text{m}}\text{Cs}$	$8 \times 10^{-2}$ $\mu\text{Ci}/\text{m}^3$	air*
$^{131}\text{I}$	30 pCi/ $\text{m}^3$	air
$^{131}\text{I}$	110 pCi/day	Total Diet (6 months child)
$^{131}\text{I}$	1000 pCi/l	cow's milk
		(Through mother to breast fed babies).
$^{137}\text{Cs}$	300 pCi/ $\text{m}^3$	air
	$5 \times 10^3$ pCi/day	Total Diet
	RURAL TYPE DIET	URBAN TYPE DIET
	$1.3 \times 10^4$ pCi/Kg	$2.6 \times 10^4$ pCi/Kg
	$12 \times 10^4$ pCi/Kg	$12 \times 10^4$ pCi/Kg
	$9 \times 10^4$ pCi/l	$2 \times 10^4$ pCi/l
	$11 \times 10^4$ pCi/Kg	$7 \times 10^4$ pCi/Kg
		Corn
		Beans
		Milk
		Meat

\*Safety factor of 1/4 was not used for calculus.

The main food for children's diet from birth to about 2 years of age is milk, but in Mexico most babies are breast fed, and weaning starts between 1 and 2 years of age substituting the mother's milk with corn or/and beans, not with cow's milk, the pathway for radioiodine being from milk and food eaten by the mother to breast fed babies. A survey made at the Instituto de Nutrición by Perez H. et al<sup>21</sup> gives an average of 400 ml milk in the mother's diet and so the derived working level or concentration guide for average individual in general population was calculated, on the basis of iodine transferred by mother to child, using data from Weaver et al<sup>22</sup> for a mother with a milk production similar to the average Mexican mother.

Strontium has a metabolism similar to calcium, but is discriminated against through its pathway from environment to man, the ratio of Sr 90 to calcium in bone needed to obtain a dose of 0.75 rems/year to bone, for individuals in the average population applying the method outlined by UNSCEAR<sup>2</sup>: is  $4.5 \text{ m rad y}^{-1}$  per pCi (gCa)<sup>-1</sup>, and dose in rems is obtained multiplying by the "relative damage factor" 5, for Sr 90 in bone, giving  $23 \text{ mrem y}^{-1}$  per pCi (gCa)<sup>-1</sup> and 33 pCi (gCa)<sup>-1</sup> for 0.75 rem y<sup>-1</sup>. UNSCEAR<sup>2</sup> gives a transfer factor of 0.12 diet to bone and 275 pCi <sup>90</sup>Sr/gCa in diet produces 33 pCi <sup>90</sup>Sr/gCa in bone.

A high proportion of calcium is obtained in Mexican diet through mineral calcium added to corn (150 mg of Ca/100g corn<sup>23</sup>), in making much of the food based on corn and specially "tortillas" which are used instead of bread by most of Mexican population. This calcium has a negligible contribution to <sup>90</sup>Sr contamination and in considering a uniform contamination of the biosphere, after UNSCEAR and C. L. Comar<sup>2,24</sup> and calcium content in food items commonly consumed in Mexico, transfer factors are shown in Table V. Doses produced by Sr<sup>89</sup> for a long period of time are 25 times lower per pCi/gCa than dose produced by <sup>90</sup>Sr, and the average concentration guides or working limits for <sup>89</sup>Sr and <sup>90</sup>Sr in food regardless of the actual quantity consumed are shown in Table VI.



T A B L E V

RELATIONS BETWEEN \*Sr-Ca OF VEGETATION (100 \*Sr-100Ca) and \*Sr-Ca OF MAN

	% Ca IN DIET		PLANT PRODUCT	DIET BODY	*Sr PER 100Ca IN BODY	
	RURAL	URBAN			RURAL	URBAN
Cereals	5	7	1	0.12	0.6	0.84
Other plants	15	19	1	0.12	1.8	2.28
Dairy products	10	35	0.12	0.12	0.14	0.5
Mineral Ca	67	36	0	0	0	0
Total					2.6	3.7

\*Sr RADIOSTRONTIUM

T A B L E VI

RADIOSTRONTIUM DERIVED WORKING LIMITS OR CONCENTRATION GUIDES FOR AVERAGE POPULATION IN MEXICO

	URBAN DIET		RURAL DIET	
	<sup>90</sup> Sr	<sup>89</sup> Sr	<sup>90</sup> Sr	<sup>89</sup> Sr
Corn	100 pCi/Kg	2x10 <sup>3</sup> pCi/Kg	130 pCi/Kg	3.5x10 <sup>3</sup> pCi/Kg
Milk	130 pCi/l	3.3x10 <sup>3</sup> pCi/l	180 pCi/l	4.5x10 <sup>3</sup> pCi/l
Beans	2x10 <sup>3</sup> pCi/Kg	5x10 <sup>4</sup> pCi/Kg	3x10 <sup>3</sup> pCi/Kg	7.5x10 <sup>4</sup> pCi/Kg
Vegetables	10 <sup>3</sup> pCi/g Ca	2.5x10 <sup>4</sup> pCi/gCa	1.3x10 <sup>3</sup> pCi/gCa	

Concentration Guides in Sea Water. The marine food chain to man is not well known in Mexico's coastal waters and the specific activity approach is likely to be the best under this circumstances. Following Kaye S.V. and Nelson D.J.<sup>25</sup> concentration guides in sea water for some radionuclides of interest are obtained using the following:

$$\text{Concentration Guide in sea water } \mu\text{Ci/l} = \frac{2.8 \times 10^{-3} Y_1 W}{\Sigma EF(RBE)n Y_2} \left[ 1 + \frac{T_b}{T_r} \right] \left[ \frac{1}{1 - e^{-\frac{0.693t}{T_e}}} \right]$$

Where:  $Y_1$  = concentration of stable element in sea water ( $\mu\text{g/l}$ );  $Y_2$  = concentration of stable element in organ of reference ( $\mu\text{g/g}$ );  $W$  = weekly dose limit (annual dose limit/52);  $\Sigma EF(RBE)n$  = effective energy in MeV per disintegration;  $T_b$  = biological half-life in days;  $T_e$  = effective half-life in years;  $t = 70$  years.

The dose limit used was not affected by the 1/4 safety factor since there are already safety factors included, in omitting the effective half-life and growth factors for every link in the pathway from sea water to man. Data used for calculation were obtained from literature<sup>1, 15, 26</sup>.

For radioisotopes with GI tract as critical organ, since the exposure is due to the absolute concentration of radionuclide in the tract, instead of using the specific activity approach, the method outlined by Aten<sup>27</sup> is used:

$$\text{Concentration Guide } (\mu\text{Ci/l}) = \frac{M PC_w \times 2200}{\left[ (0.13 \times F_c \times K_c) + (0.13 \times P_f \times K_p) \right] \times 10}$$

Where:  $MPC_w$  = maximum permissible concentration in drinking water for occupationally exposed personnel (168 h)  $\mu\text{Ci/ml}^{25}$ ;  $F_c$  = Concentration factor for shrimps<sup>26</sup>;  $K_c$  = Shrimp fraction in marine food intake (1, 0.5, 0);  $P_f$  = Concentration factor for fish<sup>26</sup>;  $K_p$  = Fish fraction in marine food intake (1, 0.5, 0); 0.13 = Marine food daily consumption in  $\text{Kg}^{12}$ .

A survey made by the Instituto de Nutrición, on food intake, of a fishing community, Alvarado, in the same state where the power reactor site is, although not in the same area, is used for calculus, food consumed were fish and shrimps, but no mention is made on the proportion of each, since both were grouped together for survey purposes. Since concentration factor from sea water to edible product are quite different for shrimps and fish, derived working levels or concentration guides were calculated considering fish 100%, shrimps 100% and 50% consumption of each. A factor of 1/10 for individuals in the critical population and 1/4 safety factor are included.

The concentration guides are presented in Table VII.

T A B L E V I I					
DERIVED WORKING LIMITS OR CONCENTRATION GUIDES FOR SOME RADIONUCLIDES IN SEA WATER					
RADIO-NUCLIDE	CRITICAL ORGAN	DERIVED WORKING LIMIT OR CONCENTRATION GUIDE IN SEA WATER			
		SPECIFIC ACTIVITY METHOD pCi/l	SHRIMP 100%	FISH 100%	SHRIMP 50% FISH 50%
<sup>54</sup> Mn	GI Tract		2.2x10 <sup>2</sup>	5.3x10 <sup>3</sup>	4.3x10 <sup>2</sup>
	Liver	6x10 <sup>2</sup>			
<sup>55</sup> Fe	Spleen	1.7x10 <sup>2</sup>			
<sup>59</sup> Fe	GI Tract		10 <sup>2</sup>	1.6x10 <sup>2</sup>	1.3x10 <sup>2</sup>
	Spleen	31			
<sup>58</sup> Co	GI Tract		8.5x10 <sup>2</sup>	4.2x10 <sup>4</sup>	1.7x10 <sup>3</sup>
	Whole body	4.6x10 <sup>2</sup>			
<sup>60</sup> Co	GI Tract		4.2x10 <sup>2</sup>	2x10 <sup>4</sup>	8.3x10 <sup>2</sup>
	Whole body	1.7x10 <sup>2</sup>			
<sup>65</sup> Zn	Whole body	1.2x10 <sup>2</sup>			
<sup>87</sup> Sr	Bone	11x10 <sup>6</sup>			
<sup>90</sup> Sr	Bone	4.7x10 <sup>4</sup>			
<sup>91</sup> Y	GI Tract			5x10 <sup>2</sup>	
<sup>131</sup> I	Thyroid	1.1x10 <sup>3</sup>			
<sup>137</sup> Cs	Whole body	8.7x10 <sup>3</sup>			
<sup>137</sup> Cs	Whole body	10 <sup>5</sup>			
<sup>140</sup> Ba	GI Tract			1.6x10 <sup>4</sup>	
	Bone	3x10 <sup>3</sup>			
<sup>144</sup> Ce	GI Tract		2x10 <sup>4</sup>	1.4x10 <sup>5</sup>	3.7x10 <sup>4</sup>

#### Action Levels

In order to set action levels, the social cost together with the expected effectiveness in enforcing the corrective measures has to be balanced against the risk reduced, in this behalf a due study has to be undertaken and each place has to be analyzed in itself, and reviewed as changes happen.

In order for the nuclear industry to include the needed safety measures in design it is considered that people are prepared to move from one state into another and in so doing their risk of accidental death will change, varying from  $2.1 \times 10^{-4}$  in Quintana Roo to  $11.1 \times 10^{-4}$  in Colima<sup>3</sup>, so for individuals in the population, an increase in 10% the previous risk due to accidental death is acceptable, and action levels for whole body irradiation, should not produce significant early effects in the individuals exposed, a limit of 25 rems to whole body for men and 10 rems for women in reproductive age, delivered in a short period of time, and for organ irradiation, the enhanced stochastic cancer risks, should not be higher than 10% the actual risk from accidental death in Mexico, about  $6 \times 10^{-4}$  in 1969. Action levels for whole body and different critical organs are displayed in Table VIII, together with data on risk estimates considered.

T A B L E V I I I			PROJECTED ACTION GUIDES	
	ACTION GUIDE	ENHANCED STOCHASTIC CANCER RISK OF DEATH PER MILLION PEOPLE EXPOSED.		
For whole body				
Women in reproductive age	10 rems			
Men	25 rems			
For Thyroid	35 rems	$3 \times 10^{-6}$ per rem for children <sup>2B</sup>		
		$10^{-6}$ per rem for adults <sup>2B</sup>		
		$1.7 \times 10^{-6}$ per rem for average mexican population*		
For bone	1.5 rem/year	$10^{-5}$ for 0.3-3 rems/year <sup>29</sup>		
For lungs	6 rems	$10^{-5}$ per rem <sup>2B</sup>		
*Mexican Population includes 33% of children under 9 years <sup>3</sup> .				

Applying action levels in order to get concentration guides in some of the links of the pathway, should be done after studying the population at risk.

### Conclusions

On assessing the dose to average population, the common procedure is to survey the environment, by measuring activity in suitable samples and data obtained must be translated into dose for people exposed, or compared with data set as based on dose limits, in any case, parameters are needed for dose assessment and the values calculated are as good as the actual numbers used.

On the other hand values for parameters are different among different people and it is important, to dedicate some effort in obtaining these parameters and governmental agencies, beside the Nuclear Energy Institute should be encouraged to do research in this field, in order to find for average Mexican population physiological and morphological data, on food consumption habits and transfer factors for common food in Mexico. This studies are of importance, specially in areas where nuclear industry will be developed.

### References

- 1.- ICRP. "Recommendations of the ICRP adopted Sept. 17, 1965". ICRP Publ. 9, (1966).
- 2.- UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION. IONIZING RADIATION: Levels and Effects, Vol. 1, United Nations, New York, 1972.
- 3.- SERIA DE INDUSTRIA Y COMERCIO, DIR. GRAL. DE ESTADISTICA. "Anuario estadístico de los Estados Unidos Mexicanos 1968-69". México, (1971).
- 4.- COLEGIO DE MEXICO. "Dinámica de la Población en México". (1971).
- 5.- BENITEZ Z.R. y CABRERA G. "Tablas abreviadas de la Mortalidad de la Población en México 1930, 1940, 1950, 1960". El Colegio de México (1967).
- 6.- MORGAN K.Z. "Health Physics and the environment". Presented at the Health Physics Topical Symposium (1971).
- 7.- INSTITUTO NACIONAL DE ENERGIA NUCLEAR. "Primer Informe a la Junta Directiva. Programa de Trabajo 1972. Proyecto de Inversiones 1972 y 1972-76". México (1972).
- 8.- PEREZ TAMAYO R. "Principios de Patología". Prensa Médica Mexicana. México (1969).
- 9.- PEDRO ARROYO et al. "Los Hábitos de alimentación en una región fronteriza, Agua Prieta y Esqueda, Sonora". Div. de Nutrición L-15 (1969).
- 10.- PEREZ H, et al. Revista de Salud Pública. México 11, 223 (1969).
- 11.- TOVAR E. et al. "Revista de Salud Pública". Méx. Epoca V, 6, 443 (1964).
- 12.- CHAVEZ A. Encuestas Nutricionales en México. Div. de Nutrición L-1 2a. Ed. Inst. Nacional de Nutrición, México (1965).
- 13.- FEDERAL RADIATION COUNCIL. "Background material for the development of radiation standards". F.R.C. Pub. 1 (1960).
- 14.- BARRY P.J. "Maximum Permissible concentrations of radioactive nuclides in airborne effluents from nuclear reactors". AECL 1624 (1963).
- 15.- ICRP. Health Physics 3, June (1960).
- 16.- BRYANT P.M. Health Physics 17, 51 (1969).
- 17.- ICRP. "Recommendations of the International Commission on Radiological Protection". ICRP Publication 10. Pergamon Press (1968).
- 18.- EBERHARDT L.L. Health Physics 13, 88 (1967).
- 19.- BUREAU OF RADIOLOGICAL HEALTH AND TRAINING INSTITUTE "Radiological Health Handbook. U.S. Dept. of Health, Education & Welfare. Rockville (1970).
- 20.- ZUBIRAN S. et al. Rev. Inv. Clínica. México 14, 359 (1962).
- 21.- PEREZ H. et al. Rev. Salud Pública. Méx. 12: 441, 1970.
- 22.- WEAVER J.C. et al. J.A.M.A. June 25, 872, (1960).
- 23.- HERNANDEZ M. et al. "Valor Nutritivo de los Alimentos". Pub. de la Div. de Nutrición L-12, 5a. Ed. Inst. Nal. de Nutrición de México (1971).
- 24.- COMAR C.L. Mentioned in Training Publ. No. 164n Div. of Radiological Health. U.S. Dept. of Health, Education & Welfare.
- 25.- KAYE S.V. & NELSON D.J. Nuclear Safety 9, 1, 53-58 (1968).
- 26.- COMMITTEE ON OCEANOGRAPHY OF NATIONAL RESEARCH COUNCIL. "Radiactivity in the Marine Environment". Washington (1971).
- 27.- ATEN A.H.W. Jr. Health Physics 6, 114. (1961).
- 28.- DOLPHIN G.W. & MARLEY W.G. IAEA-SM-117/23. Environmental Contamination by Radioactive Materials. IAEA. Vienna (1969).
- 29.- International Atomic Energy Agency. Safety Series 21. Vienna (1967).

VADOSCA: A SIMPLE CODE FOR THE EVALUATION OF POPULATION  
EXPOSURE DUE TO RADIOACTIVE DISCHARGES

L. Bramati, T. Marzullo, I. Rosa, G. Zara

ENEL  
Rome, Italy

Abstract

The code consists of two parts, one for liquid discharges (VADOSCA-LI) and one for gaseous discharges (VADOSCA-GAS), and it incorporates the transfer parameters of twenty-four radioisotopes in the case of liquid discharges, and of twenty radioisotopes in the case of gaseous discharges. It allows the evaluation of the concentrations of the various isotopes in all the compartments of the critical paths outlined in the ICRP Publication No. 7, and the evaluation of the annual doses for five critical organs (whole body, G.I. tract, thyroid, bones, lung) for the various critical groups of population, on the basis of environmental parameters, such as the time of residence in a certain area, diet, type of activity, hydrological regimen, irrigation methods, and meteorological conditions.

Although extremely simple, the code allows rapid performance of all the evaluations required to define the amount of radioactivity that can be released and the associated exposures.

Introduction

As the peaceful uses of nuclear energy expanded, a great number of criteria had been voiced on environmental protection against radioactive discharges from nuclear plant; in 1965 they were collected in an orderly form in the ICRP Publication No. 7. In the light of the acquired knowledge of reconcentration of radionuclides in the environment as a result of chemical-physical (absorption, sedimentation) and biological processes, ICRP suggested assessing the doses due to the discharges following all the possible paths from the plant to man. This approach calls for a wide knowledge of the environment and transfer parameters of all nuclides present in the discharges, and sophisticated computer programs<sup>1</sup>.

In the meanwhile the public opinion had risen against environmental degradation to the point of objecting even to the construction and operation of nuclear power stations, and especially to their effluents. The Regulatory Agencies were thus pressed to lower the limits of discharged radioactivity and to request of each station an analysis of its impact on the environment, more or less in harmony with the ICRP recommendations. This attitude is very well illustrated in a recent AEC document<sup>2</sup>, which analyzes the merits of the various types of waste treatment plants in the light of the reduction of the population exposure to "as low as practicable" values.

Likewise, in Italy CNEN has long ago adopted the criterion of issuing operating licenses containing limitations on the radioactive discharges based on the actual station requirements and on an analysis of the receptivity of the environment. ENEL, the national producer and distributor of electricity in Italy, responsible for providing to CNEN such an impact analysis for its nuclear stations, developed a computer code in two parts that simplify the evaluation of

of population exposure due to liquid and gaseous radioactive wastes (called VADOSCA-Li and VADOSCA-Gas).

### VADOSCA-Li

In its present form, the code covers twenty-four radionuclides, of which some are fission products ( $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ ,  $^{91}\text{Y}$ ,  $^{95}\text{Zr}$ ,  $^{106}\text{Ru}$ ,  $^{131}\text{I}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{140}\text{Ba}$ ,  $^{144}\text{Ce}$ ), others are activation products ( $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{32}\text{P}$ ,  $^{35}\text{S}$ ,  $^{45}\text{Ca}$ ,  $^{51}\text{Cr}$ ,  $^{54}\text{Mn}$ ,  $^{59}\text{Fe}$ ,  $^{58}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{65}\text{Zn}$ ,  $^{110m}\text{Ag}$ ,  $^{124}\text{Sb}$ ), plus an alpha emitter,  $^{239}\text{Pu}$ . The choice of these nuclides was dictated by the frequency of their presence in the station discharges and by their radiotoxicity. Nuclides having a half-life of less than a few days are not considered. However, the code can handle thirty isotopes to accommodate particular situations with isotopes that are not listed above.

When the wastes are discharged to a closed body of water (sea or lake) the main dilution considered is that due to the condenser coolant; moreover, dilution coefficients are fed to the computer for each critical path and for each single case on the basis of hydrological and thermal considerations. Introduction of the commonly used diffusion models<sup>3,4</sup> was avoided because they lack the required flexibility and they required a semi-empirical approach.

When the wastes are discharged into a river, they are considered diluted first by the condenser coolant and then by the average flow of the river itself. If the water table is involved, decontamination coefficients are fed to the computer to take into account the absorption due to the soils crossed by the liquid. Finally, additional dilution coefficients can be introduced for each pathway to take into account any other mechanisms, such as suitably large tributaries or sedimentation effects.

Fig. 1 provides a schematic illustration of the critical pathways considered for the liquid wastes. The code calculates the doses to four special groups, namely, fishermen, other workmen, local population, farmers. A separate calculation is made for the doses originated by drinking water. The doses are evaluated for five critical organs (whole body, gastro-intestinal tract, thyroid, bones, lungs) and they are expressed in mrem/year if the discharges are ex-

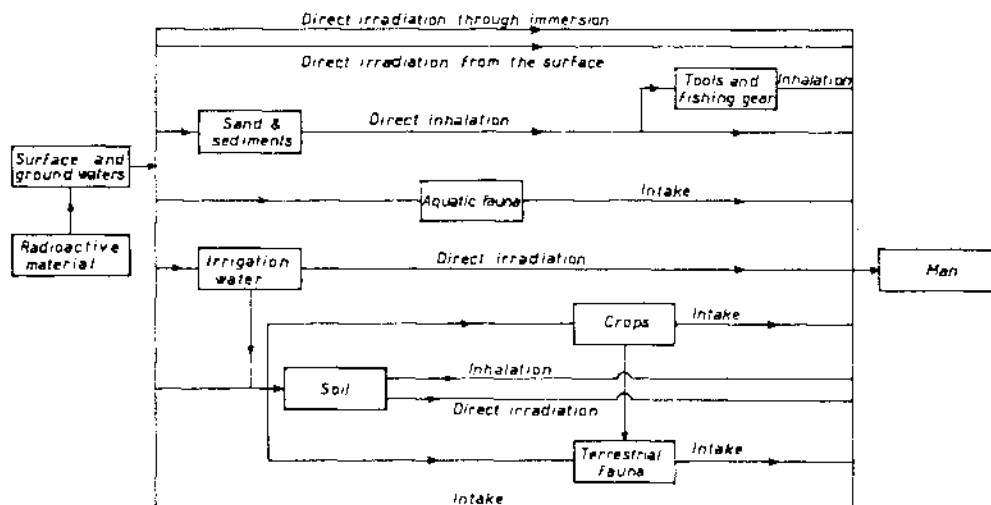


Fig. 1-Critical pathways for liquid discharge

pressed in Ci/year.

The concentrations in each compartment in Fig. 1 are obtained by means of a linear relation from the preceding compartment and with the transfer coefficients taken from the literature or from the concentrations of stable isotopes in the respective compartments or evaluated on the basis of other considerations, as illustrated below.

Marine biota and fresh water fish. Frecke's<sup>5</sup> concentration factors were used for the former, whereas reference was made to the concentration factors found in the literature<sup>6,7,8</sup> for the latter.

Irrigated crops. Six types of crops were considered, namely, rice, other cereals, leafy vegetables (including forage), radishes, other vegetables and fruit. The literature provides very scanty information on the relative concentration factors, so reference was often made to the method of specific activity; a standard concentration was assumed for fresh water and the concentrations for the vegetables were taken from the literature. When this method was not applicable, the concentration factors of chemically similar elements were used.

Terrestrial animals and products. The following critical pathways were considered: meat of herbivores (beef, rabbit, lamb, pork) and milk, poultry and eggs. The radionuclide transfer is considered from the forage for herbivores and from cereals for poultry. The transfer factors for C, P, S, Ca, Fe, Mn, I, Zn, Co, Sr were obtained by the specific activity method, the factor for Cs from the literature<sup>9</sup>. Cr, Co, Y, Zr, Ru, Ag, Sb, Ba, Ce and Pu were considered little assimilated by the GI tract, (i.e. 1% in respect of Cs).

Sediments. Lacking specific data that would any way strongly depend on local factors (meteorology, hydrology, type of river banks and soils), we assumed standard transfer factors that varied by a factor of 10 depending on the half life and type of sediment (sandy or silty). The tailrace was taken as the preceding compartment before complete mixing with the receiving body.

Irrigated tillage. The irrigating water was assumed to deposit its radioactivity in the first ten centimeters of soil and equilibrium concentration was assumed to be reached with a 15% leaching yearly.

Once the concentration in each compartment is known, the doses to the critical groups are calculated taking into account critical parameters, such as time of residence in water, on water, on sediments, on irrigated land, dietary habits, and working time. The doses due to irradiation from water and land were calculated with the formula<sup>10</sup>:

$$D_i = 1.06 \times 10^3 \times C_i \times S_i \times T$$

where  $D$  is expressed in mrem/yr,  $C_i$  is the concentration of isotope  $i$  (Ci/m<sup>3</sup>),  $S_i$  is the energy of beta and gamma rays emitted by the isotope  $i$  (Mev),  $T$  is the irradiation time (hours). The formula is based on the assumption of a plane infinite source and it overestimates the actual dose. When applied to sediments, the formula gave an excessive dose because of their high concentration factors; therefore, a more sophisticated mathematical treatment<sup>11</sup> was used, whereby allowance is made for the actual thickness of the sediments, for the overlying water layer and for the geometry effect of the source (semi-plane in the case of irradiation of a critical group residing on river banks or sea coast).

For the intake doses the CMAs given in the ICRP Publication No. 2 for the various organs, following the method proposed by Essig<sup>12</sup>, and the yearly food consumption of the various population groups were used.

The doses due to inhalation of resuspended material (sediments and agricultural soil) are factored in by assuming a standard breathing rate of 0.83 m<sup>3</sup>/hr and a content of airborne particulates of 10 mg/m<sup>3</sup>. The latter value is a maximum obtained from samples of air taken in different conditions, such as open country, construction sites, residential areas.

The code is written in FORTRAN; it needs 20 K memories and it takes approximately 15 seconds in a GE 635 computer.

### VADOSCA-Gas

Twenty nuclides are considered, of which eight noble gases (fission and activation products: <sup>41</sup>A, <sup>85m</sup>Kr, <sup>85</sup>Kr, <sup>87</sup>Kr, <sup>88</sup>Kr, <sup>133</sup>Xe, <sup>135</sup>Xe, <sup>138</sup>Xe), various isotopes as particulates (<sup>60</sup>Co, <sup>88</sup>Rb, <sup>89</sup>Sr, <sup>90</sup>Sr, <sup>131</sup>I, <sup>134</sup>Cs, <sup>137</sup>Cs, <sup>138</sup>Cs, <sup>239</sup>Pu), and <sup>3</sup>H, <sup>13</sup>N, <sup>14</sup>C in the form of vapor. Short-lived isotopes were taken into account only when experience indicated that they might be present in the discharges. At any rate, the code can accommodate other five nuclides. For each nuclide it is necessary to supply the average yearly discharge.

Atmospheric diffusion is evaluated with Pasquill's theory<sup>13</sup> adapted to the meteorological data available. In fact, two sets of information are handled by the code, namely,

Set A: Only a wind rose and an average distribution of the six stability categories are available. This is the general case described by P. Bryant<sup>13</sup>.

Set B: In this case, in addition to the wind rose, one must have the frequency distribution of the stability categories for each wind direction, subdivided by wind speed ranges. This is the sort of information used by May and Stuart for their diffusion analysis at Brookhaven<sup>14</sup>.

For simplicity, in its present form the code does not take into account particular effects such as the cloud depletion due to deposition, down draft and building effect; the particulate deposition rate is assumed constant at 3x10<sup>-2</sup> m/s.

Fig. 2 shows the critical pathways for gaseous discharges. Two sets of doses are considered: (1) doses due to irradiation and inhalation as a function of the distance from the stack and direction of plume travel; (2) doses due to intake of agricultural (vegetal and animal) produce. The former are calculated

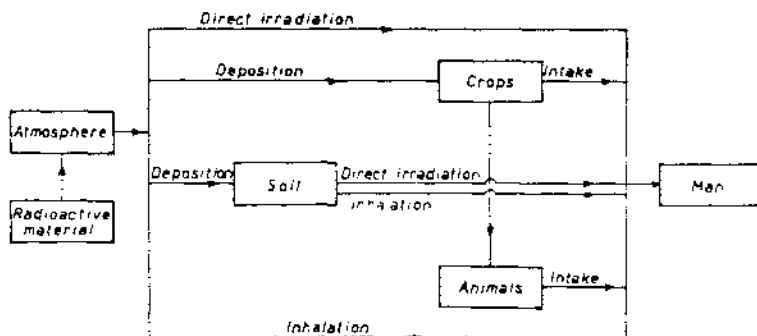


Fig 2 Critical pathways for discharges to the atmosphere

over a discrete number of distances between 0,3 and 50 km and for a maximum of sixteen sectors; the latter are calculated only at the point of maximum fallout and for two population groups (farmers and local population) and for the usual five organs.

The exposure due to the cloud is calculated with a method developed by Garmertsfelder<sup>15</sup>. This method takes into account the contribution made by a cloud of finite dimensions with a gaussian vertical concentration distribution and an average horizontal concentration.

The exposure due to inhalation is calculated from the ground concentrations, evaluated with Pasquill's formula<sup>13</sup>. The same formula is used to calculate the deposition on the crops, excluding the short-lived <sup>88</sup>Rb and <sup>138</sup>Cs.

The transfer factors for crops were calculated taking into account the growth time of the crops, the concentrations of the stable isotopes, the decontamination due to decay and processing. The related doses were calculated like those resulting from the liquid discharges.

The code is written in Fortran; it needs 60 K memories and it takes about 60 seconds in a GE 635 computer.

#### General Comments

Applied to the analysis of environmental impact, VADOSCA has proved to be a useful tool for the health physicist. While relieving him of painstaking calculations, it has compelled him to single out from the host of ecological data those that were most significant for radioprotection. In practical application, it was necessary to modify the standard transfer factors to adapt the code to the results of the surveys at the nuclear station sites. For instance, for the nuclear station on the Garigliano river at 11 km from the coast, the Cs transfer factor for fish was found to be much lower (200) than the standard value (1000). Instead, at Trino Vercellese on the upper course of the Po river the same factor was 1400 and for certain species of fish that feed on periphyton it was as high as 2500. This example is typical to illustrate the limitations of this type of code, which starts from standard values selected conservatively and then must be adapted through successive approximations.

In the negotiations with the safety authorities for the discharge permits, the availability of an agreed standard code facilitates mutual understanding and saves considerable time.

It is recognized that further improvements of the code are necessary in order to factor in all the information on radioactivity concentration and dispersion mechanisms in the environment that is building up. Moreover, the next step should be the use of dynamic models of the type already widely used in other branches of ecology<sup>1,16</sup>, but this will be warranted only when a better understanding of the aforesaid mechanisms has been acquired and more complex problems are to be dealt with.



### Bibliography

1. S. V. Kaye, R. S. Booth, P. S. Rohwer, E. G. Struxness, "Ecological Model Development for a Methodology to Estimate Doses to Human Population". Proc. of Rome Symp. (Sept. 7-10, 1971).
2. "Draft Environmental Statement Concerning Proposed Rule-Making Action: Numerical Guides for Design Objectives and Limiting Conditions for Operation to Meet the Criterion 'As Low as Practicable' for Radioactive Material in Light-Water-Cooled Nuclear Power Reactor Effluents", USAEC Report, (Jan. 1973).
3. D. W. Pritchard, A. Okubo, H. H. Carter, "Observations and Theory of Eddy Movement and Diffusion of an Introduced Tracer Material in the Surface Layers of the Sea". Proc. of Vienna Symp., (May 16-20, 1966).
4. N. H. Brooks, "Diffusion of Sewage Effluents in an Ocean Current". Proc. of 1st Int. Conf. on Waste Disposal in the Marine Environment, (1961).
5. A. M. Freke, "A Model for the Approximate Calculation of Safe Rates of Discharge of Radioactive Wastes into Marine Environments". Health Phys. 13, 743-758 (1967).
6. G. W. Bryan, A. Preston, W. L. Templeton, "Accumulation of Radionuclides by Aquatic Organisms of Economic Importance in the United Kingdom". Proc. of Vienna Symp., (May 16-20, 1966).
7. R. L. Blanchard, B. Kahn, "Pathways for the Transfer of Radionuclides from Nuclear Power Reactors through the Environment to Man". Proc. of Rome Symp., (Sept. 7-10, 1971).
8. S. E. Thompson, C. D. Burton, D. J. Quinn, Y. C. Ng, "Concentration Factors of Chemical Elements in Edible Aquatic Organisms". TID 4500, U. C. 48, (Oct. 10, 1972).
9. G. Ward, J. Johnson, "The  $^{137}\text{Cs}$  Content of Beef from Dairy and Fed-Lot Cattle", Health Phys. 11, 95 (1965).
10. K. E. Cowser, W. S. Snyder, C. P. McCammon, C. P. Straub, O. W. Kochtitzky, R. L. Herviv, E. G. Struxness, R. J. Morton, "Evaluation of Radiation Dose to Man from Radionuclides Released to the Clinch River". Proc. of Vienna Symp., (May 16-20, 1966).
11. T. Rockwell III, "Reactor Shielding Design Manual", page 353, (1966).
12. T. H. Essig, "A Method for Calculating Human Radiation Doses in the Environment". Proc. of Augusta Symp., (Jan. 24-26, 1968).
13. P. M. Bryant, "Methods of Estimation of the Dispersion of Windborne Material and Data to Assist in their Application". Report AHSB(RP)R42, (1964).
14. M. J. May, I. F. Stuart, "Comparison of Calculated and Measured Long-Term Gamma Doses from a Stack Effluent of Radioactive Gases". Proc. of Augusta Symp., (Jan. 24-26, 1968).
15. TID-24130, "Meteorology and Atomic Energy", pages 351-355, (1968).
16. R. S. Booth, S. V. Kaye, P. S. Rohwer, "A System Analysis Methodology for Predicting Dose to Man from a Radioactively Contaminated Terrestrial Environment". Proc. of Oak Ridge Symp., (May 10-12, 1971).

## MEDICAL EXPOSURE

### NATIONWIDE EVALUATION OF X-RAY TRENDS (NEXT)

Joel O. Lubenau, FDA, Northeastern Radiological Health Laboratory, Winchester, Mass. 01890; Charles M. Hardin, State Department of Health, Frankfort, Kentucky 40601; John W. Shaver, State Board of Health, Raleigh, North Carolina 27602; I. Paul Leggett, Jr., FDA, Bureau of Radiological Health, Rockville, Maryland 20852; and LaVert Seabron, FDA, Bureau of Radiological Health, Rockville, Maryland 20852.

#### Abstract

The impact of efforts by government agencies to minimize and control unnecessary patient exposure in medical x ray has been difficult to assess. Population exposure studies, such as those conducted in 1964 and 1970 of the United States Genetically Significant Dose, are expensive. Reports of the number of x-ray machines in compliance with equipment standards, such as for filtration and collimation, are traditional but are difficult to relate to patient or population exposures. Furthermore, such reports are limited to machine parameters and do not reflect other factors influencing patient dose such as operator training, film and screen selection and film processing.

A Task Force of State and Bureau of Radiological Health representatives, created in 1971, has applied the "standard man" concept as a method of evaluating the impact of government control efforts on medical x-ray exposure.

A limited number of randomly selected medical x-ray facilities are visited. The operator of an x-ray unit is requested to set the technique factors for a selected examination of a patient whose anthropometric characteristics have been standardized. Exposure data is collected using standardized procedures and equipment. Organ Dose Index values are generated for selected critical organs.

By eliminating patient size variation, the wide range of exposure technique factors currently employed in medical radiography has become readily apparent, together with the wide range of gonad doses encountered by a standard patient. It is expected that these can be correlated with governmental agency activities in radiological health and can be modified by their efforts.

The methodology of the NEXT Organ Dose Index System is simple. Fifty local, State and Federal radiological health programs in the United States are participating or planning to participate in the system which began operation in October, 1972. Some results of the first nine months of its operation are presented. The system appears to be far more efficient in assessing the impact of governmental program efforts in medical x ray than previously used methods. It avoids the complexity and cost of population exposure studies but can relate program impact in terms of patient exposure and dose.

## Radiological Health Efforts in the United States

Historically, regulatory radiological health efforts in the United States trace their origins to the state and local levels, particularly as they affect medical x-ray use. Currently 50 States and the District of Columbia conduct radiation control programs under either general public health laws or specific enabling legislation.<sup>1</sup> Technical details of regulatory control are usually not included in authorizing legislation but are delegated to a radiological health agency or advisory group. More recently, under authority of Public Law 90-602, Federal performance standards for medical x-ray equipment have been prepared.<sup>2</sup>

In 1962 the Council of State Governments, in cooperation with the United States Public Health Service and the United States Atomic Energy Commission, developed Model Regulations for Radiation Control which included a section on the "Use of X rays in the Healing Arts."<sup>3</sup> The original regulations have been updated twice and are undergoing further revision. Most States which have adopted regulations in the area of medical and dental x ray have followed this guidance thus providing some measure of uniformity. The Council of State Governments' regulations have been based upon the recommendations of various standard-setting bodies, particularly those of the National Council on Radiation Protection and Measurements.<sup>4</sup>

The major thrust of radiological health agencies' activities in the United States, to date, has been directed at upgrading the x-ray equipment used in the diagnostic healing arts to meet minimum regulatory standards. Consequently, the primary purpose of most medical x-ray inspection programs has been to determine that the equipment requirements of the regulations are being followed. However, advantage is frequently taken of the personal contact during an inspection of a medical or dental facility to discuss with personnel other items related to the use of the equipment.

For example, exposure technique and film processing may be reviewed with the operator. In such cases, the inspection visit becomes a mini-training program. As a result, in addition to a report of items which do not meet regulatory requirements, recommendations may also be provided to the user which, if followed, can improve the overall radiological health aspects of an x-ray facility.

Regular training courses are offered by many radiological health agencies, usually in conjunction with, or using the resources of, various Federal agencies. The Bureau of Radiological Health of the Food and Drug Administration has developed training packages for use by State and local agencies. Many of these agencies routinely offer the services of their staff to provide lectures for ongoing formal training programs in hospitals or other schools.

Many State and local radiological health agencies review and approve x-ray facility plans and specifications. Items such as adequacy of radiation barriers, film handling and processing facilities, type and location of radiation machines, and adequacy of ancillary equipment are subjected to critical review.

Radiological health agencies realize that ensuring that proper equipment is provided is only one parameter in the equation of optimizing the benefit of an x-ray exposure of a patient. With this in mind, three States and the Commonwealth of Puerto Rico have approved specific laws which establish minimum standards for education, training and experience for certain user groups that apply x rays to humans for diagnostic or therapeutic purposes.<sup>1</sup> The United States Public Health Service has developed guidelines for the establishment of

licensing programs for x-ray users in the healing arts.<sup>5</sup>

All of these programs are directed toward minimizing exposure in the environs of an x-ray installation and optimizing the benefits of patient radiation. But how do these agencies determine their program effectiveness? National X-ray Exposure Studies such as conducted by the United States Public Health Service in 1964 and in 1970 can and do provide information to answer this question.<sup>6,7</sup>

However, such x-ray exposure studies require considerable investments of time, money and personnel at local, State and Federal levels. This time interval - 6 years - is too long for use by agencies requiring the kind of information which is necessary to justify budget requests, to plan program priorities and to evaluate past program effectiveness. These activities are performed on an annual basis and information must be available on an annual basis.

The usual information resources available to agencies are limited to reports of inspection programs such as the number of x-ray machines in compliance, the number of x-ray machines with deficient filtration, etc. Increasingly, such data has been found unsuitable for identifying specific problem areas, for justifying existing radiological health programs and budget requests, and for program planning. Such terminology is not meaningful to many public officials responsible for planning fiscal and personnel resource allocations to the various technical programs under their jurisdiction. Instead, program effectiveness or needs must be reported in people-related terms.

#### Nationwide Evaluation of X-ray Trends (NEXT)

In May 1971 the Conference of Radiation Control Program Directors called for the formation of a Task Force to design a uniform program for surveys of x-ray facilities.<sup>8</sup> The Task Force, co-sponsored by the Bureau of Radiological Health of the Food and Drug Administration, Department of Health, Education and Welfare, was appointed in July 1971 and consisted of equal representation from State and Federal radiological health agencies.

The Task Force adopted the project name of "NEXT", an acronym for Nationwide Evaluation of X-ray Trends. In reviewing its charge, the Task Force identified four specific objectives<sup>9</sup> it wished to meet:

1. Design a system to measure the effectiveness of radiological health programs,
2. Design a system which would enable program priorities to be assigned on a rational, documentary basis,
3. Identify the optimum components of a radiation survey, and
4. Provide for the uniform collection of data related to radiological health.

The development of an Optimum Survey Procedure Manual would partially satisfy these objectives. This is currently under revision and will not be discussed here.

While the production of a manual of optimum survey procedures will fulfill a long sought need by local and State radiological health programs, the NEXT Task Force recognized that additional parameters were required to meet the objectives of providing a system to measure program effectiveness and to assign program priorities. The Organ Dose Index System (ODIS) was devised to meet these objectives.

## The Organ Dose Index System (ODIS)

The Organ Dose Index System is based on an annual survey of a statistically representative sample of the x-ray tubes within an agency's jurisdiction. The results are intended to provide the agency with a measure of its effectiveness in reducing unnecessary radiation exposure during diagnostic radiography. The system was not designed to replace compliance survey procedures now in use but is intended to be an adjunct to these procedures. The system provides specific organ doses called Organ Dose Indexes for selected x-ray procedures applied to a standard sized patient.

The term "Index" has been appended to "Organ Dose" because the calculated dose values are not an "average", nor representative of the population dose. They are the organ doses only for an individual who fits the physical characteristics of the "standard patient". (This patient was subject number 16 of the group who participated in the Johns Hopkins study to determine scatter to primary x-ray beam exposure ratios.<sup>6</sup>) Organ Dose Index, is therefore, a people-related quantity. Since it reflects a "standard patient" it removes the variable of patient size. It has the potential for evaluating variations of organ dose by type of facility, technique, operator training, beam size and shape, etc. The methodology of calculating radiation doses to the gonads used in the Organ Dose Index System is that used in the X-ray Exposure Studies of the United States in 1964 and 1970, with modifications.<sup>6</sup> Other organs have been identified by the Task Force for which calculated radiation doses are desired. These are the thyroid, lens of eyes and bone marrow. Organ Dose Indexes for these organs are awaiting development of suitable dose models.

Twelve common diagnostic radiographic examinations are included in the Organ Dose Index System:

<u>Projection</u>	<u>Body Part of Interest</u>	<u>Body Part Thickness (centimeters)</u>
Chest (P/A)	Thorax	23
Skull (Lateral)	Head	15
Abdomen (KUB)		
Scout Film (A/P)	Abdomen	23
Retrograde Pyelogram		
Scout Film (Cysto Units) (A/P)	Abdomen	23
Thoracic Spine (A/P)	Thorax	23
Cervical (A/P)	Neck	13
Lumbo-Sacral Spine (A/P)	Abdomen	23
Full Spine (A/P) (14"x36" film size only)	Chest and Abdomen	23
Feet (Weight Bearing) (D/P) (Podiatrists Only)	Foot	8
Dental Bitewing (Posterior)	Left Bicuspid and Molars	-
Dental Periapical	Central Incisor (Maxillary)	-
Dental Cephalometric (Lateral)	Head	15

These projections were selected to provide a useful cross-section of x-ray examinations encountered in private and institutional medical care facilities.

To obtain Organ Dose Indexes, a statistically representative sample of the healing arts x-ray facilities within a participating agency's jurisdiction is drawn by the participating agency on an annual basis.

During the inspection of a selected x-ray facility, the inspector determines which of the twelve selected examinations are performed most frequently on the machine being inspected. The inspector asks the operator to set the technique (milliamperage, kilovoltage, exposure time, target-to-film distance, collimation, etc.) that would be used for this standard patient. For example, if a Chest P/A is the most frequent examination performed with the machine in question, the operator is asked to set the technique that would be used for a patient having a 23 centimeter chest. Appropriate measurements of x-ray beam exposure, quality and beam size are made utilizing standardized procedures and equipment.<sup>9</sup> A Mean Ovarian Dose and a Testicular Dose are then calculated from the measurement data using computer programs.

### Preliminary Results

The NEXT Organ Dose Index System began October 1, 1972. At the end of the first nine months operation, 32 States and 3 Federal agencies were participating. Additionally, NEXT data has been processed for one foreign government and for an international health agency. At the end of nine months of operation (June 30, 1973), data for 3,431 projections had been collected in the United States and submitted for processing. Pre-edit and quality control checks designed to eliminate erroneous data are applied to all submitted data. 2,316 projections passed these checks and were entered upon the NEXT ODIS master file

The present data pool is not yet complete. Many participating agencies have not completed surveying their annual representative sample. Not only is it too early to attempt an identification of trends, but the baseline has not yet been established.

Nonetheless, preliminary analysis of existing data does seem to validate some of the concepts, and expectations of the system.

For example, data is available for 291 cases of the Lumbo-Sacral (A/P) projections. Registered radiologic technologists performed 129 of these projections, the others being performed by practitioners or other persons. For these 291 applications of this projection to our standard patient:

1. The reported kVp ranges from 50 to 110,
2. The reported mAs ranges from 10 to 400,
3. The measured tube target-to-film distance ranges from 30 to 72 inches,
4. In view of the ranges in the above 3 categories it was not surprising to find the calculated exposure at skin entrance varied by 2 orders of magnitude.
5. X-ray Field Size at the film varied from well collimated beams limited to the spinal column, e.g., 5"x16" to large circular beams, e.g., 49", and even large rectangular beams, e.g., 22"x47",
6. In consequence of these variations, the Mean Ovarian Dose Index ranged from 4 mrad to 951 mrad and the Testicular Dose Index ranged from <0.5 mrad to about 2,300 mrad.

This kind of variation is not unique to the Lumbo-Sacral Spine examination. A another example, 42 cases of the Retrograde Pyelogram Scout (Cysto only) (A/P) were collected. This is a specialized projection rarely performed outside a hospital or major private practice facility. The x-ray machine operator in 34 of the 42 cases was a registered radiologic technologist.

1. The reported kVp varied from 68 to 90,
2. The reported mAs varied from 20 to 250,

3. The measured tube target-to-film distance varied from 31 to 59 inches,
4. The calculated exposure at skin entrance varied from 205 mR to 1,200 mR,
5. Beam size at the film varied from a 25" diameter circular beam to a 10"x13" rectangular beam,
6. The mean Ovarian Dose Index varied from 45 mrad to 507 mrad and the Testicular Dose Index varied from 1 mrad to about 1,000 mrad.

It is evident that the range of variation for this projection is smaller than for the Lumbo-Sacral Spine, but it is still quite large. As noted earlier, this projection is a specialized procedure restricted to a limited number of facilities. In comparison to the Lumbo-Sacral projection, a larger proportion of the operators were registered radiologic technologists and one may speculate on the influence of this factor.

Even so it is perplexing to find such wide variation in technique for the same examination for the same standard patient.

#### NEXT, Now and Future

These preliminary results suggest that opportunity does lie ahead for reducing unnecessary diagnostic x-ray exposure by identifying facilities using high exposure techniques. The NEXT Organ Dose Index System, by eliminating patient size variation and utilizing standard survey techniques, provides an objective method of accomplishing this.

Presumably, a radiological health agency effort, directed at identifying the high exposure facilities, followed by efforts to change their techniques, if successful, should be reflected by a trend, over time, to lower average organ dose indexes. The NEXT Organ Dose Index System will monitor these trends.

The NEXT Organ Dose Index System will not provide estimates of population dose, nor will it provide per capita dose information. It will provide information that can be used to effect changes in population dose. It will monitor changes in medical diagnostic x-ray application and trends in medical x-ray exposure.

## REFERENCES

1. Lois A. Miller. Federal/State Radiation Control Legislation 1972, DHEW Publication (FDA) 73-8035 (1973), National Technical Information Service, Springfield, Virginia 22151.
2. Federal Register of August 15, 1972 (37 FR 16461, Office of the Federal Register, Washington, D.C. 20408.
3. Council of State Governments. Suggested State Regulations for Control of Radiation, Revised June, 1970.
4. National Council on Radiation Protection and Measurements. NCRP Report 33, Medical X-ray and Gamma Ray Protection for Energies up to 10 MeV- Equipment Design and Use (1972) and other NCRP reports, NCRP Publications, Washington, D.C. 20014.
5. Model Legislation for Users of Ionizing Radiation in the Healing Arts, BRH/ORO 70-8 (1970), National Technical Information Service, Springfield, Virginia 22151.
6. Population Exposure Studies Section, Bureau of Radiological Health, Population Dose from X-rays U.S. 1964, Public Health Service Publication No. 2001 (1969) U.S. Government Printing Office, Washington, D.C. 20402.
7. Joseph N. Gitlin and Philip S. Lawrence. Population Exposure to X-rays U.S. 1964, Public Health Service Publication No. 1519 (n.d.) U.S. Government Printing Office, Washington, D.C. 20402.
8. 3rd Annual National Conference on Radiation Control, DHEW Publication (FDA) 72-8021, BRH/ORO 72-2 (1971) National Technical Information Service, Springfield, Va. 22151.
9. 4th Annual National Conference on Radiation Control, DHEW Publication (FDA) 73-8003, BRH/ORO 73-3 (1972) National Technical Information Service, Springfield, Va. 22151.



# THE GENETICALLY SIGNIFICANT DOSE DUE TO MEDICAL X- RAY EXAMINATIONS IN THE NETHERLANDS

by J.A. Koen and J. Weber  
Interuniversitair Reactor Instituut

## Summary

Recent in situ measurements of the gonadal doses of 2500 male patients in seven Dutch hospitals make a reappraisal of the genetically significant dose (GSD) due to X-ray examinations in the Netherlands possible. It was found that the gonadal doses of nearly all examination types in all hospitals form a log normal distribution with a standard deviation of the same order of magnitude as the mean doses. The mean doses differ largely between the hospitals. Measurements in many hospitals will be necessary to obtain a reliable national mean dose for each examination type. The number of examinations for each type varies appreciably per district and needs careful consideration. The child expectancy, which did change drastically during the last 4 years, did not influence the GSD.

## Introduction

The Genetically Significant Dose (GSD) due to X-ray examinations in the Netherlands was estimated by Beekman<sup>1)</sup> over 1959 and by Beentjes<sup>2)</sup> over 1967. The former estimation resulted in a GSD of 7 mrem, the latter in different values, ranging from 19 - 40 mrem. Beekman used a set of relatively low gonadal doses obtained from phantom measurements with minimum beam size. In this respect her result is a minimum value. The number of examinations was obtained from the registers of all medical services in the city of Leiden and surroundings. For this confined population the total number of examinations was 0.55 per year in 1959, dental radiography and mass survey excluded. Beentjes used different sets of doses obtained from the literature and the phantom measurements of Beekman. The frequency of examinations was obtained from health insurance companies operating in different districts and covering 9 million people out of a total population of 13 million. The number of examinations per caput per year varies per district from 0.25 to 0.45 with a mean of 0.37. Dental radiography and mass radiography are excluded. The latter would add 0.21. Since, no in situ dose measurements were available in our country we decided to perform a survey in the hospitals in order to obtain dose values as they really occur. The preliminary results from measurements on male patients now available permit some conclusions about the procedure to be followed for gonadal dose measurements and their influence on the GSD estimate. They will be discussed in this paper.

## Methods

We used thermoluminescent dosimeters, consisting of three extruded lithium fluoride ribbons (1/8 x 1/8 x 0.035 inch) from Harshaw, wrapped up in plastic film. Up to this moment we performed measurements on male patients only. The dosimeter is attached to the scrotum during the X-ray examination. The measured exposure permits to calculate the testis dose if the following factors are properly taken into account.

- a) The difference between the skin exposure and the testis exposure is estimated to be about +5% in the direct beam and -5% in scattered radiation.
- b) A conversion from testis exposure to absorbed dose, chosen as 0.91 rad per röntgen.
- c) The sensitivity of LiF for diagnostic X-rays compared with 0.662 MeV <sup>137</sup>Cs radiation is estimated to be between 1.25 and 1.38.

The absorbed dose can be obtained from the exposure (calibrated with <sup>137</sup>Cs) through division by an overall correction factor, deduced from a, b and c between 1.55 and 1.40 for direct radiation and between 1.40 and 1.30 for the scattered radiation. An arbitrary value of 1.4 was chosen.

### Results of the measurements

The results of the measurements on about 2500 male patients in seven Dutch hospitals are given in table 1.

The mean gonadal dose  $d_{ij}$  of each examination type (i) is indicated separately for each hospital (j) with the number of patients  $n_{ij}$  and the standard deviation  $s_{ij}$  of the doses. The X-ray departments of the hospitals have slightly different characters. They comprise one academic hospital, one military hospital and five peripheral hospitals in different cities. The hospitals numbered 3 and 4 did not use lead shielding at all, while hospital 2 always used lead shielding for the examination types IVP, lumbo-sacral region, pelvis and hip. In hospital 5 an image intensifier was consistently used for examinations (fluoroscopy and radiography) of the lower gastro-intestinal tract.

For each examination type and hospital the distribution of the measured doses proved to be log normal. An example is given in fig. 1 and 2.

The standard deviation ( $s_{ij}$ ) is often larger than the mean dose ( $d_{ij}$ ). Since the accuracy of each measured dose is better than 10% (except of measurements at the edge of the direct beam where the localisation is uncertain) this standard deviation reflects the real difference between the individual doses. The relative standard deviation of the mean dose ( $d_{ij}$ ) is

$$\frac{s_{ij} \cdot 100}{d_{ij} \sqrt{n_{ij}}} \%$$

and it is used to calculate an upper and lower confidence limit of the doses (95% confidence) obtained in each hospital.

The mean gonadal doses for one examination type differ significantly between the hospitals. Local circumstances influence the doses to a large extent. For each examination type (i) the mean  $\bar{d}_i$  of the doses  $d_{ij}$  obtained in the 7 hospitals is calculated, together with its standard deviation  $s_i$ . From table 1 we see that  $s_i$  has the same order of magnitude as  $\bar{d}_i$ .

### Calculation of the GSD

The measured gonadal doses make a reassessment of GSD possible.

Our first measurements happened to be in the hospitals 3 and 4 where relatively high doses were obtained. These results gave us the impression that the doses of Penfil and Brown<sup>3)</sup> are appropriate for the Netherlands. However, further measurements, performed in the other hospitals, make this conclusion doubtful. It appears that the data out of a restricted number of hospitals are subject to such a large variation that this causes one of the major sources of error in the estimation of the GSD, as follows also from the calculated standard deviation  $s_i$  of  $\bar{d}_i$ .

For the calculation of the GSD due to the examination of male patients we used the mean of the doses  $d_{ij}$  weighted according to the number of measurements performed in each hospital (see table 2, column 3). The frequency of examinations of each type is taken from Beentjes and the child expectancy over 1971 is used. At the bottom of the table a correction of 10% per year for the increase of the number of examinations during the years 1967-1971 is given<sup>4)</sup>. The resultant value of the GSD, 28 mrad, is subject to a large error for the following reasons.

- 1e. The influence of errors in the doses  $d_{ij}$  on the GSD is estimated with the 95% confidence limits of the mean doses. The deviations of the contribution to the GSD of the eight examination types proved to be plus or minus 36%.
- 2c. A much larger error is to be expected due to the restricted number of hospitals used in the calculation, as is already mentioned. The standard deviation of the mean  $\bar{d}_i$  indicated that measurements in about hundred hospitals should be necessary to obtain this figure accurately, unless the hospitals can be classed into groups, for instance academic and peripheral hospitals. In order to study this point a calculation of the GSD was made with, for each examination type, the maxima and the minima of the mean doses  $\bar{d}_i$  found in all hospitals. The resultant GSD for the eight examination types was 19.8. and 1.2 mrad respectively for male patients over 1967. It seems unlikely that for male patients further dose measurements will lead to a higher GSD than 32 mrad over 1971, which is the maximum calculated in this way (table 2, column 4-7).
- 3e. The frequency of examinations of each type is not precisely known and influences the GSD to a large extent. Beentjes calculated the GSD on the basis of frequencies found in six different districts. For each examination type the contribution to the GSD in the six districts varies with a standard deviation between 30 and 40%.

We conclude that an estimate of the GSD for the whole country, obtained by multiplication of the mean doses with the mean number of examinations can lead to large errors.

Table 1. Gonadal doses of male patients per examination type (i) and hospital (j). Doses in mrad.

$d_{ij}$  = mean gonadal dose       $\bar{d}_i$  = mean of the mean doses for examination type i  
 $s_{ij}$  = standard deviation       $s_i$  = standard deviation of  $d_i$   
 $n_{ij}$  = number of patients

j Hospital no.	i Intravenous pyelography			Lower gastro- intestinal tract			Lumbosacral region			Abdomen (general)			Pelvis			Hip			Femur			Stomach		
	$d_{ij}$	$s_{ij}$	$n_{ij}$	$d_{ij}$	$s_{ij}$	$n_{ij}$	$d_{ij}$	$s_{ij}$	$n_{ij}$	$d_{ij}$	$s_{ij}$	$n_{ij}$	$d_{ij}$	$s_{ij}$	$n_{ij}$	$d_{ij}$	$s_{ij}$	$n_{ij}$	$d_{ij}$	$s_{ij}$	$n_{ij}$	$d_{ij}$	$s_{ij}$	$n_{ij}$
1	228	346	71	761	1154	15	204	324	290	65	164	45	893	901	53	-	-	-	560	630	12	6	5	100
2	86	216	238	107	104	90	30	111	127	-	-	-	99	168	8	36	24	17	23	36	6	24	30	9
3	1262	2560	102	367	728	134	-	-	-	136	264	46	-	-	-	-	-	-	-	-	-	24	28	134
4	1580	1871	64	106	71	56	1048	855	93	184	350	17	807	797	20	350	453	5	-	-	-	-	-	-
5	322	670	81	48	122	35	460	950	33	58	32	5	-	-	-	347	636	20	113	97	3	2	6	70
6	167	269	83	133	44	13	156	279	18	482	421	12	-	-	-	-	-	-	-	-	4	7	92	
7	205	416	211	59	46	5	36	31	63	164	373	6	-	-	-	-	-	-	56	61	3	-	-	-
mean $d_i$	550			226			322			182			536			244			138			12		
standard deviation $s_i$	606			259			388			156			377			180			251			11		
number of measurements $n_i$			850			348			624			131			101			42			24			405

Table 2. Different estimations of the GSD over 1971. -GSD in mrad per year, gonadal doses in mrad.

Examination type.	Measured gonadal doses						Gonadal doses of Penfil and Brown 3)		Gonadal doses of Beekman 1)		
	mean doses		set of highest doses		set of lowest doses		Gonadal dose	GSD	Gonadal dose	GSD child exp. '67	GSD child exp. '71
	Gonadal dose	GSD	Gonadal dose	GSD	Gonadal dose	GSD					
<i>Hip</i>	223	0.27	350	0.43	36	0.044	1064	1.29	3323	4.06	4.04
<i>Femur</i>	307	0.67	560	1.22	23	0.050	96	0.21	91	0.20	0.20
<i>Pelvic region</i>	793	2.91	893	3.27	99	0.363	717	2.63	157	0.58	0.58
<i>Lumbosacral region</i> (lumbar spine and abdominal aortography included)	290	1.84	1048	6.69	30	0.191	2268	14.46	60	0.32	0.32
<i>Intravenous urography</i> (retrograde urography and urethro cystography included)	411	1.43	1580	5.51	86	0.299	2091	7.29	640	2.11	2.10
<i>Abdomen (general)</i>	148	0.29	482	0.94	58	0.112	254	0.49	92	0.18	0.18
<i>Lower gastrointestinal tract</i>	229	0.49	761	1.63	48	0.102	1585	3.39	45	0.10	0.10
<i>Stomach and duodenum</i> (oesophagus included)	11.2	0.09	24	0.19	2	0.016	137	1.10	4.8	0.03	0.03
<b>Total</b>		7.99		19.88		1.177		30.86		7.58	7.55
<i>Other examinations</i>		2.01		2.01		2.01		2.01		0.03	0.03
<b>Total GSD for males</b>		10.00		21.89		3.19		32.87		7.61	7.58
<b>Total GSD for females</b>		9.13						8.88			9.13
<b>Total</b>		19.13						41.75			16.71
<b>Total corrected for increase of number of examinations '67 - '71</b>		28						61			24

4e. In our country the child expectancy is decreasing rapidly during the last years. In order to get an idea of the influence of this factor we made two calculations of the GSD (male patients), one with the child expectancy over 1967 and one over 1971. The estimations were calculated following the method of Beentjes; for both years the set of doses reported by Beekman and the frequency of examinations of Beentjes was used. Although there is a strong shift in the number of live births and child expectancy in all age classes (see table 3), the resultant differences in the calculated GSD are insignificant (7.58 and 7.55 mrad for male patients over 1967 and 1971 respectively).

Table 3.  
Male child expectancy in the Netherlands

age	1967	1971
0.75 - 0	2.644	2.232
0	2.676	2.255
0 - 4	2.720	2.307
5 - 9	2.742	2.314
10 - 14	2.748	2.319
15 - 19	2.745	2.309
20 - 24	2.564	2.071
25 - 29	1.921	1.405
30 - 34	1.080	0.809
35 - 39	.494	0.294
40 - 44	.186	.100
45 - 49	.056	.028
50 - 54	.014	.005
55 - 59	.002	--
60 <sup>+</sup>	--	--

Deduced from tables of the Netherlands Central Bureau of Statistics.

### Conclusion

In spite of 2500 gonadal dose measurements and intensive efforts to obtain the frequency of each type of examination per year the estimation of the GSD is still unreliable. Improvement is only possible if an appreciable number of measurements in many hospitals is available. Relatively large statistical errors in the dose measurements may be tolerated due to the large standard deviation in the dose distributions. In our country the rather important changes in child expectancy did not influence the GSD of the male patients significantly.

### References

- 1) Beekman, Z.M.. Genetically Significant Dose from Diagnostic Roentgenology (a study concerning a defined population in the Netherlands), Thesis, Leiden (1962).
- 2) Beentjes, L.B.. An estimate of the Genetically Significant Diagnostic Roentgen-Ray Dose in the Netherlands (1967), Thesis, Utrecht, 1969.
- 3) Penfil, R.L., Brown, M.L.. Genetically Significant Dose to the United States population from diagnostic Medical Roentgenology, Radiology 90 (1968) 209.  
Population Dose from X-rays U.S. 1964, Public Health Service Publ. No. 2001, Oct. 1969.
- 4) Puylaert, C.B.A.J.. De expansie van de röntgendiagnostiek, Ned. Tijdschrift v. Geneeskunde, 112, (1968). 1503.

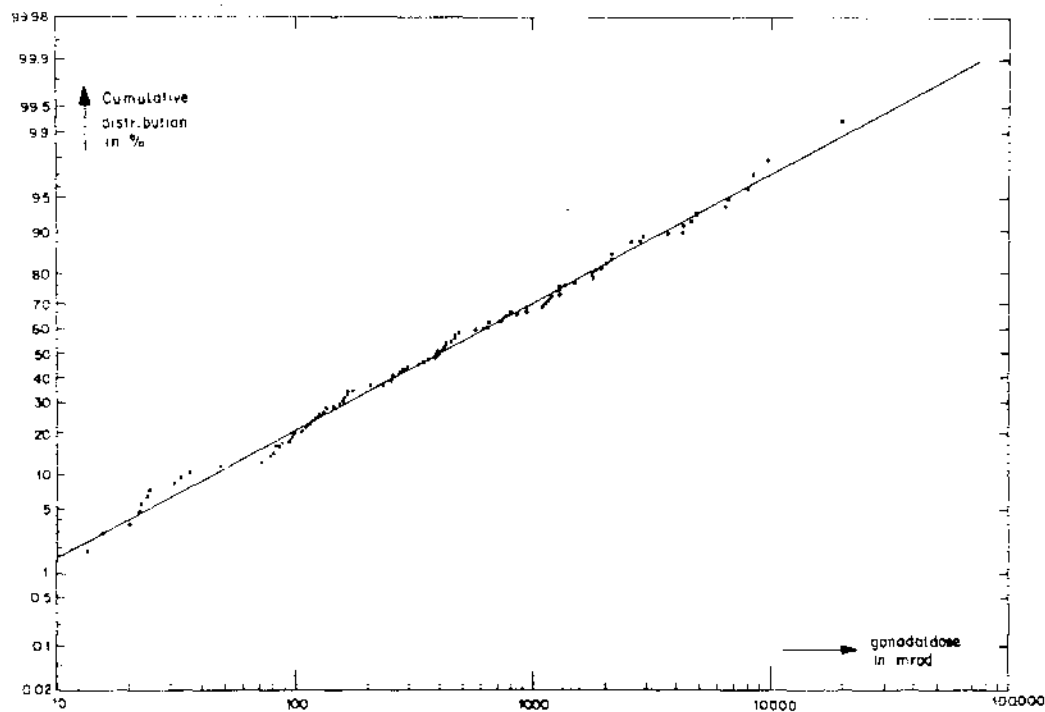


Fig. 1. Cumulative distribution of male gonadal doses from intravenous pyelography; hospital no. 3;  $d_{ij} = 1300$  mrad;  $s_{ij} = 2600$  mrad; median = 370 mrad; 102 patients.

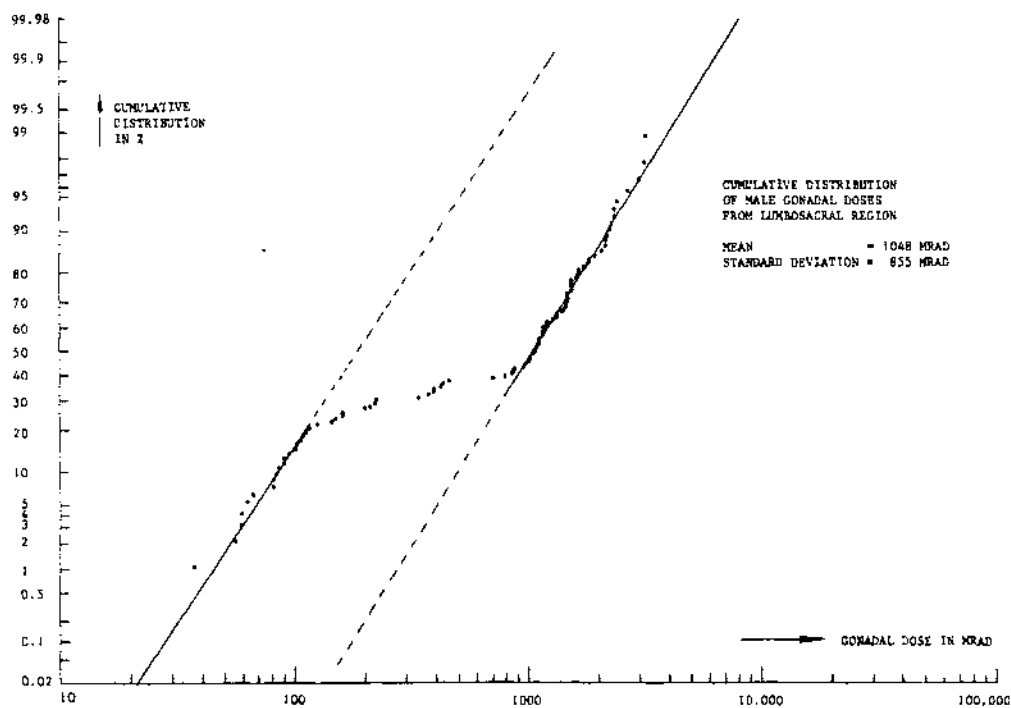


Fig. 2. Cumulative distribution of male gonadal doses from intravenous pyelography; hospital no. 4;  $d_{ij} = 1048$  mrad;  $s_{ij} = 855$  mrad; median = 1079 mrad; 93 patients. This examination type forms the unique exception on the log normal distribution found in all other types.

EVALUATION OF DIAGNOSTIC X-RAY CONTRIBUTION TO  
THE ANNUAL GENETICALLY SIGNIFICANT DOSE  
EQUIVALENT OF TAIWAN URBAN POPULATION

P.S. WENG  
Health Physics Section  
National Tsing Hua University  
Hsinchu, Taiwan 300  
Republic of China

C.Y. HUANG  
Atomic Energy Council of Executive Yuan  
Taipei, Taiwan 106  
Republic of China

Abstract

Thermoluminescent dosimeters (LiF-*teflon* discs) were distributed to clinics and hospitals using X-rays for diagnostic purposes in Hsinchu which is a medium size city of Taiwan with a total population of 213,735. The dosimeters were placed inside a small pocket stitched on a piece of cloth. Before irradiation the patient was covered with this cloth of which the location of the pocket was adjusted to be against the gonadal region of patient's body.

The detailed record of the irradiation conditions and the status of patients were provided by the clinics and hospitals concerned. Dosimeters were replaced and read in a one-month interval. Based on the formula given by the United Nations Scientific Committee of the Effects of Atomic Radiation the annual genetically significant dose equivalent was calculated with proper corrections for gonad dose of both sexes.

Since one half of Taiwan population is living in the cities nowadays, and surveys of medical radiation sources in Taiwan have been carried out twice since 1970, the annual genetically significant dose equivalent calculated for Hsinchu is extended to Taiwan urban population. It shows that the annual genetically significant dose equivalent of Taiwan urban population was in the range of 3 to 4 mrem in 1972.

Introduction

The hazard of radiation exposure of most concern has shifted from relatively high doses received by a few people to low doses received by a large segment of the population. The diagnostic X-rays belong as a contributor to the latter category. This paper is concerned with doses from diagnostic X-ray received by gonads of the Hsinchu population during an eight-month investigation period as recorded by thermoluminescent dosimeters.

Hsinchu is a medium size city of Taiwan with a total population of 213,753 (1972 census). Of all inhabitants 113,921 are male and 99,814 are female. The medical radiation sources surveyed in 1970 and again in 1972 show that there are 15 diagnostic X-ray units in operation, i.e., 70 X-ray units per  $10^6$  population.<sup>1</sup> Of the 15 hospitals and clinics equipped with diagnostic X-ray units, only 5 major hospitals or clinics have frequent use of the equipment. The other 10 hospitals and clinics have a few exposures taken every year. Based on this information, the

major hospitals and clinics were chosen for this investigation and the characteristics of their X-ray units are shown in Table 1.

Table 1. The Manufacturers, Applied Voltages, and Beam Currents of X-ray Units in Hsinchu Major Hospitals and Clinics.

Hospital No.	Manufacturer	Filter (mm Al)	Radiography			Fluorography		
			kVp	mA	sec	kVp	mA	sec
1	Arora	2.0	95	300	0.1	95	1	60
2	Shimadzu	2.0	60	300	0.5	60	2.5	180
3	Toshiba	2.5	100	500	0.05	70	3	60
4	Toshiba	2.5	70	200	0.25	70	2.5	120
5	Picker	2.5	75	100	0.05	70	3	60

#### Procedures

The 13 mm diam. x 0.4 mm thick discs of  $^7\text{LiF}$ -Teflon of Teledyne Isotopes were used because of the consideration of the maximum sensitivity. The minimum dose defined as three times the standard deviation of the background is 15 mrad. The tissue-equivalence of LiF-Teflon dosimeters enables meaningful estimates of dose in X-ray radiation from a single measurement of a dosimeter at the monitoring site. The response is independent of photon energy to within  $\pm 30\%$  down to 20 keV. In addition the LiF-Teflon discs are unaffected by extremes of humidity and environmental temperatures which do exist in Taiwan. Readout LiF-Teflon discs was performed on Teledyne Isotopes Model 7100 TLD instrument. Nitrogen was supplied to suppress the spurious thermoluminescence during readout procedure. The standard deviation was about 3.6%.

Two LiF-Teflon discs each were placed inside a small pocket stitched on a piece of cloth for radiography and fluorography, respectively. Before irradiation the patient was covered with this cloth of which the location of the pocket was adjusted to be against his-her gonadal region. The detailed record of the irradiation conditions and the status of patients were replaced and read in a one-month interval. Table 2 present the results of an eight-month investigation period in 1972.



Table 2. The Average Exposure per Capita due to Diagnostic X-rays Monitored with LiF-Teflon Discs at the Gonads.

Month	Hospital No.	mR/man-exposure		Number of Patients							
				Radiography				Fluorography			
		Radio-graphy	Fluoro-graphy	Age			Total	Age			Total
				<18	18-45	>45		<18	18-45	>45	
1	1	28	243	11	141	31	183	1	2	3	6
	2	15	303	15	28	13	56	1	4	4	9
	3	16		10	17	42	69				
	4	62	273	3	28	13	44		7	5	12
	5	25	242	5	46	5	56	5	18	14	37
2	1	32	218	32	261	42	335	2	2	2	6
	2	78	233	12	36	27	75		7	9	16
	3	11		2	35	63	100				
	4	7	453	1	103	24	128		13	6	19
	5	42	363	4	18	6	28	5	63	5	73
3-4	1	23	145	65	476	67	608		3	4	7
	2	48	414	17	90	50	157	1	16	15	32
	3	38			24	81	105				
	4	17	487	4	98	49	151		34	10	44
	5	37	212	10	77	22	109	44	229	56	329
5-6	1	30	342	83	1101	52	1236	1	2	6	9
	2	46	320	25	99	75	199	1	19	7	27
	3	18			5	22	27				
	4	22	256	1	207	82	290	1	54	18	73
	5	38	285	9	68	23	100	45	210	61	316
7-8	1	25	246	90	1320	48	1458	1	3	9	13
	2	40	381	30	110	86	226	1	20	8	29
	3	32		2	30	88	110				
	4	36	298	4	102	67	173		51	11	62
	5	40	240	13	86	31	130	46	253	61	360
Average or Total		32	298	448	4606	1109	6153	155	1010	314	1479

The annual genetically significant dose equivalent to the population is a measure of the genetic significance of the yearly dose equivalent received by the population's reproductive organs (gonads). To calculate this dose equivalent, one should consider the gonad dose and the future number of children expected by each member of the population as listed in Table 3.

Table 3. Age-Group and Average Expected Children Census

Age	Population (%)	Average Expected Children
0-18 years		
(M)	19.9	4.00
(F)	18.4	4.00
18-45 years		
(M)	23.6	2.30
(F)	21.7	2.30
> 45 years		
(M)	9.8	0.015
(F)	6.6	0.015

Table 3 was taken from population and birth rates census data released by the National Health Administration of Republic of China.

The genetically significant dose equivalent described here has the same meaning as that given in the report of the UNSCEAR.<sup>2</sup> The genetically significant dose equivalent can be calculated with the following formula:

$$D = \frac{\sum_k \sum_j (N_{jk}^{(F)} w_{jk}^{(F)} d_{jk}^{(F)} + N_{jk}^{(M)} w_{jk}^{(M)} d_{jk}^{(M)})}{\sum_k (N_k^{(F)} w_k^{(F)} + N_k^{(M)} w_k^{(M)})} \quad (1)$$

where

$D$  = annual genetically significant dose.

$N_{jk}$  = number of individuals of age-class  $k$ , subjected to class  $j$  exposure, i.e., either radiographic or fluorographic X exposure.

$N_k$  = total number of individuals of age-class  $k$ .

$w_{jk}$  = future number of children expected by an exposed individual of age-class  $k$  subsequent to a class  $j$  exposure.

$w_k$  = future number of children expected by an average individual of age-class  $k$ .

$d_{jk}$  = gonad dose per class  $j$  exposure of an individual of age-class  $k$ .

(F) = female.

(M) = male.

Since the radiation levels and exposure frequency from diagnostic X-rays are quite low, the number of expected children will be the same for individuals after irradiation as it was before. Therefore, for the purposes of these calculations  $w_{jk}$  will be assumed to be the same as  $w_k$ .

For calculation of male and female gonadal dose, the correction factors  $K_m$  and  $K_f$  can be used, where

$K_m$  = Depth dose at male gonads/skin exposure,

$K_f$  = Depth dose at female gonads/skin exposure.

The central axis depth dose factors used to calculate depth dose to gonads from air dose were  $K_m = 72\%$  and  $K_f = 11\%$ .<sup>3</sup>

During the investigation period, it was assumed that the number of exposures taken was equal to the number of patients being examined. According to the data provided by the hospitals and clinics, patients under age 18 were less than 10% of all patients concerned. Hence, it was assumed that the gonad dose was independent of age-class.

### Results and Discussion

The gonad doses thus obtained were as follows:

1. Radiography  
 $d(F) = 3.52$  mR  
 $d(M) = 23.04$  mR
2. Fluorography  
 $d(F) = 37.78$  mR  
 $d(M) = 214.56$  mR

For practical purposes, in X-ray diagnosis, an exposure of 1 R can be regarded as delivering to soft tissue a dose of 1 rad or a dose equivalent of 1 rem.<sup>4</sup> The annual genetically significant dose equivalent in Hsinchu City due to diagnostic X-ray only was 3.83 mrem in 1972.

The major cities in Taiwan are Keelung, Taipei, Taichung, Tainan, and Kaohsiung with a total population of 5,034,267 of which 2,683,264 are male and 2,351,003 are female. The number of diagnostic X-ray units is known.<sup>1</sup> Based on the data surveyed at Hsinchu City, it can be estimated that the annual genetically significant dose equivalent due to diagnostic X-rays in major cities of Taiwan was 3.64 mrem in 1972 while that of U.S.A. was 5 mrem. Since one half of Taiwan population is living in the cities nowadays, it is concluded that the annual genetically significant dose equivalent due to diagnostic X-rays of the Taiwan urban population was in the range of 3 to 4 mrem in 1972.

### References

1. WENG, P.S., CHENG, C.H., WEI, V.R., and LIU, K.Y., "Survey of Medical Radiation Sources in Taiwan," Health Physics, 23, 549-553 (1972).
2. Report of the United Nations Scientific Committee of the Effects of Atomic Radiation (1958, 1962).
3. BROYLES, M., NEELY, O., SLINKMAN, R., and TIERFELDER, C., "Evaluation of Television Contribution to Annual Genetically Significant Radiation Dose of the Population," Radiological Health Data and Reports, 7, 363-370 (1971).
4. "Protection of the Patient in X-ray Diagnosis," ICRP Publication 16, Pergamon Press, Oxford, England (1969).

## THE ROLE OF THE HEALTH PHYSICIST IN REDUCING MEDICAL RADIATION

by

Richard E. Gross  
Maria E. Marks  
Michael F. Audet  
James S. Benson

Division of Training and Medical Applications  
Bureau of Radiological Health  
Rockville, Maryland

### Abstract

Medical use of x ray continues to be the greatest manmade source of exposure to the United States population, in spite of substantial improvements in equipment and technique. Reduction of this exposure is possible through training and motivation of allied health professionals.

In addition to proposed x-ray equipment standards and current upgrading of technologist proficiency criteria, increased involvement of health physicists is needed. This paper examines the role of health physicists in the health care environment, stressing their potential for effecting exposure reduction through close working relationships with allied health professionals.

Health Physics emerged as a child of the nuclear age even before the first atomic bomb was exploded. As the use of atomic energy rapidly increased, so did the concern for the hazardous consequences of misuse and carelessness. Although the term Health Physicist may not have been used as it is today, scientists and engineers became increasingly involved in the problems of radiation protection and control. This was a logical development in view of the potentially high levels of radiation exposure to man, not only from the obvious threat of nuclear warfare but also from the rapid increase in radiation use by industry and research.

It is interesting to note, though, at the beginning of this nuclear age, medical radiation had already been in use for half a century, ever since Wilhelm Roentgen in 1895 accidentally discovered the mysterious rays that could penetrate matter including human tissue, and despite the extensive use of medical x rays for many years, only in the past few decades has there been an emerging medical specialization from the ranks of the radiation physicists.

In the scientific community, specifically that portion concerned with the uses of radiation, it is generally accepted that the medical and dental use of x rays constitutes the greatest single source of manmade radiation exposure to the general population. Yet, a relatively small percentage of the overall effort for safe, effective use of ionizing radiation is directed toward medical use. Since the Health Physicist has selected a profession which is dedicated to the prevention of unnecessary exposure to people, it follows that the Health Physicist would want to do something about the source of 90%<sup>1</sup> of the manmade radiation exposure. In 1971, there were approximately 3,000 members of the Health Physics Society. A statistical analysis of 2,862 members was done from application form information. An item asking for area of professional interest by first, second, and third choice resulted in the following figures: As a first choice, only 4.5% indicated the medical field, and ranked behind Health Physics (52.3%), Physics (8.7%), Biology (6.5%), Chemistry (5.7%), Engineering (4.8%), and other areas not specifically listed (6.0%). Even as the second and third choice, medicine was only 5.4% and 5.9% respectively.<sup>2</sup>

Dr. Dade Moeller, who conducted this analysis, offers this general comment:

"It would appear that a Society with such a high percentage of members with undergraduate backgrounds in physics would have many of the qualifications necessary for making a major impact in the field of medical radiation physics and the control of associated x-ray exposures. This is particularly true in light of the fact that the Society has such a high percentage of people with Doctoral degrees (18%) of whom well over a fifth are medical doctors and dentists. Undoubtedly, the reason that the Society has not been more active in this field is that most graduate programs in radiation protection, as currently organized, simply do not offer the opportunity for Health Physicists to receive the specialized training required for professional work in Medical Physics."<sup>3</sup>

We would add that perhaps an equally important reason is a general lack of communication between the health physics profession in general and the majority of those scientists (be they Physicists, Physicians, Biologists or whatever) presently working primarily in the area of medical radiation. There is a need for greater awareness of and active involvement in medical radiation exposure problems by Health Physicists.

A recent manpower study<sup>4</sup> estimates that in 1971 there were approximately 500 physicists working in the medical field and that double this amount would be required by the end of this decade.

Considerable public awareness and concern has been generated regarding radiation pollution of the environment by nuclear power plants. Much effort and money has been spent in the analysis of real and potential hazards of radiation exposure from nuclear power. But how does this compare to medical radiation exposure? The following statements from a study by the National Academy of Sciences (BEIR Report) can give one an idea of the relative magnitude of the two problem areas:

"Based on experience to date and present engineering judgement, the contribution to radiation exposure averaged over the U. S. population from the developing nuclear power industry can remain less than about 1 mrem per year (about 1% of natural background) and the exposure of any individual kept to a small fraction of

background. . . . ." [ Provided certain controls are maintained.]  
[Whereas] "In the foreseeable future, the major contributors to radiation exposure of the population will continue to be natural background with an average whole body dose of about 100 mrem/year and medical applications which now contribute comparable exposures to various tissues of the body."<sup>1</sup>

We derive great benefit from medical radiation in terms of public health and therefore we accept a certain degree of risk. This is a reasonable argument which will usually meet with little resistance and which is applicable to many areas of public concern such as the use of automobiles or the taking of drugs. The criteria, though, in each case, is to maintain an imbalance in the benefit/risk scale--that is, the benefits must outweigh the risks. There must be a constant effort to minimize the risks without adversely affecting the benefits.

The increase in numbers, types and complexities of medical radiation procedures challenges the allied health professionals just to keep pace with the medical and logistic considerations before one even considers the radiation protection problems which may be involved. We believe that increased efforts by Health Physicists who can develop good working relationships and communication lines with the medical professionals can further tip the scale to the benefit side.

The benefits of medical radiation are well known and need no elaboration. But just what are the risks? The answer, of course, is unknown for a particular individual involved in a particular type of medical radiation procedure. One can only talk about probabilities when large numbers of people are subjected to low levels of radiation, such as those used in the range of medical procedures. Even then, lack of sufficient human data precludes accurate predictions.

The BEIR Report contains this consensus regarding risks from radiation:

"Until recently, it has been taken for granted that genetic risks from exposure of populations to ionizing radiation near background levels were of much greater import than were somatic risks. However, this assumption can no longer be made if linear non-threshold relationships are accepted as a basis for estimating cancer risks. Based on a knowledge of mechanisms (admittedly incomplete) it must be stated that tumor induction as a result of radiation injury to one or a few cells of the body cannot be excluded. Risk estimates have been made based on this premise and using linear extrapolation from the data from the A-bomb survivors of Hiroshima and Nagasaki, from certain groups of patients irradiated therapeutically, and from groups occupationally exposed. Such calculations based on these data from irradiated humans lead to the prediction that additional exposure of the U. S. population of 5 rem per 30 years could cause from roughly 3,000 to 15,000 cancer deaths annually, depending on the assumptions used in the calculations. The Committee considers the most likely estimate to be approximately 6,000 cancer deaths annually, an increase of about 2% in the spontaneous cancer death rate which is an increase of about 0.3% in the overall death rate from all causes."<sup>1</sup>

Adopting the assumption of linear relationship between dose and biological damage then, the prudent course is to minimize all unnecessary or unproductive exposure. Two predominant types of unproductive radiation exposure in

the medical realm are radiation from procedures considered medically unnecessary, as in the case of patient self-referral, and radiation from improper performance of equipment and/or operator.

Both types of unproductive radiation exposure are sensitive and highly controversial issues that can be dealt with and eliminated without a decrease in medical benefit to the patient.

The use of radiation in medicine is usually divided into three general categories: Diagnostic Radiology, Radiotherapy, and Nuclear Medicine. Reference 3 indicates that Physicists working in medicine devote the greatest effort to Radiotherapy, with the remaining effort about equally divided between Diagnostic Radiology and Nuclear Medicine. The remainder of this paper is concerned specifically with the area of Diagnostic Radiology. Diagnostic x rays involve the greatest segment of the population.

Approximately one hundred and thirty million persons had one or more diagnostic x-ray examinations done in 1970\* with an estimated 660 million radiographic films being taken. Data taken from the 1970 X-ray Exposure Study is now being compared with the earlier 1964 Study. Preliminary figures seem to indicate that average exposure to the population may have been reduced. It is comforting to know that progress has been made. However, we believe that much greater reduction is still possible.

In the scope of a diagnostic x-ray examination, there are three main functions:

- 1) selection of the patient
- 2) performance of the examination
- 3) interpretation of the results

Health Physicists can, by active involvement, be the catalyst in improving the use of medical radiation on the public. By investigating and analyzing uses and abuses of medical radiation in these three functions of an examination, physicists can and must take decisive action to influence changes as required to minimize exposure to the population. One of the immediate areas that requires change is the chest x-ray screening procedures for cardiopulmonary disease. As you well know, this method of screening has been used for many years. It has recently been clearly identified as a procedure that should not be done. In view of the undesirability of using radiation without clear evidence of significant benefit, the liaison committees of the American College of Radiology, the American College of Chest Physicians and the United States Public Health Service issued on February 18, 1972, a policy stating:

"Community chest x-ray surveys among the general population as a screening procedure for the detection of tuberculosis, other pulmonary disease and heart disease are not productive and should not be done."

With reference to the detection of tuberculosis, the policy states that chest x-ray examinations should be restricted to individuals evidencing a positive reaction to the tuberculin skin test. Mass chest screening procedures should be eliminated also, because they are examples of the practice of patient

---

\*Preliminary estimates from the U. S. Public Health Service 1970 X-ray Exposure Study.

self-referral, that is the patient himself, not a qualified physician, decides he should have an x-ray examination. This is undesirable because the physician is much more capable of weighing the benefits to be derived against potential risk.

This policy statement has been effective in discontinuing mass screening surveys in some areas, but has not completely eliminated the situation. Health Physicists should investigate the practices in their own areas and individually and through their societies make sure that the mobile chest vans are located in those areas only where there is the possibility of high incidence of chest disease.

Another area which should be given strong consideration by the physicists is the extensive use of pre-employment chest x rays. This again is the use of x rays primarily for the detection of tuberculosis and is used extensively as a prerequisite in hiring of food handlers. This is another situation where the x radiation is applied to people indiscriminately, many of them very young, without prescription by a physician. Although there is admittedly a higher risk of the spread of tuberculin infection through food handlers, the criterion for a chest x ray should still be a positive skin test. By coordinating their efforts the physicists can affect this situation by investigating the reasons for pre-employment chest x rays and suggesting methods that would obtain the same results but without the use of radiation.

The second function of a diagnostic examination "performance of the examination" is where the scientific and technical knowledge of the Health Physicist can be put to very great use. The taking of a radiograph involves operation of complex equipment usually with many options and variables under the control of the operator. In many cases, although the operator may be knowledgeable and skilled in particular areas of health care, he or she may have little or no knowledge of the physical principles involved in the production of x radiation. If there is no real understanding of what happens when the button is pushed, this operator cannot effectively exercise the options available in order to obtain maximum benefit with the least radiation exposure to the patient.

Health Physicists can improve this situation by exercising their role as teachers and educators. They should become an integral part of the education of all those who apply radiation to other humans. If greater awareness of and concern for radiation safety can be instilled at the grass roots level through teaching in the medical, dental, and x-ray technology schools, the more difficult task of correcting poor practices can be considerably lessened.

A similar issue that Health Physicists could become involved in is that of credentialing of operators of radiation emitting equipment. In the United States presently there are only 3 States that have mandates requiring the licensure of x-ray machine operators. Although the licensing procedures vary in these States, all of them require by law that operators meet certain minimum educational requirements and possess a knowledge and skills required to deliver health care x-ray services with minimum amounts of radiation. Health Physicists can play an important role in this issue by becoming aware of the provisions in the licensure bills on the State and Federal levels and assume the responsibility to influence these bills with regard to reducing unnecessary exposure.

The third function "interpretation of the results" - that is making a diagnosis from a radiograph - is the responsibility of the physician. However, the physician's interpretation is limited by the quality of the radiograph which he interprets. Proper performance of the operator and the equipment are essential to quality radiographs. It is true that one can use poor radiation



safety practices and still obtain diagnostically acceptable radiographs. However, most methods used to minimize exposure will also improve the film quality. This is an important fact which much be emphasized to the users of diagnostic x rays. A false assumption by many is that a reduction of exposure to the patient is automatically accompanied by a reduction in quality or diagnostic information on a radiograph. Here again, the Health Physicist through educational methods, can help to modify such beliefs.

We have talked primarily about exposure to the general population meaning the patients undergoing radiographic examinations. With the development of higher energy equipment and complex procedures which require both a large number of successive radiographs and the presence of greater numbers of health care specialists, occupational exposure becomes an increasing problem. In many of these procedures, serious medical conditions exist and far outweigh radiation exposure hazards to the patient. However, the health professionals who perform these procedures on a daily or weekly basis, can be subjected to extremely high exposures unless constant safeguards provided through innovative techniques, equipment modifications and procedural analyses are established and maintained. For the professional, the cost of the exposure would of course not be compensated for by the benefit of the treatment.

A fourth area where Health Physicists may be very effective is in the area of assistance to the State and local radiation control programs. An effective dialogue between the two groups can result in an interchange of experience which can lead to the effective solution of common problems and assist the local radiation control programs in broadening their efforts beyond facility compliance with State codes. A mutual effort at working with users of medical x ray can lead to a significant reduction in the unnecessary exposure received by the population.

The challenge is there. We believe that the Health Physicist with a desire to enter the medical field can meet it. He need only take heed of the following principles:

1. Be sure that the problem being tackled is recognized as such by others involved.
2. Try to find solutions to exposure problems which also improve the quality of the radiological service.
3. Be sensitive to changes in attitudes of the medical profession toward recognizing the health benefits of improved radiological practices.
4. Be persistent but not with the "hard sell" approach.

In general, the medical profession is open to many changes in radiologic practice, even more so when the changes result in meaningful improvements in the quality of the service they perform. Sell yourself as a service and making the changes which they recognize as valuable to their department will make the elimination of unnecessary exposure easier to achieve.



#### REFERENCES

1. The Effect on Populations of Exposure to Low Levels of Ionizing Radiation - report of the Advisory Committee on the Biological Effects of Ionizing Radiations, Division of Medical Sciences, National Academy of Sciences, National Research Council, Washington, D.C. (report published and sponsored by the Environmental Protection Agency and the Department of Health, Education, and Welfare).
2. Statistical Information on Members of the Health Physics Society - by Dade Moeller, Ph.D., Health Physics Journal, April 1972 (study supported in part through research contract CPE-R-70-0034 between Harvard University and the Bureau of Radiological Health, FDA, DHEW).
3. Reproduced from Health Physics 22, 313-326 (1972) by permission of the Health Physics Society.
4. Status and Future Manpower Needs of Physicists in Medicine in the United States - by the Joint Committee on Manpower Needs in Medical Physics, July 1971 (unpublished). A study by members of the American Association of Physicists in Medicine (AAPM) and the American College of Radiology supported by the Bureau of Radiological Health, FDA, PHS, DHJW contract no. PH 86-67-202.

DENTAL TECHNIQUE NORMALIZATION:  
DEVELOPMENT AND PILOT TEST

Kenneth L. Travis, Northeastern Radiological Health Laboratory, 109 Holton Street, Winchester, Massachusetts 01890; John R. Stanton, New Hampshire State Department 03301; James E. Hickey, Rhode Island Department of Health, Davis Street, Providence, Rhode Island 02908; Bruce M. Burnett, Northeastern Radiological Health Laboratory, 109 Holton Street, Winchester, Massachusetts 01890

Abstract

A two phase program designed to help control excessive radiation exposure in dental radiography is described and the results of a two state pilot test presented. The first phase is a data collection system for measurement of radiation exposure from dental x-ray machines and the identification of machines producing excessive exposures. It was found that 54% of the machines surveyed have outputs within "acceptable" exposure limits. Personal in office visits to the other 46% constitutes the second phase of the program. The cause of the excessive exposure was identified and proper radiographic techniques were presented. The result was a reduction of 80.4% in the average exposure at end of cone (mR/film).

Introduction

The Bureau of Radiological Health, FDA, in conjunction with the radiation control agencies of several States has developed and tested a program using thermoluminescent dosimeters (TLD's) to help control excessive radiation exposure in dental radiography. The magnitude of this problem has been discussed and documented in literature. The skin entrance exposure for the average adult dental bite-wing examination has been shown to vary tremendously from facility to facility.<sup>1</sup> The PHS X-ray Exposure Study of 1970<sup>2</sup> indicated that dental exposures range from 20 to 13,000 mR even in facilities which comply with appropriate local State and federal radiation standards. These programs have traditionally been oriented to equipment and facility regulation and reduction of occupational exposure. As a result the user and his impact on the patient dose has not been directly affected. This program has been developed to deal directly with the user to improve his performance, and lower patient x-ray exposure.

The program described in this paper has been designed for use by radiation control agencies and provides methodology for reducing population exposure from dental radiography to levels in accord with good radiographic technique.

This program consists of two phases: identification and action. The major emphasis of this discussion concerns the data collection system for measurement of radiation exposure from dental x-ray machines and the identification of machines producing excessive exposures. An action program designed to reduce these excessive exposures will also be presented. The development of the system and the results of a pilot test in two states are presented.

### Methodology

The thermoluminescent dosimeter in the form of lithium fluoride (LiF) chips has, in recent years, found many applications in the field of radiation measurement. This project was undertaken to determine the effectiveness of mailing TLD's in a data collection and screening system for dental x-ray exposures. Specifically we wished to determine whether the information collected by such a method could be used to determine and set priorities in such a way as to accomplish the greatest exposure reduction with the most efficient use of time and resources of state radiation control personnel.

### Acceptable Dental Exposure Range

As a first step in the development of this program, it was necessary to determine acceptable exposure levels for diagnostic dental radiographs. A search of the literature was conducted and a laboratory investigation performed to establish these levels.

Travis and Hickey<sup>3</sup> have empirically determined values for the exposure at the tip of the cone which are useful as a guide to the production of diagnostic quality dental radiographs. They reported a single tip of cone exposure value, capable of producing a diagnostic quality dental radiograph, for selected operating kVp values.

It is known, however, that for a given kVp and properly filtered x-ray beam, there exists a range of tip of cone exposure values which produce radiographs of acceptable diagnostic quality. In order to determine this range, laboratory studies were conducted on a dental phantom consisting of the mandibular and maxillary sections of a human skull imbedded in a transparent, non-granular plastic which has the same absorption and secondary radiation-emitting characteristics of living tissue. The part of the phantom that would correspond to the inside of the mouth was notched to accept a standard (1-1/4" x 1-5/8") dental x-ray film positioned behind the teeth to represent the bite-wing examination.

A series of radiographs were then produced at selected kVp settings from 45 to 90 kVp to represent the operating kVp found on most dental units in dental offices. The exposure range at each kVp setting was broad enough to produce films too light to be of diagnostic quality at the lower exposure and too dark to be of diagnostic quality at the upper exposure levels. The tip of cone exposure was measured using Harshaw LiF (TLD-100) chips and read on an Eberline Model TLR-5 Reader. Exposure values were recorded for each radiograph. A panel of 24 dentists read these radiographs and selected those they felt were diagnostically acceptable. All radiographs were produced using speed group "D" dental film and were processed using fresh solutions and the time and temperature developing technique recommended by

the film manufacturer. Speed group "D" film was used because of its wide acceptance by the Dental Profession, and its ability to produce high quality diagnostic radiographs at reduced patient exposure. Figure I illustrates the range of cone tip exposure that yields diagnostic radiographs as a function of operating kVp. The upper curve is the maximum exposure. With proper development, overexposure results in a black film and underexposure produces a film which is too light. Figure I clearly indicates that there is a finite exposure range which will produce dental radiographs of acceptable diagnostic quality.

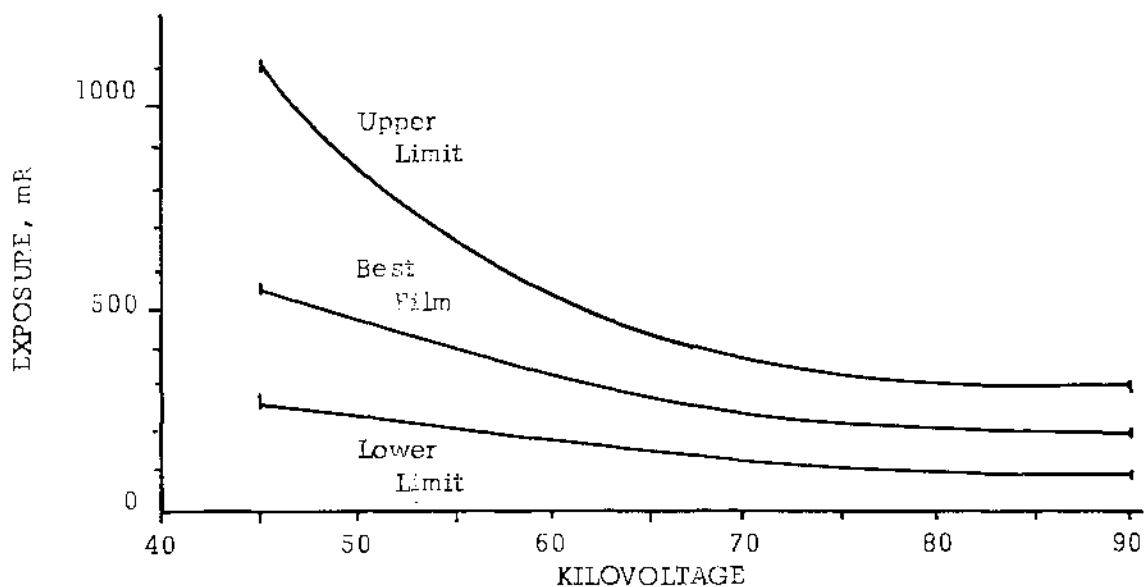


FIGURE 1 DENTAL EXPOSURE RANGE TO PRODUCE RADIOGRAPHS OF DIAGNOSTIC QUALITY

#### Identification Phase

The program described in this paper was developed to answer two questions: 1) how many dental x-ray machines used in the routine practice of dentistry fall within the acceptable exposure range; and 2) how can these specific machines producing excessive radiation output be efficiently identified so corrective action can be taken? To find the answers, Radiation Control Program Directors in two states, Rhode Island and New Hampshire, were invited to participate with the Northeastern Radiological Health Laboratory in the development and testing of a dental inspection program which made use of a mailable card system to obtain cone tip exposures with thermoluminescent dosimeters.

A 5" x 7" card (folded to 3-1/2" x 5") was designed to carry three TLD's. An identification number, such as the machine registration number, was written on the card before mailing to relate the exposure to a specific machine. A card was sent for each x-ray machine registered by the dentist in the participating states. Instructions and a sketch indicating how to properly make the end of cone exposure were printed on the card and spaces for the exposure data were provided. The data collected includes the radiographic

technique and film type normally used. A cover letter of instructions was included briefly describing the program and inviting the dentist's participation. Upon receipt of the card, the dentist was requested to expose the TLD's attached to the card and return the card or cards, with TLD's still attached, in a self-addressed stamped envelope to the Health Department for reading and evaluation. For this pilot test all TLD's were read at the Northeastern Radiological Health Laboratory.

#### Action Phase

Once the dental exposures had been determined, an action program to correct deficiencies was begun. In this study, the individual State Radiological Health Program Directors identified the exposure level at which corrective action was to be taken. This action point could be a constant value for all operating kVp or a sliding scale depending on the exposure range required for a given kVp. Any machine producing an exposure greater than required for diagnostic radiographs based on the operating kVp could be visited. Priority in this system normally was given to those facilities having the potential for the greatest exposure reduction to the population.

Field visits to offending x-ray units showed that the chief causes of excessive radiation are: 1) use of slow speed film; 2) overexposure and underdevelopment (sight development) of film; or 3) use of depleted developers. During a field visit, the inspector identified the cause of the excessive exposure, informed the dentist of the problem and took the necessary corrective action. The success of this action phase of the program lay in the approach the inspector used during the visit. The dentist was instructed in how to produce quality radiographs at lower patient exposure and was left with an improved radiographic technique.

During the inspection visit the x-ray machine output was adjusted to produce an end of cone exposure which fell within the acceptable exposure range presented in Figure 1. The inspector made the necessary adjustments of the exposure time, x-ray tube current (mA), voltage (kVp), or installed additional aluminum filtration to obtain the required exposure. A recommended technique was left for each machine after it had been adjusted. The new technique would produce an acceptable radiograph for all common dental exams when processed in fresh developing solution at 68° for 5 minutes.

The value of the new technique was reinforced by asking the dentist to witness a demonstration. Two bite-wing radiographs were taken of the dental phantom; first using the equipment, exposure technique, film and processing technique routinely employed by the dentist; the second radiograph was made using speed group "D" film and the newly recommended exposure and processing techniques. Before the films were processed the dentist's darkroom was checked to insure that adequate conditions for film development were present. If not, the inspector would develop the film produced by the new technique, using fresh solutions in small portable developing tanks which he carried. The film which had been exposed using the pre-adjusted equipment and timer settings specified by the dentist, was developed according to the dentist's customary manner of development. The group "D" film taken with the correct exposure factors was developed by the time and temperature technique specified by the manufacturer of the film.

Films exposed to the proper amount of radiation, as determined in Figure I, and developed according to the recommended time and temperature technique proved to be consistently superior to those improperly exposed and developed. The fact that the dentist could observe a film developed in his own facility which was of both low patient exposure and good diagnostic quality, served to insure that the recommended techniques left by the inspector would be followed.

### Results

The dental profession in the two pilot test States have responded enthusiastically to this new program as indicated by both the number of cards returned and the active endorsement by the Dental Associations in both States. The return of the TLD cards ranged from 75% in New Hampshire with a modest follow-up effort, to 99% in Rhode Island with a comprehensive follow-up effort. The results from both States were compiled within a 3-month period.

State Radiation Control Program Directors' acceptance of this program is primarily due to the ease and convenience with which it can be performed without significant increased cost of program personnel and time.

### Identification Phase

The exposure data obtained by the cards was compared to the exposure ranges shown in Figure 1. Table I lists by operating kVp, the number of machines that fell within the exposure range prescribed. Fifty-four percent of the dental x-ray machines surveyed in this two State pilot study had outputs which fell inside the acceptable range. Forty-six percent of the machines surveyed were found to produce excessive radiation and contribute to the unnecessary exposure of the population even though with few exceptions, these dental facilities comply with existing local, State, and Federal regulations.

TABLE I

Results of the TLD Card Survey of Two States

Operating kVp	Acceptable Range mR	Total No. of of Machines Surveyed	Machines Inside Range		Machines Outside Range	
			No.	%	No.	%
90+	100-315	179	124	69.3	55	30.1
70-89	115-330	152	92	60.5	60	39.5
60-69	190-570	410	200	48.8	210	51.2
50-59	250-800	93	37	39.8	56	60.2
	Total	835	453	54.3	381	45.7

### Action Phase

The action phase of this program has been completed in New Hampshire, and the results are encouraging. The point at which action was initiated in New Hampshire was 800 mR. A total of 105 dental x-ray units were found to lie above this limit and were visited by inspection personnel. The average

exposure at end of cone, mR/film, was reduced by 80.4% for the x-ray machines visited during the action phase. A total of 269 dental x-ray machines were surveyed in the identification phase of the project in New Hampshire, the net reduction in the average exposure at end of cone, mR/film, for all of the machines involved in the study is a very respectable 56%.

#### Conclusions

The dental profession in the two pilot test States responded enthusiastically to this new program as indicated by both the number of cards returned and the endorsement by State Dental Association in both States. Radiation Control Program Directors' acceptance of a card system has been accomplished through the ease and convenience with which the program is conducted without significant increased cost of program personnel and time. Based upon the results of this pilot study it is estimated that one Radiological Health Specialist working one-quarter time and one Secretary working one-half time can complete a 1,000 unit program in 3 months. The estimated cost of such a survey of 1,000 units is \$4.50 per machine. This cost includes the purchase of all needed equipment, printing, postage and personnel. This cost would be greatly reduced in those States already having TLD capabilities.

It has been demonstrated that TLD's sent through the mail can be used as effective screening devices to identify dental x-ray equipment producing excessive patient exposures. With this information, Radiation Control Agencies are able to plan their program efforts to lower exposure to the population from dental radiography according to a defined priority system.

#### Bibliography

1. "A Survey of Dental X-ray Equipment and Radiological Practices in Nassua County, New York", by L. Pental, et al., Health Physics, Vol. 20, pp. 59-71, January, 1971.
2. Personal conversation with the Chief, X-ray Exposure Study Branch, BRH, FDA, 1973.
3. "A State Program for Reducing Radiation Exposure from Dental X-ray Machines", K. L. Travis, B.S., F.A.P.H.A., and J. L. Hickey, M.S. American Journal of Public Health, Vol. 60, No. 8, August, 1970.



## NASHVILLE DENTAL X-RAY STUDY

Orlen N. Johnson, D.D.S., C. Larry Crabtree, D.D.S., Division of Training and Medical Applications, Bureau of Radiological Health, 5600 Fishers Lane, Rockville, Maryland 20852; S. Julian Gibbs, Division of Radiation Therapy, Vanderbilt University, Nashville, Tennessee 37203.

Division of Training and Medical Applications  
Dental Branch

U.S. Department of Health, Education, and Welfare  
Public Health Service  
Food and Drug Administration  
Bureau of Radiological Health

### I. INTRODUCTION

In January of 1972, personnel from the Bureau of Radiological Health, U.S. Public Health Service and members of the Department of Radiology, Vanderbilt University, met with representatives of the Nashville, Tennessee Dental Society to discuss dental radiological health practices. When comparing data from the 1964 and 1970 National X-ray Exposure Studies (XES)<sup>1,2</sup> it was pointed out that the use of slow and intermediate speed dental film decreased substantially, yet the average incident skin exposure remained disproportionately high. In 1964 about half of the dentists were using the slow and intermediate film (speed group A, B and C) and 50% were using fast film (speed group D). In 1970 only 20% of the dentists used slow or intermediate film while 80% were using the fast film. However, the mean incident skin exposure in 1964 was 1,138 mR, whereas in 1970 it was 910 mR, a decrease of only 20%.

The group raised the following questions: "What are dentists doing which contribute to overexposure of their films and their patients? Are they using good equipment, following recommended procedures, and exercising good judgment? Furthermore, can the dentist be motivated to correct any deficient findings or improper radiographic practices through an educational approach?"

In an effort to answer these questions, a pilot project was initiated by Vanderbilt University, under contract with the Bureau of Radiological Health, and in cooperation with the Nashville Dental Society. An effort would be made to test the effectiveness of a non-regulatory educational approach to dental x-ray exposure reduction. The primary objective of the project would be to motivate the participating dentist toward improved dental radiological practices with emphasis on efficient and effective use of X-radiation.

The project was divided into three phases. Phase I was a detailed assessment of radiology practices and equipment in a random sample of private dental offices in the Nashville area. Phase II consisted of a continuing education program individualized to each office. Specific recommendations based on Phase I information were given to each participating dentist for modification of his x-ray equipment and radiology practices to achieve optimum diagnostic quality with minimum exposure. Phase III was a follow-up survey, one year after Phase II, to determine if this educational effort to improve radiographic practices was successful.

Phases I and II of the project have been completed and are the subject of this report. Data are being further evaluated to develop a profile of dental radiographic practices in Nashville. Such information will be useful in setting priorities for corrective action programs locally and possibly in indicating nationwide problems. A more complete report will be available from the Bureau of Radiological Health in the near future.

## II. METHODOLOGY - PHASE I

Phase I consisted of a physical survey of 72 dental facilities selected at random from a total of 209 offices operated by 250 members of the Nashville Dental Society conducted during the summer of 1972. Offices having intraoral x-ray equipment were contacted by telephone and appointments were made. The surveyors were trained by members of the Radiology staff at Vanderbilt University and members of the Bureau of Radiological Health. The two man survey teams collected such information as: number of x-ray machines; cone types; speed of intraoral dental films; processing procedures; and, film mounting and viewing. Physical measurements were made of the beam size, filtration, exposure/film, and stability of the tube head.

## III. RESULTS AND DISCUSSION - PHASE I

A total of 72 dental facilities were surveyed, involving 80 dentists and 110 x-ray machines. The following tables show some of the more significant findings.

Table 1.

<u>BEAM SIZE</u>	
<u>INCHES</u>	<u>MACHINES</u>
2.0-2.4	15
2.5-2.8	85
2.9-3.1	8
3.2+	2
TOTAL	<u>110</u>

The beam diameter was measured by making an exposure on direct print paper. Results from Table 1. show 100 machines have a beam diameter of less than 2.8 inches. Only 2 of the 110 machines had a beam diameter larger than the Tennessee State regulation of 3.0 inches. The largest beam diameter recorded was 3.4 inches.

Table 2.

## OPERATING kVp BY CALCULATED HVL (mm Al)

kVp	Machines	HVL (mm Al)		
		< 1.1	1.2-2.0	> 2.1
< 50	1	--	--	1
50-70	75	3	47	25
> 71	34	--	9	25
TOTAL	110	3	56	51

The operating kVp was determined by the setting on the dial of the x-ray machine used by the dentist for a periapical exposure. The half value layer (mm Al) was determined using the Organ Dose Index System<sup>3</sup>. Table 2. shows that 76 machines operate up to 70 kVp and 34 machines operate over 70 kVp. Twelve of the 110 machines were deficient in filtration according to the proposed Federal Standards<sup>4</sup>. Three 50-70 kVp machines had a HVL of  $\leq 1.1$  mm Al and nine machines operating over 70 kVp only had a HVL of less than 2.0 mm Al.

Table 3.

## FILM SPEED - FACILITIES

	Speed Group				Total
	B	C	D	Unk.	
Periapical	1	2	71	--	74*
Interproximal	1	2	70	1	74*

\*Two facilities used two different film speeds.

Table 3. shows that over 95% of the dental offices in Nashville are using speed group D dental film. This is considerably better than the national average of 80% as reported in the 1970 National X-ray Exposure Study. The two offices using speed group C film also used speed group D.

Table 4.

## AVERAGE mR/FILM

All	542
Sight Develop	730
Occ. Sight Develop	536
Never Sight Develop	404

The exposure per film at the end of the position indicating device (cone) was determined by making three exposures on low energy dosimeters using the Organ Dose Index System<sup>3</sup>. Table 4. shows the average incident skin exposure for all 72 facilities was found to be only 542 mR. This is low compared to the national average and is to be expected when one considers the high percentage of dentists using the fast film. However, it was also noted that 43 of the 72 offices sight-developed, 24 routinely. These "sight developing" dental offices are overexposing and underdeveloping dental films. Consequently, the average exposure per film was calculated for the sight developing offices, the occasional sight developing offices and the never sight developing offices. The results are seen in Table 4; sight developing offices 730 mR, occasional sight developing offices 536 mR, and the never sight developing offices only 404 mR.

This is felt to be significant because apparently by just properly exposing the film and changing the processing technique to one of time-temperature, the sight developing dental office can reduce the exposure to its patients by at least 45%.

Table 5.

mR/FILM (INTERPROXIMAL) BY kVp

mR/film	Machines	kVp			
		< 50	50-60	61-70	> 70
55-200	23	--	3	10	10
201-400	37	--	--	23	14
401-600	17	--	--	10	7
601-999	15	--	1	13	1
1,000+	18	1	4	11	2
TOTAL	110	1	8	67	34

Table 5. is a cross tabulation of mR/film by kVp listed by x-ray machine. Seventy-seven of the 110 machines were producing an exposure of 600 mR or less. The exposures ranged from less than 100 mR to slightly over 2,500 mR. Although the average exposure was 542 mR, the median exposure was 404 mR. The kVp was determined by the setting on the dial of the x-ray machine used by the dentist for an interproximal exposure. Table 5. indicates that in general, the dentists operating higher kVp equipment are giving their patients less skin exposure than dentists using lower kVp techniques. It is well established that incident skin exposure increases as kilovolt peak is reduced, but it is not known exactly what the effect is on the integral absorbed dose to the patient.

Table 6.

POSITION INDICATING DEVICES (CONES)

Type	Machines
Pointed	46
Open lined	50
Open unlined	13
Unknown	1
TOTAL	110

A breakdown of position indicating devices (cones) on equipment showed 46 pointed cones; 50 open lead-lined cones; 13 open unlined cones; and 1 unknown. There is more scattered radiation associated with pointed cones than with open end cones. Tennessee State regulations require that after July 1, 1973, all x-ray apparatus designed for intraoral radiographic use shall be equipped with open end cones (cylinders).

Table 7.

<u>TUBE DRIFT AND VIBRATION</u>		
	<u>Drift</u>	<u>Vibration</u>
	<u>Machines</u>	<u>Machines</u>
Yes	34	18
No	76	92
TOTALS	<u>110</u>	<u>110</u>

Tube head drift was determined by fully extending the tube head and noting any drifting or pulling back of the head from its set position. Thirty-one percent demonstrated drifting. Drifting can cause inferior radiographs by cone cutting and/or improper angulation of the primary beam.

Vibration was determined by extending the tube head to near maximum and noting any vibrations. Sixteen percent of the tube heads vibrated. Excessive vibration can cause increased penumbra and blurring of the radiograph.

#### IV. CONCLUSIONS

After analyzing the preceding data, one can conclude that in general the dental x-ray equipment in Nashville, Tennessee, meets recommended standards. Most dental x-ray machines have properly collimated and filtered beams. Within the near future all dental x-ray equipment in Tennessee will have open end cones (cylinders). Almost all of the dentists are using the fastest speed film available. Yet the incident skin exposure is higher than necessary because the dentists fail to use proper processing techniques. Instead of overexposing films and using a sight developing technique, dentists should properly expose the films and use the recommended time-temperature processing procedure.

#### V. METHODOLOGY - PHASE II

At the completion of Phase I, the results from each survey form were read and reviewed. Dental radiology experts from Vanderbilt University and the Bureau of Radiological Health took one facility at a time and in detail listed the deficiencies in the office on a Check List Form. Then recommendations were made for improving radiographic practice on a Reporting Form. The report also included the approximate cost involved to bring the facility up to the recommendations.

Phase II consisted of a second visit to the dental office by a consulting team composed of one of the dental students and a dentist knowledgeable in dental radiology from Vanderbilt University or the Bureau of Radiological Health. As in Phase I, the telephone was used for making definite appointments. It was requested that the dentist and all his staff attend this meeting.

While in the dental office, the consulting teams performed the following: presented and reviewed the findings of the initial survey; discussed the written recommendations for any changes to improve the dentist's radiographic practice and left a copy of the "Reporting Form" and "Survey Check List"; answered all questions from the facility personnel; gathered from the dentist certain judgmental factors, such as criteria and frequency for making examinations; collected missing or questionable information from the initial survey; and left with the dentist selected reprints and pamphlets for future reference.

Phase III was started in July of this year with a follow-up visit to the original 72 dental offices where the equipment was resurveyed. Analysis of the data will determine the effectiveness of the project in motivating the Nashville dentists to correct any machine deficiencies and to improve their radiographic practices. If the project proves successful, the Bureau may assist other interested dental societies, universities, and health agencies in implementing similar educational programs.

#### BIBLIOGRAPHY

1. Gitlin, Joseph N. and Lawrence, Philip S., Population Exposure to X-rays U.S. 1964, U.S. Public Health Service Publication #1519.
2. Gitlin, Joseph N., Preliminary Estimate from U.S. Public Health Service, 1970 X-Ray Exposure Study, presented at American Roentgen Ray Society, Boston, Massachusetts, September 29, 1971.
3. Nationwide Evaluation of X-Ray Trends, Organ Dose Index System, Radiographic, Instruction Manual, U.S. Department of Health, Education, and Welfare, PHS, FDA, September, 1972.
4. Federal Register, Volume 37, Number 158, page 16466, Tuesday, August 15, 1972.

## ABSORBED DOSE TO SELECTED INTERNAL ORGANS

### FROM TYPICAL DIAGNOSTIC EXPOSURES \*

J. W. Poston and G. G. Warner  
Health Physics Division  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee 37830

Estimates of dose to internal organs from exposure to diagnostic x-ray beams are usually based on measurements of the entrance and exit dose and/or the use of depth dose curves for homogeneous media. This paper presents the results of a series of Monte Carlo calculations which mock-up typical diagnostic x-ray procedures. Results are presented for 22 internal organs as well as red and yellow bone marrow for two typical procedures. The calculations employ measured x-ray spectra from 45 kVp, 1-mm Al to 105 kVp, 2-mm Al and are for a field size of 14" x 17". In addition, depth dose profiles in various sections of the heterogeneous phantom are presented for each x-ray beam.

#### Introduction

It is well established that x-rays, particularly medical and dental x-rays, contribute the largest exposure to the population of any man-made source of ionizing radiation. The fundamental objective of the medical use of radiation is to obtain optimum diagnostic information with minimum exposure to the patient, and the radiological personnel concerned, and the general public. However, the problems posed when one attempts to estimate the doses received by various organs of the body from a medical exposure are among the most difficult problems the radiological physicist must face. The geometrical complexities and inhomogeneities of the body and the various organs make experimental simulation of the human body extremely difficult and usually unsatisfactory.

Monte Carlo techniques currently in use on high-speed digital computers have greatly facilitated the solution of these complex problems. These techniques have gained wide use in the field of radiation protection because the method allows one to perform an experiment by use of the computer. Many experimental arrangements and physical parameters, which can be described mathematically, can be operated on by the computer to produce the desired results.

---

\*Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corporation.

A Monte-Carlo-type computer program has been developed at the Oak Ridge National Laboratory, which can be used to estimate dose due to external photon beams typical of those employed in x-ray diagnosis.

#### Description of the Study

Two diagnostic x-ray procedures were simulated on a computer for these studies. Eight computer runs consisted of a set of exposures each of which simulated a chest x-ray and eight runs consisted of a set of exposures each of which simulated a G. I. x-ray.

The target for these studies was an anthropomorphic phantom, which may be considered to be two coexistent phantoms. One is that of an adult human body and some of its internality. It has been variously described<sup>1,2,3,4,5</sup> and in its present form represents a worthwhile target for these studies. It contains 23 internal organs including gonads, lungs, and four parts of the G. I. tract; it has ten skeletal parts with provisions for red and yellow marrows; and there is skin and there is tissue which includes muscle. The bone marrow and the bone are mixed homogeneously in the skeleton of this phantom.

The other phantom is called a geometric phantom. Whereas, in toto, it has the same outer dimensions and the same mass and composition of the human phantom, it is divided into dose regions by cutting planes and curves. For example, the trunk of the phantom has five layers, is divided into five concentric cylinders, and is cut by four vertical cross planes. This results in 85 subregions in which depth dose may be determined.

Both phantoms are heterogeneous by virtue of their composition which consists of 3 distinct media: tissue, lung, and bone with their concomitant densities and attenuation and absorption properties.

Each of the 16 exposures consisted of a collimated 36 cm x 44 cm (14 x 17 in) beam of 120,000 parallel photons incident on the posterior (P-A) of each phantom. In these calculations the source input was a set of eight measured x-ray energy spectra due to Epp and Weiss<sup>6</sup> at the Sloan-Kettering Institute for Cancer Research in New York City. The spectra range from 45 kVp, 1-mm Al filtration to 105 kVp, 2-mm Al filtration. The energy of each photon was determined from a normalized distribution of relative photon fluences per unit energy interval between 10 keV and 102 keV. Monte Carlo methods were used to follow the transport of each photon through the phantoms, determining the scattering angles, absorption sites, etc., and permitting the estimates of absorbed dose in units of absorbed dose per unit incident exposure (rad/R). The absorbed dose was calculated in the internal organs of the



adult human phantom as well as in the volume elements of the geometric phantom.

Depth dose distributions in the trunk for the simulated chest x-ray exposures are presented in Figure 1. These data are for 36 x 44 cm beams incident on the posterior of the phantom. Illustrated are the effects on dose of the reduction in average energy of the beams and the attenuation of the beams as they pass through the phantom. For the high energy beam, the dose from the back to front drops off by a factor of 10. For the low energy beam, it is reduced by a factor of about 130. The average dose in the first 2 cm of tissue for the 105 kVp beam is 1.7 times higher than for the 45 kVp beam. Near the exit surface the dose for the 105 kVp is 23 times higher than that for the 45 kVp beam. Data for the simulated G. I. exposure are similar in magnitude and ratio negating the necessity to discuss these results in detail.

Often the radiologist uses the dose at 5 cm depth as an indicator of the average dose to the red bone marrow. Table I presents such a comparison of the data derived from the simulated chest exposures. The last column of the table is the ratio of the red bone marrow dose to the 5 cm depth dose and shows that the indicator mentioned above might lead to a 40% error.

Figure 2 shows dose to selected organs for a simulated chest x-ray as a function of average beam energy. There appears to be three pairs of curves. The pair with the highest dose represents organs definitely within the beam. The next highest pair, the upper large intestine and the thyroid gland, represents organs outside but near the edge of the beam. The last pair represents organs definitely outside the beam.

Lowering the beam location to a position which simulates a G. I. exposure caused a 50% increase in the dose from the low energy beam and a 20% increase in dose from the high energy beam to the red bone marrow. This result is due to the exposure of the pelvis which contains about 32% of the red bone marrow and was outside the beam during the simulated chest exposure. The dose to other organs, such as the uterus and the upper and lower large intestine, was increased by at least a factor of three at this lower exposure.

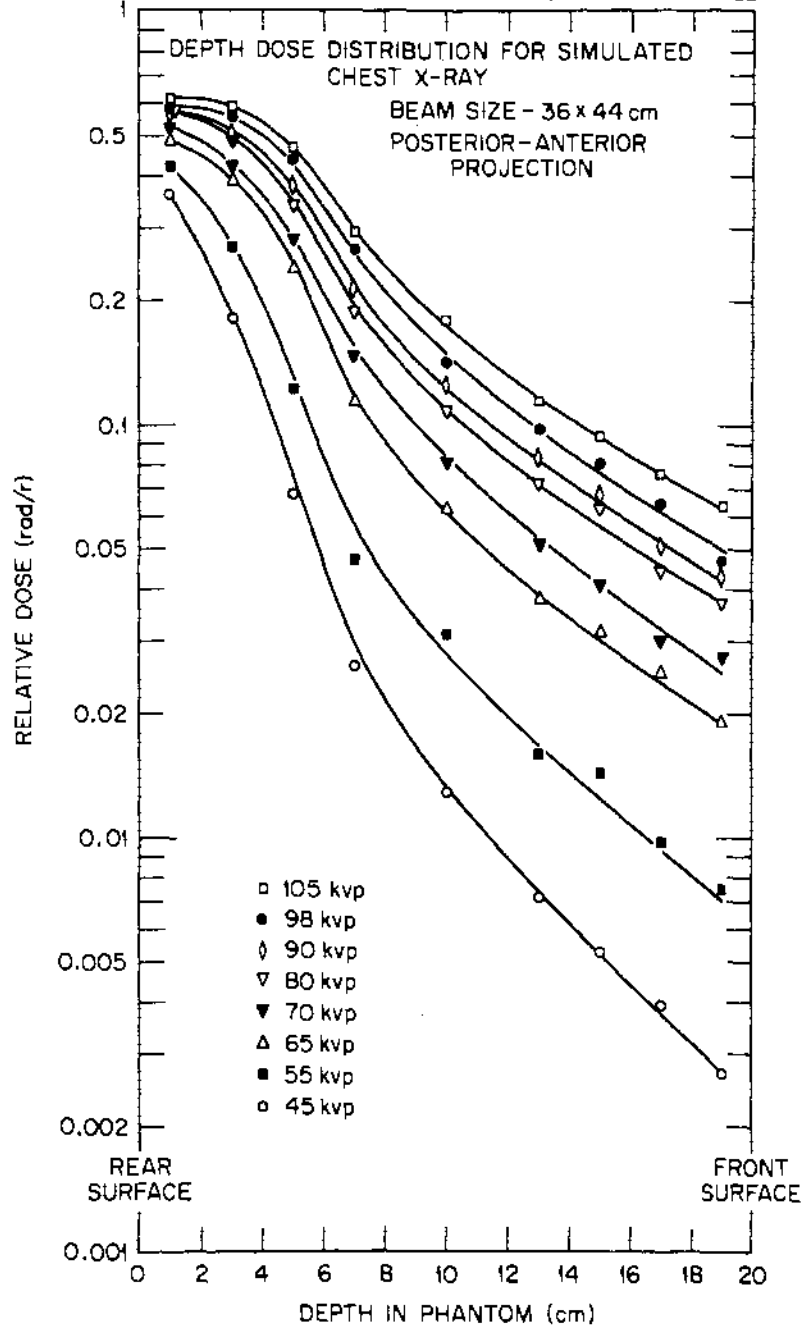
This study represents only a beginning in that it demonstrates the versatility of Monte Carlo techniques in the simulation of diagnostic procedures. The computer programs used allow various source descriptions, such as point sources located at various source to skin distances, divergent beams, etc. In addition, the beam size, shape, and angle of incidence on the phantom may be specified.

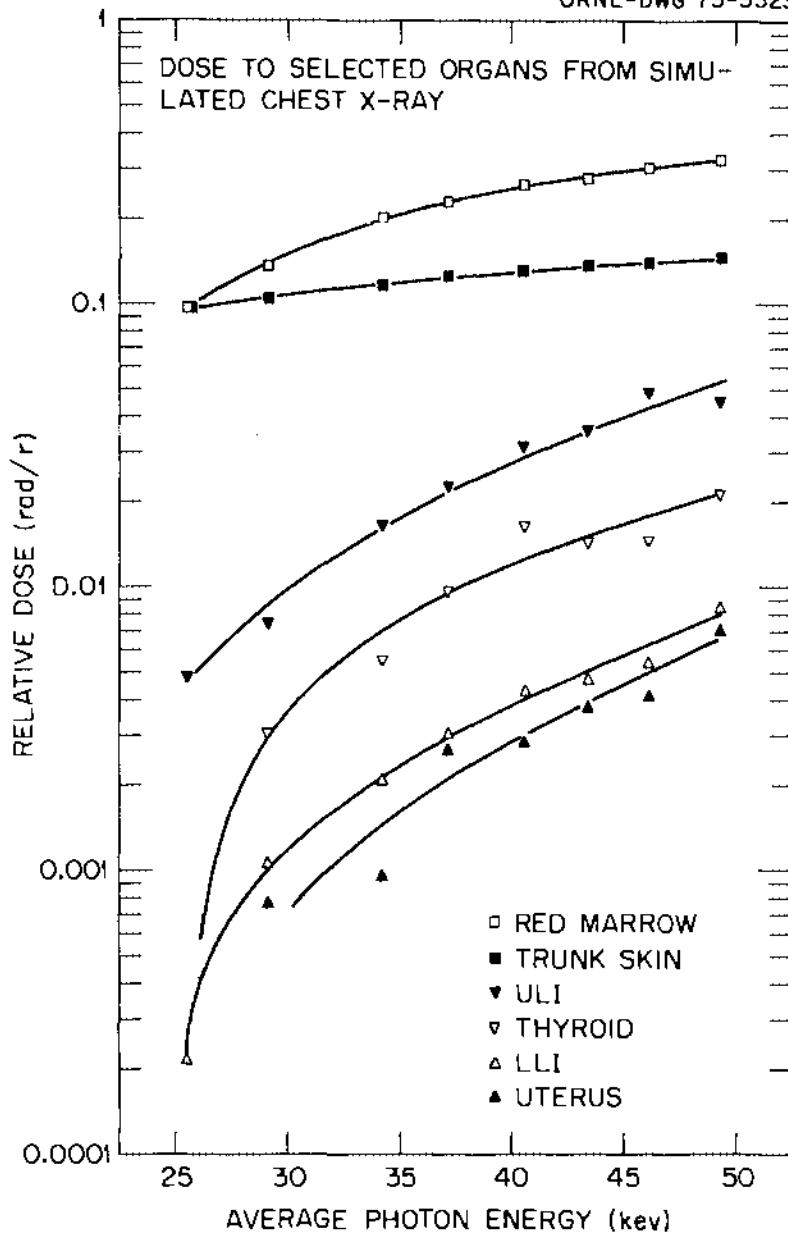
References

1. W. S. Snyder, M. R. Ford, G. G. Warner, and H. L. Fisher, Jr., "Estimates of Absorbed Fractions for Monoenergetic Photon Sources Uniformly Distributed in Various Organs of a Heterogeneous Phantom," J. Nucl. Med. Suppl. No. 3 10 (August, 1969).
2. M. J. Cook, W. S. Snyder, G. G. Warner, Health Physics Division Annual Progress Report for Period Ending July 31, 1970, ORNL-4584, pp. 200-03.
3. J. W. Poston and G. G. Warner, Medical X-Ray Dose Estimation Program Progress Report, ORNL-TM-3986, October 1972.
4. G. G. Warner and W. S. Snyder, Mathematics Division Annual Progress Report for Period Ending December 31, 1972, ORNL-4851, p. 29.
5. J. W. Poston and G. G. Warner, Medical X-Ray Dose Estimation Program Progress Report, ORNL-TM-4217, April 1973.
6. E. R. Epp and H. Weiss, Experimental Study of the Photon Energy Spectrum of Primary Diagnostic X-Rays, Phys. Med. Biol., 11 (2), 225-238 (1966).

TABLE I. Comparison of Average Absorbed Dose at 5 cm Depth to Average Absorbed Dose to Red Bone Marrow

Avg. Beam Energy (keV)	Avg. Dose at 5 cm Depth in the Beam (rad/R)	Dose to Red Bone Marrow (rad/R)	Ratio
25.5	0.164	0.242	1.48
29.1	0.303	0.343	1.13
34.2	0.564	0.503	0.892
37.1	0.707	0.573	0.810
40.6	0.836	0.657	0.786
43.4	0.944	0.696	0.737
46.1	1.08	0.754	0.698
49.3	1.18	0.805	0.682





HANDY DEVICE FOR PROTECTION OF  
THE TESTICLES IN X-RAY EXAMINATIONS

Erik Lindholmer and Ole Berg  
X-Ray Dept. 1,  
Bispebjerg Hospital, Copenhagen  
and  
State Institute of Radiation Hygiene,  
National Health Service of Denmark.

Abstract

We have modified the Picker Lead plastic gonad capsule by mounting leaf springs in the two ducts along its opening slit, thus making it "self-supporting" and able to adjust itself to the scrotum so that the testicles are covered as completely as possible.

Results are given of an investigation of the practical applicability and the shielding effect of the new device.

It is demonstrated that the shielding effect of the modified Picker lead plastic gonad capsule is as good as that of the common two-piece lead capsule device. From a practical point of view it is far better, because it is easily applied and causes the patient no discomfort.

Introduction

It is generally admitted by now that the gonads in men in the reproductive age should be protected by shields of capsule type whenever X-ray examinations exposing areas close to the gonads to the primary beam are carried out, i.e. by shields which fit tightly around the scrotum in order that the testicles may be efficiently protected against primary radiation as well as against scattered radiation coming from the body volume exposed to the primary beam. Even so, it is our impression that this type of shielding of the gonads is used only rarely, at least in Denmark. The main reason is probably that application of the common two-piece lead capsule device in general is considered inconvenient in the routine.

Accordingly, we tried in 1970 to find other means of protection and chanced to find a lead plastic gonad capsule produced by the firm Picker; it is provided with a slit-formed opening the edges of which are in the form of two ducts. It was suggested that it might be desirable if the ducts were provided with leaf springs which tentatively were fitted in. The result was a capsule which for one thing remains in place automatically, no matter the patient's movements, secondly it fits around the scrotum, its opening being the smallest possible. The original Picker capsule and the modified type are illustrated in Fig. 1. The problem of hygiene is solved by a disposable plastic bag which prior to each application is to be inserted into the capsule and turned over its edges.

A few tests on patients showed beyond doubt that the new device was by far more convenient than the solid two-piece capsule and besides, the patients found

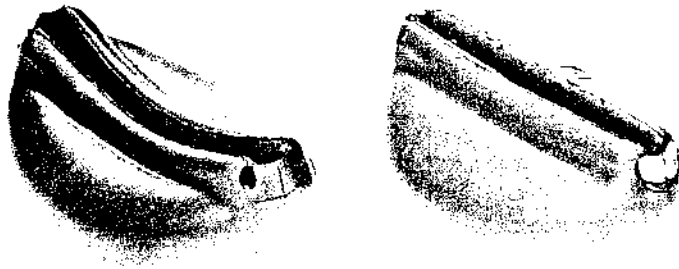


Fig. 1. Picker's original lead plastic gonad capsule and the modified capsule.

that they might easily apply it themselves. Thus, it seemed as if we actually had found a gonad shield which was more handy than the two-piece capsule and, in fact, everybody who at present have reported on the experience gained in the use of the new capsule have shared our opinion. Fig. 2 illustrates how patients in standing position manage to apply the capsule.



Fig. 2. Patient demonstrating how to apply the modified gonad capsule.

The question remained, however, whether the shielding effect of the new capsule was sufficiently satisfactory. Its lead equivalent is stated to be only 0.5 mm in contrast to an equivalent of 1 mm in the case of the two-piece capsule made by Mavig and consequently, it provides less protection against the primary beam; on the other hand, the two-piece capsule must be assumed to provide a less satisfactory protection against scattered radiation because its opening around the scrotal basis is about 15 cm<sup>2</sup> (large model) at optimal fitting while the new capsule leaves an opening of only 10-15 cm<sup>2</sup>, dependent on the anatomy of the patient. The shielding effect of the new capsule was therefore tested, partly by measurements during urography of hospitalized patients and outpatients, partly by phantom-measurements. The new capsule was finally tested in practice, mainly in order to obtain an impression of the patients' capacity to apply the capsule correctly.

#### Dose Measurements on Patients

In one series of patients the shielding effect was examined by measurement of doses accumulated during 10 routine urography examinations using no shielding and during another 10 examinations using each of the following three types of shielding: common lead rubber sheet, Mavig's two-piece capsule, and the new capsule. LiF thermoluminescence dosimeters were used for the measurements; the results are recorded in Table 1.

As regards the three groups in which shielding was used, a dosimeter was placed on top of the shielding device in addition to the gonad dosimeter on the scrotum; the former dosimeter recorded approximately the dose to be given to

	No shield	Lead rubber	Mavig capsule	Modified Picker capsule
No. of urographies	10	10	10	10
No. of films used	56	61	60	56
Mean dose externally on shielding device	(130)	380	130	85
Mean dose to gonads	130	40	24	8

Table 1. Mean gonad doses in mrad/urography with different types of shielding.

the gonads under the said conditions in the absence of shielding.

It appears from the table that the dose to the gonads was remarkably low if the new capsule was used. Even though the measurements involve a high degree of uncertainty owing to the non-standardized experimental conditions, it gave us reason to believe that the shielding effect of the new capsule was sufficient.

In order to obtain a further insight into the individual variations in doses, we continued our experiments in a minor series in which doses were measured separately during each urography, the latter including five exposures. The dosimeters were arranged as described above. The results appear from Table 2 and are to be interpreted to the effect that the two capsules are of equal value.

The conclusion to be drawn on the basis of the two tests on patients is that our measurements during urography failed to disclose any significant difference in shielding effects of the two-piece capsule and the new lead plastic capsule. The tests are described in further detail in<sup>4</sup>.

Patient	Mavig capsule							Modified Picker capsule				
	1	2	3	4	5	6	7	8	9	10	11	12
Dose externally on shield	138	73	108	125	71	85	40	110	35	75	170	30
Gonad dose	34	24	47	29	14	35	14	23	15	8	90	12
Mean dose to gonads	28							30				

Table 2. Individual gonad doses in mrad during 7 urographies using the Mavig two-piece capsule and during 5 urographies using the new capsule.

#### Dose Measurements on Phantom

Phantom-measurements were subsequently performed in order to determine the shielding effect under reproducible conditions. A therapy equipment with Greinacher coupling was used for the exposures. The Alderson-Rando phantom which for the occasion was provided with a gonad phantom is depicted in Fig. 3 in midline sectional view.

Two LiF dosimeters were placed centrally in the gonad phantom. The thickness of the stalk of the latter, on which the size of the opening of the new capsule depends, was chosen at 20 mm, providing about the maximum size of openings of capsules applied to patients, namely 15 cm<sup>2</sup>, and thus an opening similar to that of the two-piece capsule. Owing to the construction of the Alderson-Rando phantom, the opening of the capsules is unfortunately turned in the posterior-cranial direction and hence their orientation is not quite in agreement

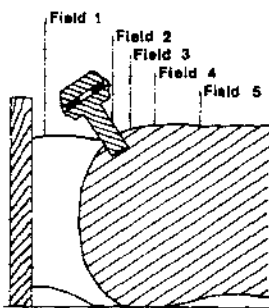


Fig. 3. Phantom in midline sectional view. Caudal field edges are sketched in.

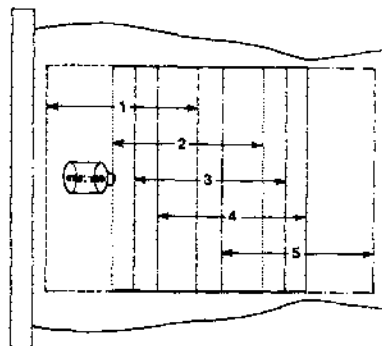


Fig. 4. Location of fields on the anterior surface of the phantom.

with that of capsules used on patients. A treatment applicator providing a field size of 17 x 25 cm on the anterior surface of the phantom was used at a distance of 70 cm from focus. The position of the fields used appears from Fig. 1.

At field 1, the gonad phantom is in the primary beam; as regards fields 2-5, their caudal field edges are at distances of 2.5, 5, 7.5, and 12.5 cm, respectively, from a point centrally between the dosimeters. Doses were measured at 60, 90, and 120 kV (total filtration equivalent to 4 mm Al). The results are recorded in Table 3, except the results obtained by common lead rubber sheet since these values, when outside the primary beam, were almost the same as those obtained without shielding. The doses are expressed in  $\mu\text{rad}/\text{mAs}$  at the given focus-skin distance.

	No shield			Mavig capsule			Modified Picker capsule		
	60 kV	90 kV	120 kV	60 kV	90 kV	120 kV	60 kV	90 kV	120 kV
Field 1	8200	21000	38000	75	350	920	42	270	700
Field 2	550	1900	3700	72	270	570	39	165	400
Field 3	270	1050	2200	48	230	470	43	150	350
Field 4	135	540	1150	19	73	200	16	71	170
Field 5	32	155	350	3	20	61	3	20	50

Table 3. Gonad doses in  $\mu\text{rad}/\text{mAs}$  measured on phantom.

It will be noted that the shielding effect of the new capsule was not in any case found inferior to that of the two-piece capsule. It should be mentioned that the results are impaired by some uncertainty because due regard has not been paid to the exact reproducibility of the orientation of the capsules. This may explain why the dose at 60 kV with the new capsule was found to be higher in Field 3 (43  $\mu\text{rad}/\text{mAs}$ ) than in fields 1 and 2.

The diagram in Fig. 5 illustrates the results obtained at 90 kV, including the results obtained by lead rubber sheet shielding. As mentioned, the shielding effect of a lead rubber sheet against scattered radiation is seen to be almost negligible.

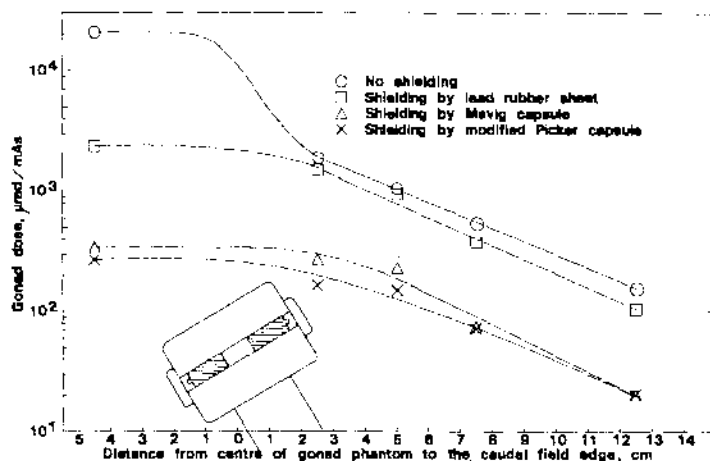


Fig. 5. Graphical representation of the results obtained by measurements on phantom at 90 kV.



The results obtained by the Mavig two-piece capsule and recorded in Table 3 have been compared with those of similar measurements performed by STEVE<sup>2</sup> in an autopsy series; The reduction factors in our series (ratio of dose measured externally on the capsule to gonad dose) were generally found to be 2-3 times higher than those reported by STEVE. According to our opinion, part of the explanation may be that, as already mentioned, the opening of the capsules in our phantom set-up is turned more backwards than is normally the case if they are used on patients.

It is not easily comprehensible why the shielding effect of the new capsule appears to be superior to that of the two-piece capsule, taking into consideration that the lead equivalent of the latter is highest (1 mm versus 0.5 mm) and that the sizes of openings were almost identical during measurements on phantom. The explanation may be that the form and orientation of the openings of the two capsules have a marked influence on the amount of scattered radiation to pass through the opening and that doses contributed by radiation through the capsular walls at 0.5 and 1 mm lead equivalent are negligible as compared with the doses contributed by scattered radiation through the opening.

#### Experience gained in Practice

The results obtained by testings in practice of the new lead plastic capsule are discussed below. The main object was to learn how many patients would manage to apply the new capsule correctly on themselves. With this end in view, a brief, illustrated instruction was prepared. The tests were performed on out-patients who met for examination in a diagnostic X-ray hospital department throughout two months; it must be admitted that it proved impossible to include all patients who appeared during the said period, partly because one physician, always the same, had to be present and supervise that application was correct in all cases, partly because we were interested primarily in the applicability of the device among young patients. It must also be admitted that the patients were not selected at random in the statistical sense of the word since selection was dependent on various practical circumstances. As the object of the investigation merely was to obtain an impression of the applicability of the device, without aiming at a direct collation with other gonad shields, the bias thus introduced is hardly of any significance.

A total of 46 patients received a lead plastic capsule - provided with a disposable plastic bag - and the written instruction immediately after they arrived in the changing room. Two or three minutes later, the examiner would appear and supervise that the capsule had been correctly applied. Thirty-one out of the 46 patients managed to apply the capsule completely correctly within the allowed interval of time. Seven patients found application rather difficult or they applied it slightly incorrectly which, however, had no essential influence on its shielding effect. Application was unsuccessful in three cases, either because of some genito-anatomical deviations or because surgery recently had been performed on the scrotum; in two of these cases, the examiner managed to apply the capsules to the patients in supine position. Two elderly patients failed to apply the capsule because they had not brought their glasses and could not read the instruction. Three patients had applied the capsule in such a way that one testicle was above the opening although there was no anatomical explanation of the phenomenon. They were all able to apply the capsule correctly after they had been told of their mistake. Not a single patient refused to participate and all tried to apply the capsule (except the two patients who had not brought their glasses). Nobody found it inconveniencing to wear the capsule, and nobody complained of having found it too difficult to apply. It is our impression, however, that application might be facilitated in some cases if the capsule were a little larger, but if so, its capacity to remain in place might be reduced in other cases and thus, we cannot recommend any changes in size until further experiments have been carried out. Application may be facilitated if the scrotum and the disposable plastic bag are sprinkled with talc pow-

der prior to application; it is not necessary, however, and was not done in any case in the present investigation.

Thus, the result of the testings in practice was that about 80% of the patients (38 out of 46) managed personally to apply the capsule sufficiently correctly. If the instruction could be revised on the basis of the experience gained, the results would probably be better.

#### Conclusion

The new lead plastic capsule seems to fulfill all reasonable requirements to a gonad shield to be used by men and it fulfills also the first five out of the six requirements set up by STILVE<sup>2</sup>: (1) It must be suitable for all types of examination and hence, it must fit tightly around the scrotum; (2) The opening admitting the root of the scrotum must be as small as possible; (3) It must be easily applicable, preferably by the patient himself; (4) It must be as small as possible; (5) It must be hygienic in use. The sixth requirement set up by STILVE, namely that the gonad shield must attenuate primary radiation to 2%, is not fulfilled, however, since this would require a lead equivalent of 1 mm at 150 kV in stead of the 0.5 mm in the lead rubber capsule. In consideration of the applicability of the capsule, it might be reasonable to be content with the 0.5 mm which is apparent also from the results of measurements performed in the present investigation from which it may be inferred that the gonad dose contributed by primary radiation through the lead rubber wall, even at high voltages somewhat beyond 120 kV, is not of great consequence as compared with the dose inevitably contributed by scattered radiation through the capsular opening.

As already mentioned, it is our impression that gonad shields of capsule type are used only on too rare occasions during X-ray examinations of areas close to the gonads. We are of the opinion that one reason is that the hitherto used capsules are highly inconvenient in use and another that a certain sense of modesty may be in evidence. Such obstacles are apparently eliminated by the new capsule which patients may apply to themselves. Accordingly, there is no longer any excuse why an effective gonad protection should not be used in all cases in which areas close to the gonads in men in the reproductive age are exposed to the primary beam. In this context, X-ray examination of areas close to the gonads refers to all types of X-ray examination in which the gonads either are in the primary beam or are less than about 10 cm from the edge of the beam.

#### References

1. E. Lindholm and G. Berg: Proc. 3rd Nordic Radiation Protection Conference, Copenhagen 1971, p. 51 - 57 (in Danish).
2. F. E. Stieve: Fortschr. Röntgenstr. 1959, 90, 1, p. 373 - 386.

## DOSE REDUCTION BY ELECTRONIC RADIOGRAPHY

Donald Sashin, Ph.D., E.R. Heinz, M.D. and E.J. Sternglass, Ph.D.  
Department of Radiology, School of Medicine, University of Pittsburgh  
Pittsburgh, Pennsylvania 15261

### ABSTRACT

High resolution electronic radiography has been developed for use in diagnostic radiographic and fluoroscopic procedures. These methods are being developed for IVP pelvimetry, intrauterine transfusions, gastrointestinal examinations, stereotaxic procedures, and selective catheterization. Preliminary clinical data show that dose reduction can range from 10 to 10,000 times depending on the resolution requirements of the particular examination.

#### I. INTRODUCTION

The use of new electronic image amplifiers (1) television systems, and electronic recording devices represents a major advance in radiological techniques by allowing a significant reduction in radiation dose and an improvement in the diagnostic quality of images. The medical radiation dose in diagnostic procedures (2) has become of increasing concern as the result of mounting evidence for serious somatic effects of diagnostic x-rays such as an increased incidence of leukemia in young children whose mothers received diagnostic x-ray examinations during pregnancy, (3) as well as increasing evidence for a growing genetically significant dose from diagnostic procedures. Recent calculations have estimated that from 3,000 to 6,000 cancer deaths annually are caused by exposure of the American public to present levels of diagnostic x-rays. In addition, ill health results from genetic damage caused by the exposure. (4)

At resolutions adequate for most diagnostic purposes the amount of radiation required for radiography, even when utilizing the best intensifier screens and fastest films, is far in excess of what would be required if x-ray quanta could be utilized as efficiently as in present electronic image intensifiers

for fluoroscopy. This arises from the fact that although one or more grains are typically sensitized for each x-ray photon absorbed in the intensifying phosphor screen, the presence of unavoidable fog at low densities causes these grains to be lost in the statistical fluctuations of the background grains.

With a high gain x-ray phosphor and television camera system, a detectable signal above noise can be produced for single x-ray quanta absorbed in the phosphor screen. This high efficiency of visible photon conversion in the photo-electric effect, amounting to as much as 65%, is constant for all fluxes of x-ray photons, independent of both of accumulated dose and dose-rate, unlike the case of film where the quantum efficiency for visible photons is generally less than 0.1 percent due to the inherent non-linearity of the photographic process since 5 to 10 visible-light photons must be absorbed by a given grain before it becomes developable.

Theoretical considerations and experimental evidence indicate that the ultimate limitations to the reduction in x-ray dose set by quantum fluctuations of the x-ray flux permit substantial reductions far below the doses presently realized with films. Even at resolutions of about 4 lp/mm and a contrast of 5 to 20% actually utilized in most clinical radiographs, significant dose reductions should be possible by the use of electronic radiography which maximizes the utilization of x-rays while minimizing the radiation exposure to the patient in diagnostic procedures. This method permits the radiologist to reduce the patient exposure depending on the resolution requirement of a particular examination. With film relatively large dose are required to attain adequate densities which

reduces the image quality because of focal spot and motion blurring. In film radiography the inability to achieve short exposures with very small focal spots limits magnification techniques, as well as the "air-gap" technique. Contrast enhancement by the use of heavy selective filtration to produce monochromatic radiation from the continuous spectrum of x-rays is severely limited by the high radiation exposure required by film. All these limitations are reduced by the substitution of the more efficient electronic technique for recording radiographic images, improving the diagnostic value of radiograms beyond that possible with chemical photography.

Basically, electronic radiography is an x-ray recording technique which involves the substitution of the highly effective photo-electric effect for the relatively inefficient photo-chemical effect taking place in film.

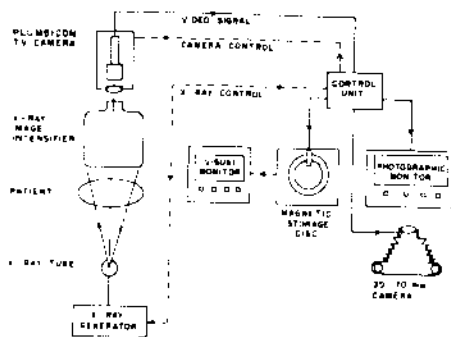


Figure 1: Block Diagram of the Apparatus

Electronic radiographs are recorded as follows (Figure 1). X-rays produced in the usual manner are incident on the object, and those that penetrate impinge upon an x-ray luminescent phosphor screen of an x-ray image intensifier. Light emitted by the phosphor screen releases photo-electrons from a thin photo-cathode in optical contact with the x-ray phosphor. The photo-electrons released are accelerated by some 30,000

volts and focused onto a small output phosphor where they give rise to a greatly brightened image which is viewed by a television camera. The optical image on the output phosphor of the intensifier is converted to an electronic charge pattern on the target of the camera tube. The charge pattern produced on the target is converted to a video-signal by means of a scanning electron beam and recorded in three alternative methods. (1) The electrical signal of one television frame or a series of frames may be recorded on a magnetic disc and then immediately replayed over a flicker-free frozen radiographic image for continuous viewing without appreciable deterioration. (2) The signal representing an individual frame may be recorded on a silicon image storage tube and immediately replayed over a TV monitor. However, this image gradually deteriorates and is completely erased after 5 to 10 minutes of continuous viewing. (3) A single video frame or many frames may be photographed by camera focused on a remote television monitor.

Systems incorporating these components have increased diagnostic information, reduced procedure time, decreased patient trauma, reduced radiation dose and permitted new types of procedures to be undertaken. Electronic radiography is used in gastrointestinal examinations (5) selective catheterization (6) pelvimetry (7) and repair of intracranial aneurysms and arteriovenous malformations (8).

In addition to improving the image quality, electronic radiography eliminates some other disadvantages of film radiography such as chemical processing time and expenses and permits a reduction of radiation exposure to the absolute minimum dictated by the quantum noise limit for the particular degree of detail and contrast needed for a given purpose.

## II. ELECTRONIC SPOT IMAGING FOR GASTROINTESTINAL FLUOROSCOPY

We have developed a high resolution technique of electronic spot imaging, to replace standard spot-filming in gastrointestinal fluoroscopy. The system consists of a standard

fluoroscopic unit electronically interfaced to a magnetic disc recorder. Key components of the system are: (1) a small focal spot x-ray tube, (2) a CsI intensifier tube, (3) a Plumbicon television camera, and (4) a 400 track magnetic disc recorder. The principle of operation is the storage of single television fluoroscopic frames on individual tracks of a magnetic disc. Once recorded, the signals can be reviewed over a television monitor at the end of the examination and later photographed for a permanent record.

At the start of an examination the unit is placed in the fluororecord mode which allows monitoring of dynamic motion. A standard television fluoroscopic image is first generated by depressing a foot pedal. Further depression of the pedal closes a second switch which automatically boosts x-ray output, and records a single television fluoroscopic frame on the magnetic disc. In contrast to normal spot-filming this recording technique does not interrupt the fluoroscopic sequence and permits the fluoroscopist to monitor the patient continuously. At the termination of an examination, the fluoroscopist can review the images by turning the hand switch to reverse. Then, every time the foot pedal is depressed, the recording head of the disc recorder is moved one track in the reverse direction and the monitor displays a frozen radiograph previously recorded as a flicker-free electronic spot image. After arriving at the initial image of a given examination the fluoroscopist by switching to the forward mode, may then leisurely study each image in the order that it was recorded. Image brightness and contrast can be adjusted on the television monitor. After checking the recorded images for completeness, the fluoroscopist is ready to proceed with the next examination. At the end of a day's fluoroscopy, the disc recorder is moved to the reporting area, where corresponding radiographs are displayed. The electronic spot images and overhead films are then

reviewed before rendering a final diagnostic report. Key electronic spot images are photographed to provide a permanent record while allowing reuse of the magnetic disc.

The radiation exposure for a single electronic spot image is about 1/50 of that required for conventional film-screen cassettes and 1/5 of the requirement for 105mm. spot-filming respectively. The exact dose depends on the resolution and contrast requirements of the particular examination.

We have compared the electronic technique to 105mm. spot-filming with regard to diagnostic accuracy in a clinical study (Fig. 2,3). Small ulcers (3-5mm. in diameter), varices, colonic polyps, various diverticula and other pathology were readily diagnosed by both techniques. In a double blind study six radiologists independently arrived at diagnoses in 21 patients. Their interpretations were then scored against those of the fluoroscopist and evaluated using the analysis of variance. The average correct scores were 76.5% and 75.5% for the electronic spot imaging and 105mm spot-filming methods respectively. Although both methods yield clinically acceptable results, we have found electronic spot imaging to be a more convenient technique of recording and reviewing fluoroscopic information.



Figure 2: Electronic Spot Image of Splenic Flexure Taken During Double Contrast Barium Enema

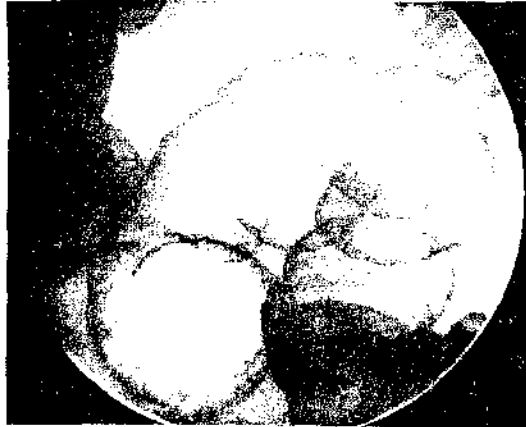


Figure 3: Electronic Spot Image  
Taken of Sigmoid Colon  
III. PELVIMETRY WITH SMALL RADIATION  
EXPOSURE

The apparatus for polaroid television technique consists of a polaroid camera focused at a remote television monitor of a standard television fluoroscopic unit. The principle of the method is to photograph an entire fluoroscopic exposure on the polaroid film which is rapidly developed. Since television fluoroscopic units have high gain image intensifiers and television cameras, the image produced on the television screen can easily be adjusted to be noise limited. Then, the radiation exposure to the patient would be reduced to a minimum which is about 1/100 of that for standard film radiography.

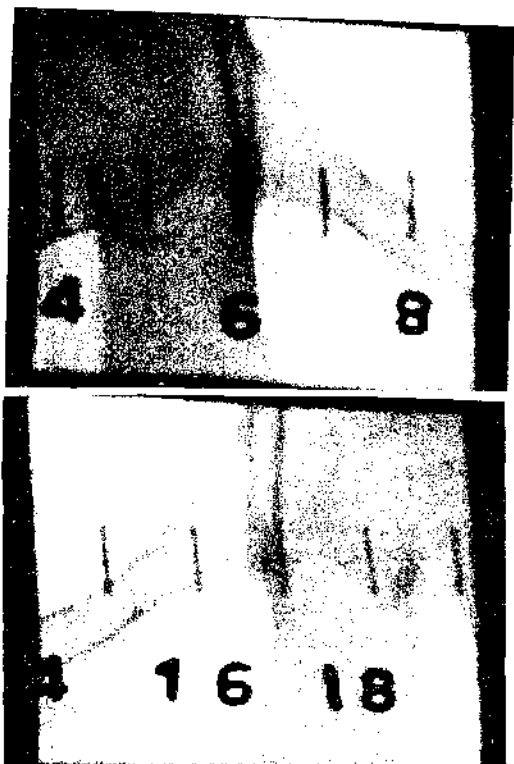
The polaroid television technique is achievable with low-cost modification of most standard television fluoroscopic units. The polaroid image is obtained in the following sequence of operations: (1) the foot-pedal is compressed, initiating the x-ray exposure, (2) the camera shutter is manually opened for a 1/2 second exposure, (3) the camera shutter automatically closes, (4) the foot-pedal is released terminating the exposure, (5) the polaroid film is developed in 15 seconds.

Since in most television chains the automatic gain control built into the television camera keep the average video output constant, the image quantum noise can be varied by changing the KVp and ma setting in the fluoroscopic exposure.

In our technique which is adapted from the orthometric pelvimetry used with standard film, a radiopaque ruler is placed between the patient and the intensifier tube. The apparatus is positioned to be centered first on one and then the other side of the anatomical structures to be measured and a polaroid fluoroscopic television image is taken. Because the field size is small, the geometric distortion is limited for each picture, and the radiation exposure is reduced from scattered radiation. The measurement of the pelvic inlet and mid-pelvis in lateral and AP views are obtained by imaging the ruler and the anatomical structure at their intersection and subtracting the two readings to obtain the distance of interest.

The fluoroscopic apparatus was slightly modified to facilitate accurate and quick measurement of the pelvis. We developed a set of clamp on lights to act as a collimator during the orthometric pelvimetry. These lights which could easily be removed at the termination of the examination, were held in place by springs and by a permanent magnet. To support the radiopaque ruler for the pelvic measurements, a plastic table was positioned on the fluoroscopic table top.

In the clinical setting, the polaroid television images of the mid-pelvis and pelvic inlet had adequate detail for the required calculations, (Fig. 4, 5). The average mid-pelvic radiation exposure to the mother in the direct beam is 1/100 of that for film casset radiography while the fetal exposure is reduced another factor of 100. The fetal exposure reduction is a result of removing the fetus from the direct beam and from using a small field size of 4 cm x 4 cm which reduces the scattered radiation. By further reducing the field size to 2cm x 2cm for each polaroid image. We can further lower the exposure. Since the polaroid fluoroscopic television technique for pelvimetry is an inexpensive and simple method that gives adequate clinical data with a radiation exposure much reduced compared to standard radiography.



Figures 4 & 5: Polaroid Television Images of Mid-Pelvis  
IV. STEREOTAXIC POSITIONING FOR NEUROSURGERY

Electronic radiography has been developed for neurosurgical procedures by coupling a two track magnetic disc recorder to a portable television fluoroscopic unit. The apparatus, enables rapid stereotaxic needle positioning for the treatment of intracranial aneurysms.

In the clinical setting for the repair of the intra-cranial aneurysm, the C-arm was placed in the AP position centered over the approximate location of the aneurysm. First, the apparatus was switched into the "MAP" mode and an electronic angiogram showing the location of the aneurysm was made. Then with the apparatus switched into the mode "STORED + MAP" successive electronic images were taken showing the needle guide superimposed with the frozen image of the aneurysm as the guide was moved into position, (FIG. 6). After positioning had been completed, the stereotaxic needle was inserted through the skull into the aneurysm. To monitor this insertion, the apparatus was placed in a lateral position and a new angiogram, taken

in the MAP mode, was superimposed with the needle as it was advanced through the brain into the aneurysm using the FLUORO - MAP mode. Tissue adhesive was injected into the aneurysm via the needle. Using the FLUORO mode the extent of occlusion of the aneurysm was evaluated by injecting contrast material into the carotid artery to outline the vessel.

The electronic technique permits successful localization and treatment of an intracranial aneurysm with procedure time and patient trauma markedly reduced compared to the traditional craniotomy.



Figure 6: Electronic Radiograph of Stereotaxic Needle Guide Superimposed with the Electronic Angiogram of the Aneurysm

The radiation dose to the patient for each electronic radiograph was 1/100 of that for one minute of continuous fluoroscopy. By superimposing single electronic radiographs on a stored map, the surgeon required only a few images, each of the order of a milliroentgen significantly reducing the radiation exposure to personnel in the operating room.

#### V. SELECTIVE CATHETERIZATION

By coupling a 150 track disc recorder to a plumbicon television fluoroscopic unit, we have developed electronic radiography for selective catheterization to reduce procedure time, radiation exposure, and the volume of injected radiopaque contrast material. There are three levels of refinement of electronic catheterization which can be applied depending on the degree of difficulty encountered in the procedure. These

methods have been clinically applied to selective catheterization of both neuro and visceral vessels.

#### A. Individual Electronic Radiograph

In this technique the angiographer takes a single electronic radiograph which he studies with the x-rays off to determine the amount and direction the catheter should be manipulated to achieve successful selective catheterization. These images showing the catheter relative to bony landmarks such as vertebrae and ribs substitute for live fluoroscopy. The progress of the manipulated catheter can be recorded on successive electronic radiographs.

#### B. Electronic Radiograph Guided by Map

An electronic angiogram is recorded during an aortic injection of radiopaque contrast. The arterial map showing the aorta with the branch vessel is electronically combined with successive individual electronic images of the catheter and the combined image is displayed over the TV monitor. By studying the position of the catheter tip relative to the branch blood vessel, the angiographer can evaluate the amount and direction of repositioning required for successful selective catheterization.

#### C. Arterial Map with Live Image Superimposed

This method, as in the previous one, uses the electronic angiogram (arterial map) as a guide for selective catheterization. In this method however the live fluoroscopic image of the catheter is electronically superimposed on the map. In a preliminary clinical trial in 20 patients, the three variations described were found to simplify the procedure and reduce the radiation dose. By obviating the need for repeated test injections of radiopaque contrast, the electronic arterial maps greatly decreased the total amount of contrast injected during catheterization. By utilizing a series of images stored on a magnetic disc, electronic radiography replaces continuous live fluoroscopy by the production of electronic images requiring much less radiation than that for

conventional fluoroscopy. This technique reduces procedure time, radiopaque contrast, and the radiation exposure.

#### VI. CONCLUSION

In the proper clinical setting electronic radiography will reduce the radiation exposure from 10 to 10,000 times of that with standard techniques in diagnostic radiology. The principle advantages of this method are that the clinical information determines the level of radiation exposure required and as a consequence of the efficient use of radiation various image enhancement techniques will yield a final electronic image with higher resolution and contrast than standard spot films in some cases. In addition to these advantages electronic radiography is convenient and opens the way for new treatment methods.

#### References

1. D. Sashin, et. al., "Resolution Contrast and Dose Reduction Performance of Electronic Radiographic Systems", 2nd Intl. Conf. on Med. Physics, Boston, Mass., 1969.
2. Ionizing Radiation: Levels and Effects, U.N. Vols. 1 & 2, E.72.IX.18, N.Y., 1972.
3. A. Stewart, G.W. Kneale, "Radiation Dose Effects in Relation to Obstetric X-rays and Childhood Cancers", *The Lancet*, 11:1185, 1970.
4. The Effects on Populations of Exposure to Low Levels of Ionizing Radiation, Environ. Prot. Agency & Dept. of H.E.W., Washington, 1972.
5. D. Sashin, W. Short, E.R. Heinz, and E.J. Sternglass, "Electronic Radiology for Spot Filming in Gastrointestinal Fluoroscopy", *Radiology*, 106:551-553, 1973.
6. D. Sashin & K. Bron, "Electronic Selective Catheterization", *Radiology* (to be published).
7. D. Sashin, S.S. Yaniv, J. Mazer & E.J. Sternglass, "Pelvimetry with Negligible Fetal Radiation Exposure" *Proc. of Symp. in Health Physics in the Healing Arts*, Dec. 1972.
8. D. Sashin, et. al., "Electronic Radiology in Stereotaxic Thrombosis of Intracranial Aneurysms and Catheter Embolization of Cerebral A-V Malformations", *Radiology*, 105:359-363, 1972.



## MEDICAL RADIATION PROTECTION IN THE EASTERN MEDITERRANEAN REGION

G. Gómez-Crespo, Adviser on Radiation Health  
WHO Regional Office for the Americas  
and  
D.R.E. Ernborg, WHO Technical Officer  
Regional Office for the Eastern Mediterranean

### Abstract

Six hundred and fifty diagnostic X-ray installations, representing over 50 percent of the existing ones, were surveyed in sixteen countries of the Middle East in 1969/70. A number of technical and human deficiencies were observed. Sixty-five percent of the X-ray units surveyed lacked one or more of the following radiological safety features: Adequate filtration; beam no larger than needed to cover the X-ray film; adequate operators' protection; and adequate protection of neighbours and all other personnel. The measures taken to tackle these problems will be described.

### Introduction

The Eastern Mediterranean Regional Office of WHO undertook early in 1969 to assist the X-ray departments in hospitals, and medical and dental departments in the countries of this Region in the evaluation and eventual improvement of the radiation safety of patients and of medical and auxiliary personnel exposed to ionizing radiation in the course of diagnostic or therapeutic procedures.

From 14 February to 7 November 1969, the WHO Technical Offices in X-rays visited ten countries of the Eastern Mediterranean Region.

During the stay in these countries he has visited 154 institutions which included medical schools, hospitals, cancer centres, tuberculosis clinics, dental schools, dentists, and private physicians (Table I); surveyed and measured field radiation levels on a total of 334 X-ray installations (Table II); instructed and demonstrated practical means of reducing dose levels to 744 X-ray operators which included radiologists, physicists, and technicians (Table III); repaired, adjusted, and recalibrated approximately 50 X-ray units; instructed dark-room personnel on processing methods leading to improvement of film quality; discussed with public health and hospital administrators the need for introducing radiation protection legislation and for establishing film-badge services; and at construction sites advised responsible authorities on the design and construction of adequate premises to accommodate new X-ray installations.

### Observations

The main shortcomings observed were:

- 2.1 human deficiencies
- 2.2 technical deficiencies

## 2.1 Human Deficiencies

The medical and para-medical personnel the Technical Officer normally met were (Table III) radiologists, radiation health physicists, X-ray engineers, and X-ray technicians.

Of the 104 radiologists met, approximately forty were expatriate doctors employed on government contracts. It is difficult to estimate precisely the shortage of radiologists. It is likely however, that the number of radiologists required is twice the number presently available.

In the ten countries visited, only five qualified radiation health physicists are available. No information could be obtained on the number of health physicists undergoing training abroad.

Radiological health inspectors do not exist in these countries.

Only four x-ray engineers are employed by their respective governments in the ten countries visited by the WHO Technical Officer. A few commercial firms keep qualified engineers on their staff in a few countries.

Training schools for X-ray technicians are operating in four of the countries visited and an attempt at training assistant X-ray technicians is now starting in a fifth one.

Approximately one third of the 634 X-ray technicians met have attended training courses varying in duration from six months to two years. As the diagnostic radiology departments are heavily dependent on the activities of this category of personnel, at least twice the number presently available is required to adequately cope with the current workload.

Low standards of exposure control and collimation were prevalent in the radiograms performed by technicians or non-radiologists. Considerable effort in improving this situation should be made and educational programs directed to this end are of great importance. Fluoroscopic examinations were often carried out by non-radiologists and even by X-ray technicians. Referring physicians often request radiological examinations without sufficient reasons, thus the yield in terms of diagnostic information is very little and patients are unnecessarily exposed to radiation.

Where radiologists are not available and other medical officers must perform radiological examinations, an adequate radiological training should be required from them.

## 2.2 Technical Deficiencies

### 2.2.1 Radiological

Sixty-five percent of the X-ray units surveyed lacked one or more of the following safety features: (Table IV)

- adequate filtration
- beam no larger than needed to cover the X-ray film
- adequate operator's protection
- adequate protection of neighbours and all other personnel (Table IV)

A number of these defects were actually rectified in the course of the visit. In most cases this could be done at small cost. Only in a few cases did the Technical Officer advise to stop further operation of the X-ray units as the

defects were highly dangerous from the point of view of radiation and/or electrical safety.

### 2.2.2 Electrical

Approximately 40% of the X-ray units seen were connected to electrical mains supply which could not provide the required power. It was also noticed that the X-ray units were connected to the same lines as other high consumers of electricity (elevators, sterilizers, etc.) thus being subject to gross power fluctuations. Most of the electric outlets (plugs) in wards where portable units are connected (bedside radiography) lacked a proper earth wire, thus exposing operators and patients to electrical hazards.

### 2.2.3 Dark Rooms

About 40 to 50% of the dark rooms have serious defects. Among the most frequently seen: lack of ventilation, light leaks, unprotected electrical fixtures, no safe-lights or incorrect filter used. Some do not have running water.

More than 80% of the dark rooms lacked one or more of required accessories, i.e., thermometers, timers, driers. Damaged cassettes, intensifying screens, hangers were often seen.

## Discussion

The analysis of the data contained in this report leads to some considerations on the adequacy of:

1. Radiological Services (Table V)
2. Radiation Protection (Tables IV, VI, VII)

### 1. Radiological Services

Table V shows in a very striking way the insufficiency of radiological services in the ten countries so far surveyed.

There is an average of 72,000 people (range 11,250 to 317,000) for each diagnostic X-ray unit, as compared with 1,000 people/unit in the United States.

The estimated average film consumption in the countries surveyed, 0.063 films/person-year, represents only one-fortieth of the average film consumption in the U.S.A. (2.46 films/person-year).

This should be kept in mind in order to place the radiation hazards to the population at large into a proper perspective.

### 2. Radiation Protection

We have seen in Table IV that only 48% of the operators and 58% of all other personnel occupationally exposed to ionizing radiation could be adequately protected by suitable structural or movable shielding, lead-glass screens, distance, etc. And yet only one-fourth of the operators (physicians, radiographers) in the ten countries surveyed (Table VI) are equipped with personnel monitoring devices. Since the number of people occupationally exposed (operators and all other personnel) is much larger, the personnel being monitored represents only a small proportion (perhaps less than 10%) of those exposed to ionizing radiation. In the U.S.A. about one-third of the personnel occupationally exposed are equipped with personnel monitoring devices.

Table VII pools the results of the survey of 334 installations in ten countries showing the percentage of units complying with some of the most essential radiological safety features.

Conclusions and Recommendations

The insufficiency of radiological services both in personnel and equipment in some of the countries surveyed is obvious.

The careful study of this report leads to evident conclusions regarding some of the remedial measures that should be taken without delay. They are as follows:

A. Stepping Up Training:

1. Of radiologists and radiological physicists.
2. Of X-ray technicians, through national courses.
3. Of X-ray technician-tutors and of technicians specialized in the maintenance and repair of X-ray equipment.
4. Of radiological health inspectors.

B. Promulgating Radiation Health Legislation:

Empowering the Ministries of Health:

1. To establish a system of registration, inspection, and licensing of X-ray, radioisotope teletherapy, and unsealed radioisotope sources and their users.
2. To promulgate rules, codes of practice, and regulations for the safe use of radiation sources.

C. Setting Up or Expanding National Services

1. For monitoring of personnel occupationally exposed to ionizing radiation.
2. For radiological health inspections.

TABLE I  
TYPE AND NUMBER OF INSTITUTIONS VISITED

Country	Private Physician	Dentist	Hospital	Tuber- culosis Clinic	Total
1)	-	-	4	-	4
2)	-	-	7	1	8
3)	-	-	18	3	21
4)	-	-	6	2	8
5)	10	-	7	2	19
6)	3	1	23	1	27
7)	-	-	21	3	24
8)	-	-	15	4	19
9)	2	2	5	1	10
10)	-	-	12	2	14
Total					154

TABLE II  
TYPE OF INSTALLATION SURVEYED

Country	Dental	Fixed Rad.	Fluoro.	Therapy	Portable	Photo-fluoro.	Combined Rad. and Fluoro.	Other	Total
1)	-	2	-	-	-	2	5	-	9
2)	1	-	1	-	-	2	7	-	11
3)	1	6	4	-	1	3	21	-	36
4)	-	-	1	2	2	2	9	1	17
5)	-	6	4	4	1	2	22	-	39
6)	1	7	-	4	-	2	38	-	52
7)	-	7	17	2	7	2	29	-	64
8)	-	2	5	6	16	4	23	1	57
9)	4	2	1	2	4	1	6	-	20
10)	2	3	3	3	3	-	14	1	29
Total									334

TABLE III

Country	Radiologists	Physicists	X-ray Operators
1)	1	0	22
2)	0	0	27
3)	12 (f)	0	79
4)	10	2	42
5)	15	0	92
6)	27	1+1 WHO	107
7)	14	1	120
8)	10 (f)	0	92
9)	6	1	30
10)	9 (f)	0	23
Total	104	6	634

TABLE IV

Country	A			B			C			D			Total units surveyed	% of units in which one or more of A,B,C,D, features were missing
	Filtration adequate			Beam no larger than needed to cover X-ray film			Operator can be adequately protected			All other personnel within permissible limits				
	Yes	No	% Compl.	Yes	No	% Compl.	Yes	No	% Compl.	Yes	No	% Compl.		
1)	6	3	66.5	4	4	50	6	3	67	4	4	50	9	50
2)	3	8	27	6	5	54.5	5	6	45	3	8	27	11	73
3)	28	8	78	26	3	89	24	12	67	26	12	68.5	36	33
4)	9	8	53	9	2	82	11	6	65	16	1	94	17	47
5)	12	27	31	17	10	63	21	18	53	28	9	76	39	69
6)	20	31	39	39	6	87	29	23	56	47	4	92	52	61
7)	7	52	12	28	13	68	29	35	45	29	35	45	64	88
8)	13	44	23	27	13	68	23	34	40	12	38	33	57	77
9)	9	11	45	11	5	69	6	14	30	13	6	68.5	20	70
10)	9	20	31	15	3	83	6	23	21	5	24	17	29	83
Total	116	212	35 (Av.)	182	64	74 (Av.)	160	174	48 (Av.)	183	141	56 (Av.)	334	65 (Av.)

TABLE V  
RADIOLOGICAL SERVICES

Country	Population (1)	Estimated no. of diagnostic X-ray units	Population per diagnostic X-ray unit	No. of physicians (2)	No. of physicians per X-ray unit etc	No. of radiologists	No. of radiographers	No. of operators/unit etc	Estimated annual X-ray film consumption	Estimated no. films/person-year
1)	260,000	15	17,333	117	7.8	1	22	1.5	98,500	0.378
2)	2,755,000	20	137,750	86	4.3	0	27	1.3	171,500	0.062
3)	23,782,000	75	317,093	320	4.3	12	79	1.2	464,500	0.019
4)	2,251,000	20	112,550	505	25.0	10	42	2.6	182,750	0.081
5)	5,724,000	50	114,480	978	19.5	15	92	2.1	266,250	0.046
6)	2,588,000	230	11,252	2,025	8.8	27	107	0.58	574,250	0.221
7)	4,463,000	100	44,630	666	6.7	14	120	1.3	590,250	0.132
8)	1,675,000	100	16,750	530	5.3	10	92	1.0	519,500	0.310
9)	620,000	40	15,500	460	11.5	6	30	0.9	165,250	0.266
10)	5,100,000	35	145,714	84	2.4	9	23	0.9	30,500	0.025
Total	49,218,000	685	71,851 (Av.)	5,771	8.4 (Av.)	104	634	1.08 (Av.)	3,113,250	0.063
USA	200,000,000	206,560 (3)	970 (Approx.)	307,422 (3)	1.83			1.5 (3)	506,000,000 (4)	2.446

- (1) Demographic Yearbook, 1966, UN.  
 (2) World Health Statistics Report, Vol. 21, No. 2-3, 1968, WHO.  
 (3) Lawrence R. Fess, Summary of Diagnostic X-ray Statistics Relating Facilities, Equipment, and Personnel by Healing Arts Professions, Radiological Health Data and Reports, Vol. 10, No. 9, Sept. 1969, pp. 379-380.  
 (4) John H. Knowles, Radiology - A Case Study in Technology and Manpower, New England Journal of Medicine, 280, 1271-1276, (19).

TABLE VI  
PERSONNEL MONITORING  
(Film-Badges)

Country	Total No. of Operators	Personnel Monitored	%
1)	23	0	0
2)	27	0	0
3)	91	21	23
4)	52	52	100
5)	107	0	0
6)	134	68	51
7)	134	16	12
8)	102	6	6
9)	36	23	63
10)	32	2	6
Total	738	188	26

TABLE VII  
MAIN RADIOLOGICAL SAFETY FEATURES  
Pooled Results of Ten Countries  
(% Units Complying)

EQUIPMENT	Filtration adequate	35.4
	Beam no larger than needed to cover film	74
	Tube housing leakage within normal limits	100
	Table top dose < 10 R/min	96
	Fluorescent screen interlocked with tube	91
	Fluoroscopic shutters adequate	92
	Lead glass on fluorescent screen adequate	97
OPERATORS	Operator can adequately be protected	48
	Exposure of all other personnel within permissible limits	56

## SOURCES OF UNNECESSARY IRRADIATION DURING FLUOROSCOPY.

N.T.Racoveanu, Georgeta Modoran and S.Suma

(Institute of Hygiene and Public Health, Radiation Hygiene Laboratory, Dr.Leonte str. nr.1-3, Bucharest 15, Romania)

### Abstract

644 gastrointestinal and 731 chest fluoroscopies performed by 6 radiologists were investigated concerning : the duration in time, the area exposed, magnitude of exposure in X-ray beam, the gonadal and bone marrow dose.

The patient exposure was found to depend on : 1.mean time duration of the examination, which varied for the six different radiologists, from simple to double (from 169 to 259 sec. for gastroduodenal fluoroscopy, 48 to 80 sec. for intestinal, and 11 to 62 sec. for chest fluoroscopies) ; 2. quality and exposure rate of primary beam (the exposure) rate ranged from 3 to 12 R/min.) ; 3. area exposed during fluoroscopy (cross-section of primary beam varied from 26 to 230 cm<sup>2</sup> for gastroduodenal, from 38 to 383 cm<sup>2</sup> for intestinal and from 47 to 490 cm<sup>2</sup> for chest fluoroscopy ; 4. the value dose area product proved to be the most interesting parameter, variations between 420 and 3160 R.cm<sup>2</sup> for gastroduodenal, 124 to 2091 R.cm<sup>2</sup> for intestinal and 100 to 473 R.cm<sup>2</sup> for chest fluoroscopies were recorded, the corresponding integrated dose values were from 3.5 to 26.8 kg. rad for gastroduodenal, from 4.0 to 16.6 kg. rad for intestinal and from 0.8 to 4.0 kg. rad for chest fluoroscopies.

The diagnostic informations being practically the same, the variations of the operational parameters encountered to the six investigated radiologists, evinces the existence of at least three sources of unnecessary irradiation : 1. the use of a too large beam (large surface and integrated dose) ; 2. the dose rate of primary beam too high ; 3. too long time of irradiation ; this situation maybe related to the knowledge level of the radiologist.

### 1. Introduction

Previous research has shown that fluorescopic examinations represent the major contribution to medical irradiation of the population in Romania. Due to the great number of fluoroscopies performed in this country, over 300 per 1000 inhabitants, instead of 80-110 in other countries, we were interested in the assesment of the sources of unnecessary irradiation during fluoroscopies.

Fluoroscopy is a nonstandardizable type of X-ray examination, its performance being entirely dependent on accuracy of the radiologist's knowledge and on the interest he pay, to the benefit vs.risk ratio.

The higher benefit/risk ratio maybe obtained by performing the fluoroscopy in that way to find maximum of diagnostic information with minimum of irradiation dose to the patient.

Objectivation of sources which contribute to lowering the above mentioned ratio in view of their control is the main aim of this paper.

## 2. Material and method

Six radiologists working in three outpatient clinics in Bucharest were subjects of the present investigation.

Measurements were done during 644 gastrointestinal and 731 chest fluoroscopies concerning : 1. time duration ; 2. cross-section of usefull beam ; 3. dose area product ; 4. dose rate of X-ray machine ; 5. doses received by some tissues and organs (skin, bone marrow, gonads, thyroid, eyes, a.s.o.). Technical parameters (KV, mA) used by the radiologist were also recorded.

For measurements we have used : 1. the Diamantor (PTW Pychlau) for dose area-product ; 2. the VA-J-15A Dosimeter (Vakutronick) for dose rate ; 3. thermoluminiscent dosimeters TLD 100 (readed at a 2000 Harshaw apparatus) for tissue and organ dose. All dosemetric facilities were calibrated at the WHO-IAEA Regional Reference Centre for Secondary Standard Radiation Dosimetry in Bucharest.

## 3. Results and discussions

Table 1 is showing the mean values data obtained for g.i. fluoroscopies.

Table 1.

Time, dose, area product, exposed area, integrated dose and dose rate used by six radiologists in Bucharest during gastrointestinal fluoroscopies (mean values).

	Dr.P.	Radiologist				
		Dr.G.	Dr.I.	Dr.L.	Dr.T.	Dr.S.
<u>I. Upper g.i. fluoroscopy:</u>						
Time (seconds)	82.5	212.3	161.8	258.8	206.2	194.7
R.cm <sup>2</sup>	1880.0	3160.0	1300.0	2430.0	900.0	420.0
cm <sup>2</sup>	210.0	95.0	220.0	230.0	53.0	26.0
kg.rad	15.0	26.8	10.4	19.3	7.4	3.5
dose rate (R/min.)	7.3-12	10.6-11.3	3.0	3.0		5.0-8.2
<u>II. Lower g.i. fluoroscopy:</u>						
Time (seconds)	51.9	-	48.1	64.1	79.8	36.9
R.cm <sup>2</sup>	2091.2	-	556.9	735.7	480.6	124.1
cm <sup>2</sup>	383.2	-	337.5	222.8	67.7	37.6
kg.rad	3.9	-	4.5	6.0	3.1	0.8



Data obtained for chest fluoroscopies are shown in table 2.

Table 2.

Time, dose, area product, exposed area, integrated dose and dose rate used by six radiologists in Bucharest during chest fluoroscopies (mean values).

	Radiologist					
	Dr.P.	Dr.G.	Dr.I.	Dr.L.	Dr.T.	Dr.S.
Time (seconds)	11.2	27.3	42.8	62.4	54.3	25.4
R.cm <sup>2</sup>	473.2	432.0	356.0	494.0	371.0	99.7
cm <sup>2</sup>	490.0	122.8	302.4	363.9	83.6	47.1
kg.rad	3.9	3.6	2.9	4.0	3.1	0.8
dose rate(R/MIN)	5.2	7.7	1.6	1.6	4.7	5.0

Both tables 1 and 2 are displaying the same phenomenon - the irradiation of the patient during g.i. and chest fluoroscopy is due mainly to cross-section of useful beam and not to the intensity and quality of the beam used.

If we compare, for the six radiologists we have studied, the dose area product, exposed area and integrated dose with time and dose rate the above mentioned phenomenon becomes more evident.

For g.i. and chest fluoroscopies the differences between maximum and minimum values for dose area product, exposed area, integrated dose, time and dose rate are the following :

	upper g.i.	lower g.i.	chest
dose.area product (R.cm <sup>2</sup> )	420 to 3160 7.9	124 to 2091 16.9	99.7 to 494 4.9
exposed area (cm <sup>2</sup> )	26 to 230 8.8	37.6 to 383.2 10.2	47.1 to 490 10.4
integrated dose (kg.rad)	3.5 to 26.8 7.7	0.8 to 6.0 7.5	0.8 to 4.0 5.0
time (seconds)	82.5 to 258.8 3.1	36.9 to 64.1 1.7	11.2 to 621 5.6
dose rate (R/min.)	3.0 to 12.0 4.0		1.6 to 7.7 4.8

From the above figures results that the greatest difference between minimum and maximum mean values obtained for the six radiologists was that of exposed area. An order of magnitude is separating the minimum and maximum values for the above mentioned parameter both in g.i. and chest fluoroscopy.

The dose.area product which reflect not only the cross-section but also the intensity of the primary beam is also very relevant about the unnecessary exposure of patient. For lower g.i. fluoroscopy especially dr.S. has obtained the diagnostic information with 1/17th part of the roentgen area product used by dr. P. Concerning upper g.i. and chest fluoroscopies the differences are smaller but still important (1/8 and 1/5).

Integrated dose, measuring the true tissue dose, is much

more important from the radiological point of view. This parameter also reveals differences between absorbed dose in patient of 5 fold in chest fluoroscopy and 7.7 or 7.5 in upper and lower g. i. fluoroscopy.

By comparison the differences concerning the duration of procedures were only 1.7 to 5.6 fold and the dose rate of primary beam 4.0 - 4.6 fold.

Data above discussed are consistent in showing that the main sources of unnecessary irradiation of the patient during fluoroscopy are the following, in order of their importance :

1.- the cross-section of the primary beam-expressed in our study as the exposed area of the patient ;

2.- the intensity of the primary beam ; use of higher ma values produces, together with larger beam, greater dose.area products and integrated dose values ;

3.- the duration of exposure, which results from the fact that the radiologist try to obtain as many information is possible from fluoroscopy and not from combining the use of fluoroscopy and radiography.

A realistic programme for control of unnecessary irradiation during fluoroscopy requires the improvement of the knowledge of radiologists concerning :

a) - use of smallest possible cross-section of primary beam during fluoroscopy except for a very brief initial general view ;

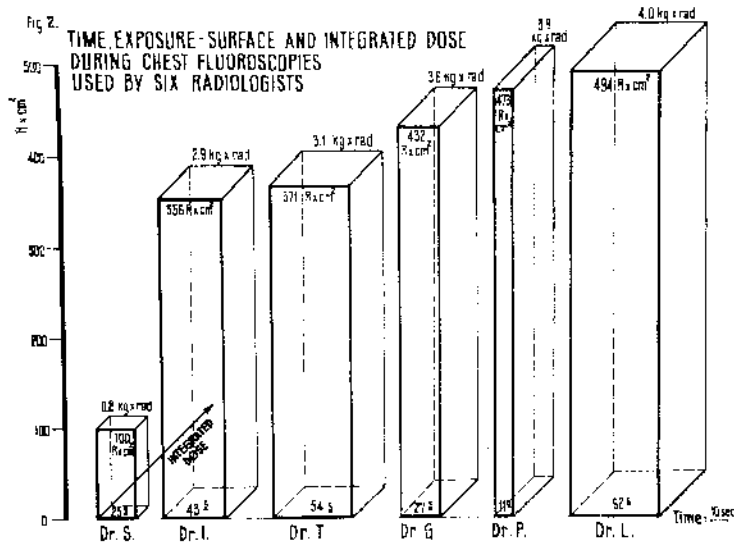
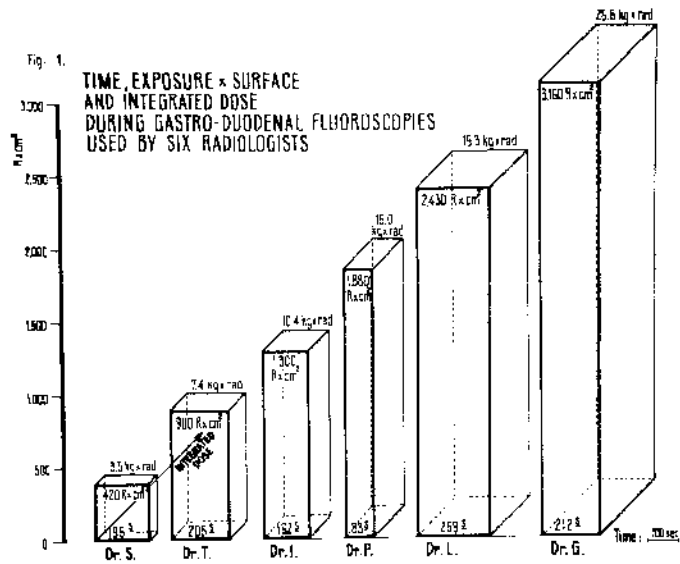
b) - decrease of intensity of tube current and increase of  $KV_p$  in view of obtaining the lowest dose rate for the useful beam

c) - take of radiographs during fluoroscopy in cases which are not clear enough on fluoroscopic screen avoiding the prolongation of the patient exposure.

As dose.area product and integrated dose are also a measure of the irradiation received by the radiosensitive tissues and organs (bone marrow, gonads, a.s.o.) the significance of these parameters from the viewpoint of radiation protection is highly relevant. The decrease in roentgen.area product and kg.rad values have a real value in control of unnecessary irradiation during radiological diagnostic procedures.

#### References :

- 1.- Ardren G.M. and H.E.Crooks-Checking diagnostic X-ray beam quality, Brit.J.Radiol.41, 193-98, 1968.
- 2.- I.C.R.P.: The evaluation of risks from radiation, a report prepared for ICRP Committee 1, ICRP Publ.8, 1966.
- 3.- I.C.R.P.: Protection of the patient in X-ray diagnosis, a report prepared by a task group of Committee 3, ICRP Publ.16, 1970.
- 4.- Weil F., J.F.Bonneville et J.P.Ricatte-Regards sur le problème de l'irradiation diagnostique, La presse Méd.76, nr.34, 1968.
- 5.- W.H.O.: Report of a WHO Seminar on the use of medical radiological apparatus and facilities, held in Singapore, 9-21 Nov 1970, Geneva- May, 1971.
- 6.- Stewart C.Bushong - The development of current radiation protection practices in diagnostic radiology, Chim.R.Comp., 1971



MEDICAL IRRADIATION OF THE POPULATION IN ROMANIA  
DURING 1970

N.T.Racoveanu, Cornelia Diaconescu, Georgeta Modoran  
Corina Vasilescu-Fulea, P.Plavosin and C.G.Milu  
(Institute of Hygiene and Public Health, Bucharest-Romania)

Abstract

X-ray examinations have increased in Romania between 1953-1970 from 429 to 1,012 per  $10^3$  inhabitants, photofluorographies (54 to 452) and radiographies (37 to 238) being developed while radioscopies remained at the same level (338 and 322).

Age group 16 to 30 and over 31 years are the most X-rayed averaging 1.22 examinations/inhab./year. Males are predominately investigated at all ages and the most frequent examinations concern the chest, G.I.tract, upper and lower members, vertebral column, pelvis, teeth, a.s.o.

Were calculated for 1970 the genetically significant dose and the mean bone marrow dose, the value obtained been 28.5 and 382 mrad respective. Radiographies of the addominal and lumbar regions in female had the main contribution to genetically significant dose and the G.I.series, chest fluoroscopies and photofluorographies to mean bone marrow dose.

1. Introduction

Medical use of ionizing radiations is still the major source of artificial irradiation of the population throughout the world.

The present work has attempted to estimate population's genetically significant dose and mean bone marrow dose due to medical irradiation in Romania.

2. Increase of the X-ray diagnostic procedures in Romania

Statistical records concerning all medical activities were developed in Romania and since 1953 we have yearly data refering X-ray procedures in all medical units throughout the country.

The data show the fact that radiographies and photofluorographies have increased 6.4 and 8.4 times during the last 18 years; at the same time radioscopies have increased very slowly until 1964 and are decreasing after that year.

3. Distribution of X-ray diagnostic procedures.

Using data recorded during 1970 by 98 different X-ray diagnosis departments throught the country we have established on 145,525 radioscopies, 115,271 radiographies and 231,600 photofluoroscopies the distribution of X-ray procedures on: sex, age

group (0-15, 16-30 and over 31 years) and body area exposed. The result of the examination coded as positive and negative was also mentioned.

Due to the shortage of space we are obliged to present only in a very condensed form these results.

The number of examinations per person in total population were as follows :

- up to 15 years	- boys	0.50 examinations
	- girls	0.48 examinations
- between 16-30 years	- male	1.36 examinations
	- female	1.09 examinations
- more than 31 years	- male	1.27 examinations
	- female	1.17 examinations

Data show that the age group 16-30 years, which is very interesting from the point of view of genetically significant radiation dose, performs an increased number of examinations, especially the men, fact which must be taken into account in any programme of reduction of medical irradiation.

#### 4. Exposure of patient during the radiological examinations

The exposure received by the patients was determined by direct measurements on the patient during different kind of examinations, using pocket ionization chambers and TLD-100 Harshaw dosimeters.

The measurements were performed in 62 medical X-ray diagnosis units, during 5,370 radiographies, 8,750 radioscopies and 9,370 photofluorographies.

The dosimeters were calibrated in the energy range from 60 KVp (0.056 mm Cu HVL) to 120 KVp (0.400 mm Cu HVL) at the WHO IAEA Regional Reference Centre for Secondary Standard Radiation Dosimetry in Bucharest.

The measurements of the exposure received by patients during X-ray examinations were done by positioning the dosimeters at several points on the surface of the body. The points for gonadal dose determinations were the anterior and posterior projections of the ovary on the skin in female and testes in male. Bone marrow determinations were done at the most important skeletal part with red bone marrow, in the vicinity or directly in the primary beam. The values obtained in such a way are skin doses and we used during the calculation a correction factor of 0.3 (experimentally determined) for the true bone marrow dose (table 2).

For a realistic estimation of patient dose during radioscopies, the average time duration of radioscopies measured during our field investigations was used : 68% of chest fluoroscopies were until 30 seconds and 11% over 60 sec., as for G.I. fluoroscopies 41% were until 80 sec., 54% between 80-240 sec. and 5% more than 240 sec.

#### 5. Genetically significant dose

The following simplified equation for the genetically significant dose (GSD) was used (see also reference 1) :

$$GSD = \frac{\sum D_i N_i P_i}{\sum N_i P_i}$$

where :

- $D_i$  = mean gonadal dose from certain type of examination received by a patient of age group "i" ;
- $N_i$  = number of persons of age group "i" who were exposed to the specified type of examination during 1970 ;
- $P_i$  = expected number of children per person of age group "i" and mentioned sex ;
- $N_i$  = number of persons of age group "i" and mentioned sex in the total population.

Using data from chapters 3 and 4 we found the gonadal dose of the population in Romania during 1970 (table 1, part I).

The calculus of genetically significant dose using the equation mentioned above and data from table 1 gives a value of  $GSD=26.5$  mrad/year/inhabitant, which is comparable with the values obtained in others countries as USA (1964)-55 mrad, Sweden (1955)-38 mrad, Japan (1960)-39 mrad, Denmark (1956)-22 mrad, United Kingdom (1957-1958)-14 mrad and New-Zeeland (1963)-12 mrad.

#### 5. Mean bone marrow dose

The mean bone marrow dose was calculated in a simple way by integrating of all values for bone marrow dose produced during a certain type of investigation in persons of a specified age group and dividing by the total number of persons of that age group.

Using data from chapters 3 and 4 and values of distribution of population from table 1, we found the bone marrow doses of the population in Romania during 1970 given in table 2 and finally as result, the mean annual bone marrow dose per inhabitant per type of examination (table 2, last column).

As it can be seen, the bone marrow dose averaged over entire population resulting from various X-ray examination was 382 mrad/year, in comparison with 32,4 mrad/year estimated in United Kingdom by Auriar Committee (1957-1958, reference 5).

The mean bone marrow dose arising from chest radioscopies was 92 mrad/year/inhabitant in comparison with 50 mrad (Austria), 10 mrad (Belgium), 600 mrad (France), 8 mrad (Spain) and 14 mrad (Switzerland).

Bone marrow dose seems to be the most important health consequence of the medical irradiation. We have not a good estimation of leukaemia expectancy in Romania so that we couldn't calculate the leukaemogenic significance of bone marrow irradiation.

The fact that age group 16-30 years, which has enough large life expectancy (in 1970 in Romania the mean life expectancy was 66 years for men and 70 for women) receive an important part of medical irradiation, gives us the real idea referring the programme for reduction the unnecessary irradiation.

## 7. Conclusions

The results of our work concerning the medical irradiation of the population in Romania allow to point out the following conclusions :

a.- medical irradiation in Romania constantly increases; an increase of 2.4 times in 1970 can be presented for total X-ray diagnosis procedures in comparison with 1953 (6.4 times for radiographies and 0.4 for photofluorographies) and a slowly decrease for radioscopies (322 radioscopies in 1970 per  $10^3$  inhabitants instead of 337 in 1953) ;

b.- the number of examinations carried out during 1970 by one inhabitant varied with age group and sex. It was 0.50 for boys between 0-15 years, 0.48 for girls, 1.36 for men between 16-30 years, 1.27 for men over 31 years, 1.09 for women between 16-30 years and 1.17 examinations for women over 31 years ;

c.- the genetically significant dose in Romania during 1970 was 28.5 mrad per inhabitant, a medium value in comparison with those established in other countries in the world;

d.- the mean bone marrow dose per inhabitant in Romania during 1970 arising from X-rays diagnosis was 362 mrad, a value which put in evidence the necessity of a programme for reduction of the unnecessary medical irradiation.

## References :

1. Arduran G.M. and H.E.Crooks-Checking diagnostic X-ray beam quality, Brit.J.Radiol. 41,193-98,1968.
2. Baker, P.M. - The genetical significant dose from diagnostic X-ray in Canadian public hospitals, Rad.Prot.Div.Dp.Health and Welfare, RPD-31, feb.1963, p.95.
3. Brown M.L., P.L.Roney and A.W.Hillberg-Estimates of gonadal and genetically significant dose to the US population from diagnostic radiology. Amer.Jourr.Publ.Health, vol.58, nr.12 p.2267-74, 1968.
4. ICRP : Protection of the patient in X-ray diagnosis, a report prepared by a task group of Committee 3, ICRP Publ.16, 1970.
5. Radiological hazards to patients : final report, Ministry of Health, H.M.S.O, 1966.
6. Seelentag W. - On the importance of the radiation burden of a population with special reference to the genetically significant dose from application of radiation in medicine. Progress in Nuclear Energy. Series XII, Health Physics, vol.2, part I, p.125-156, 1969.
7. Stewart C.Busaong - The development of current radiation protection practices in diagnostic radiology, Chemical R. Comp., 1971.

Table 1. Gonadal dose of the population (rad), distribution of population and children expectancy, per age group and sex in Romania during 1970.

	Sex	Age group			Total
		0-15 years	16-30 years	over 31 years	
		Gonadal dose (rad)			
I. Type of examination:					
A. Radiographies	M	4,100	19,475	101,680	125,255
	F	26,035	164,000	775,130	966,165
B. Radioscopies:					
Chest	M	410	1,743	3,075	5,228
	F	717	1,743	4,305	6,765
G.I.	M	615	4,100	12,505	17,220
	F	1,845	5,535	18,450	25,830
C. Photofluorographies					
	M	3,075	1,743	2,562	7,380
	F	7,175	7,790	14,555	29,520
	Total	43,972	206,129	933,262	1.183,363
II. Distribution of population (x10 <sup>3</sup> )					
	M	2,972	2,578	4,920	10,270
	F	2,829	2,338	5,063	10,230
	Total	5,801	4,916	9,983	20,500
III. Children expectancy					
	M	2.953	2.798	0,453	
	F	2.885	2.402	0,310	

1148



Table 2. Bone marrow dose of the population (rad) per age group and mean marrow dose per inhabitant (mrad) in Romania during 1970.

1149

Type of examination	Age group			Total (rad)	Mean annual bone marrow dose per inhabitant (mrad)
	0-15 years	16-30 years	over 31 years		
A. Radiographies	22,872	62,839	299,974	385,685	19
B. Radioscopies :					
Chest	241,427	559,373	1,112,915	1,886,715	92
G.I.	100,990	495,274	3,160,082	3,756,346	183
C. Photofluorographies	86,778	638,800	1,083,133	1,808,711	88
T o t a l	425,067	1,756,286	5,655,104	7,837,457	382

## Use of Medical X-Ray Diagnostic Units in Iraq

MOUSA S. ABBAS  
Radiation Control Office  
Ministry of Health  
Baghdad - Iraq

### Abstract

The medical use of X-rays represents a major source of population radiation exposure. A special consideration is given to assess the genetically significant dose due to diagnostic radiology.

All medical institutions, hospitals, private physicians and dentists in Iraq, were visited to record: the number and type of X-ray units, the frequency and kind of X-ray diagnostic examinations and number of workers in such units. In addition the protection of the workers from the radiation and the services of personnel monitoring were also observed.

It was found that, up to the end of 1972, there are 407 X-ray units, all over the Republic used for whole body medical roentgenodiagnosis and 146 X-ray units for dental radiography, carrying out more than  $4 \times 10^6$  and  $15 \times 10^4$  examinations per year, respectively.

The study gives a clear map of the distribution of the X-ray units in the different parts of the country and can be used as a guide for future radiological health programs. It might also serve as a model for radiation survey and inspection. Also, the figures could be useful in the estimation of the annual genetically significant dose received by the 10 million Iraqis from exposure to diagnostic X-rays.

### Introduction

X-ray diagnosis in the application must be used only when highly specifically indicated. The radiation dose received on an individual ground varies with the examination used, the number of exposures during their reproductive period of life and the techniques used.

Diagnostic X-rays started to be used for medical purposes in Iraq since 1917 / 1 /. No scientific measures, concerning protection of personnel and patients were taken until 1972 when a highly specialized committee was established to study this problem.

A legalized regulations were issued by this committee applying what the ICRP suggested for radiological protection and permissible doses / 2-3 /.

Therefore in response to the need of radiological health program for an effective method of surveying of all radiation sources, the Radiation Control Board requested all medical institutions and hospitals, to register radiation sources and give full details of their uses.

The survey covered approximately 99% of the radiation machines, which constitute the subject of this paper for the year 1972.

## Materials and Method

Regulations were issued restricting the sale and import of diagnostic X-ray films or any spare parts without permission. Such permission is now given only after a visit to the X-ray machine in order to record the number and type of the unit, the frequency and type of X-ray examinations performed, output measurements at various tube voltages and the different age groups of the patients of both sexes. In addition, the protection of the workers from the radiation and the services of personnel monitoring were also observed.

With regard to the dose measurements, skin dose estimation either of the critical organs was made directly on patients during different types of X-ray examinations. Film badges type E.R.P. 30 Black Spot, such as personnel monitoring film badge with kodak radiation-monitoring films were used for these measurements for practical reasons and availability. The contribution of radiography to gonadal dose is a complex function of applied peak kilovoltage, tube target to skin distance, volume of the tissue in the primary beam, the sex and age of the patient, / 4 /.

The dose received by gonads in 70 patients exposed to radiation for diagnostic purposes was calculated. The genetically significant dose was obtained from the following formula / 5 / :

$$D = \frac{\sum_k \sum_j N_{jk}^{(F)} W_{jk}^{(F)} d_{jk}^{(F)} + \sum_k \sum_j N_{jk}^{(M)} W_{jk}^{(M)} d_{jk}^{(M)}}{\sum_k \frac{N_k^{(F)}}{k} W_k^{(F)} + \sum_k \frac{N_k^{(M)}}{k} W_k^{(M)}}$$

where

D = (annual) genetically significant dose.

$N_{jk}$  = (annual) number of individuals of age-class k, subjected to class j exposure.

$N_k$  = total number of individuals of age-class k.

$W_{jk}$  = future number of children expected by an exposed individual of age-class k subsequent to a class j exposure.

$W_k$  = future number of children expected by an average individual of age-class k.

$d_{jk}$  = gonad dose per class j exposure of an individual of age-class k.

(F) and (M) denote "female" and "male" respectively.

### Results

#### A. Number of machines and area distribution :

A total number of 553 X-ray machines were used in Iraq for diagnostic purposes (Table I), 407 conventional X-ray units are used for general diagnostic medical purposes, the remaining 146 units are used only for dental purposes. Some of these machines are quite old (since 1934).

30.5% of the conventional X-ray machines are found in private clinics, while in the case of dental X-ray machines the number of the machines in the private clinics were higher than the governmental machines (53.3% of the total dental machines).

With regard to the area distribution, Baghdad (the capital) has the higher number of machines; Basrah came the next and Nineva came the third. Other provinces are more or less similar in the number of the X-ray machines used.

Table 2. shows the different firms of both conventional and dental machines used in Iraq. Since there are approximately 10 millions inhabitants in Iraq at the end of 1972 / 6 /, it means that the average is 1 medical diagnostic X-ray machine for each  $25 \times 10^3$

inhabitants and 15 dental X-ray machines for each million inhabitants.

Table I. Distribution of X-ray machines by Governorates ( Muhafadha ).

Muhafadha	Inhabitants %	Diagnostic			Dental		
		Public	Private	Total	Public	Private	Total
Duhok	11.1	4	-	4	1	-	1
Nineua		18	9	27	6	5	11
Arbil	4.5	12	2	14	1	-	1
Kirkuk	5.9	13	6	19	2	3	5
Sulaimaniye	4.7	11	3	14	3	-	3
Diyala	4.5	9	1	10	1	-	1
Baghdad	25.5	107	79	186	29	53	82
Al-Anbar	3.9	11	1	12	2	-	2
Babylon	5.7	11	2	13	1	2	3
Karbala	4.2	11	4	15	4	3	7
Al-Radisiye	5.8	9	2	11	1	1	2
Al-Muthana		3	1	6	1	-	1
Wasut	4.2	7	2	9	2	-	2
Thecare	6.4	9	1	10	4	3	7
Measan	4.3	12	1	13	3	1	4
Basrah	8.4	34	10	44	7	7	14
Total	100	283	124	407	68	78	146
		69.5%	30.5%	100.0%	46.6%	53.4%	100%

Table 2. Distribution of X-ray machines in Iraq by manufactures

Manufacturer	Diagnostic			Dental		
	Public	Private	Total	Public	Private	Total
Siemens	53	46	99	6	7	13
Phillips	48	30	78	1	7	8
General Electric	29	10	39	11	-	11
Generay	83	-	83	-	-	-
Watson	30	7	37	-	-	-
Tur	29	6	35	-	-	-
Explor	-	-	-	-	29	29
Lavo	-	-	-	24	-	24
Honda	-	-	-	11	-	11
Ritter	-	-	-	1	5	6
Other Types	8	15	23	6	25	31
Unknown	3	10	13	8	5	13
Total No.	283	124	407	68	78	146
%	69.5	30.5	100	46.6	53.4	100

B. Number of X-ray Examinations :

In the estimation the number of radiographs taken during fluoroscopic examinations were included in the list under radiography and one examination means one exposure. All types of examination concerning radiography and fluoroscopy were classified to 10 forms as shown in Table 3, which also illustrates details regarding the age, sex and area examined in a total number of one thousand patients examined in different diagnostic X-ray machines except

mass miniature radiographies and dental, because it was difficult to know the sex of patients in these last two types of examinations.

Table 3. Frequency in thousand of diagnostic examinations by age, sex and type examination.

Type of Examination	15		16-20		21-30		31-45		46-50		50+		Total		%
	M	F	M	F	M	F	M	F	M	F	M	F	M	F	
Hands	12	5	5	1	15	3	3	3	3	1	1	3	39	16	5.5
Head/Neck	12	25	10	12	37	27	13	22	3	3	8	17	83	1.6	18.9
Feet	7	3	2	7	5	7	7	3	5	1	1	1	27	22	4.9
Chest	25	25	5	32	27	35	23	37	15	10	32	8	127	147	27.4
Vertebral Col.	3	3	3	10	22	7	17	7	5	7	12	7	62	41	10.3
Gall Bladder	1	1	1	1	3	3	1	5	1	5	1	1	8	16	2.4
Stomach	1	1	3	15	32	25	25	22	7	8	12	5	80	76	15.6
Urinary Tr.	15	2	8	10	17	18	11	13	2	11	3	6	56	60	11.6
Pelvis	3	3	1	3	1	5	1	1	1	1	2	1	9	14	2.3
Pregnancy	-	-	-	5	-	5	-	1	-	-	-	-	-	11	1.1
Total	M	79	38	159	1.1	42	72	491	1000						
	F	68	96	135	114	47	49	509							
%		14.7	13.4	29.4	21.5	8.9	11.1	49.1	50.9	100%					

27.4% of the total examination in Table 3 were performed for chest X-ray examination, while the head and neck X-ray examination came to be second in frequency (18.9%), stomach and the surrounding organs (gall bladder and liver) constituted about 18% of the total X-ray examination done. Pelvis X-ray diagnosis constituted about 3.4% of the total X-ray tests. The urinary tract X-ray diagnostic tests constituted 11% of the total examinations.

With regard to age grouping, Table 3 shows that 14.7% of the test were done on patients under the age of 15 years of both sexes. While 13.4% of the tests were done on patients between 16-20 years old.

The age group 21-45 years which constitute the active reproduction age specially in women, reached 51.9% of the total number examined in this table. With respect to the other older age groups, it appears that a relatively small percentage of patients were examined. Regarding the total male to female ratio of the one thousand patients examined in this work, the ratio was nearly one. The official census of the population indicates that there are 5,073,600 males and 5,000,600 females in Iraq at the end of 1972/6/.

The total number of radiographic examinations was  $4.2 \times 10^6$  diagnostic examinations. Nearly 21% them were mass miniature radiographies. While the annual total number of dental X-ray examination was about  $150 \times 10^2$  examinations. The estimation shows that the patients performed in average 2.2 examinations each visit. Therefore it appeared that the frequency is 2 persons from 10 inhabitants were undergone diagnostic examinations yearly. Moreover, about 15 persons out of 1000 had dental X-ray examinations annually.

Table 4a shows the total number of X-ray examinations done on different parts of the body of both sexes and the gonad dose measured from each particular examination.

The man-rad/year received, was the highest in performing the abdomen, which is in the range of  $7.8 \times 10^5$  man-rad/year. The mass chest X-ray miniature constituted the second man-rad/year received. Other clinical examinations gave a significant decrease in man-rad/year dose in comparison with the above mentioned diagnostic tests. The total man-rad/year for all types of

examinations  $8 \times 10^5$  man-rad/year.

Table 4a. Total gonad dose in man-rad due to examinations of both sexes (Figures are taken 1000 exm.per year).

Type of Examination	Male		Female		Total	
	No. exm.	gonad dose man-rad	No. exm.	gonad dose man-rad	No. exm.	gonad dose man-rad
Mass miniature					820	2.700
Extremities	225	0.220	129	0.130	354	0.350
Head & Neck	282	0.280	350	0.360	642	0.640
Chest	423	0.860	500	1.000	922	1.860
Abdomen	751	329	741	445	1492	774 99%
Dental					150	0.300
Total	Gonadal dose $779.85 \times 10^3$ man-rad/year.					

Table 4-b represents the genetically significant dose received by both sexes of the Iraqi population. It is apparent that the dose received from the use of the dental machines is significantly smaller than the dose received from the conventional X-ray diagnostic procedure. The calculation of the annual genetically significant dose resulted in a value of 52 m rad for 1972.

The accuracy of this result is probably of the order of 60%. The number and technical data for measurements are reported by the author in a separate paper/7/.

#### Discussion

Table 4b. Genetically significant dose by sex (mrad/person per year).

There is no general standard system to distribute the X-ray machines over the different parts of Iraq. But one can make his own conclusion from table I, which reflects the relation

Sex	Type of exam.		Total G.S.D. mrad/year	%
	Diagnos.	Dental		
Male	25.62	0.15	25.77	49.65
Female	26.03	0.15	26.18	50.35
Total	51.65	0.30	51.95	100

between the density distribution of the population and the X-ray units. The relatively high number units of (45.7% and 56.1%) of the total conventional diagnostic and dental units respectively in Baghdad area is due to the number of its inhabitants (25.5% of the total population of the country) and the same situation is true in Basrah and Kineva. Another reason for this distribution may be attributed to the preference of most doctors to live and work in large cities. On the other hand, wherever the official numbers of units are high, the number of private units are high too. This may be due to the permission is given only to specialist doctors in radiological fields to possess X-ray units in private clinics. This permission is given to any dentist. It is found that the private specialists in medical radiology themselves are mainly the official ones. Sometimes specialists with high qualification in other fields might be granted a permission too.

A total number of 407 conventional X-ray machines performing an annual frequency of 420 X-ray examinations per 1000 persons. This figure of examinations is similar to the figures obtained in many other countries which had carried out comprehensive survey /5/ while our number of instalations per 1000 of total populations are less. From Table 2 it is shown that there are only 6 firms

which supplied about 90% of the conventional units and 7 firms supplied about 70% of the dental X-ray machines. This situation makes it easy for determination of the dose received from all machines. The average operating peak kilovoltages has been found to be 70 KVP in conventional machines and (50-60) KVP in dental.

Data of Table 3 included sex categories by sex and age. Each category encompassed the types of examinations. It seems that 20% of the total patients are less than 30 years old. The last percentage of patient is to be considered when discussing genetic effects.

The abdominal (stomach, vertebral column, gall bladder and urinary tract) examinations which comprised 33.7% of all the examinations, give exposure values representing about 99% of the average gonadal exposure as shown in Table 4a. In estimation of genetically significant dose, the individual gonad dose is weighed with a factor taking in to account the future number of children expected. However the total gonadal dose is about  $780 \times 10^3$  man-rad/year, the genetically significant dose resulted is 52 mrad/year which is a high figure in comparison to that of other countries having frequencies between 8 and 44 mrad/year/4, 5, 8, 9/. However, this dose is still below the dose recommended by ICRP, which is 5 rem over a period of 30 years.

#### Conclusions

1. The study gives a clear picture of the X-ray unit distribution in different parts of Iraq, which is somewhat satisfactory.
2. The annual number of radiographic examinations is expected to be soon more than  $5 \times 10^6$ . It is thought advisable to start a local film production. As long as films are, for the time being, imported, this study will help very much in the estimation of the annual need of these films.
3. Since the genetically significant dose (52 mrad/year) is higher than that in other countries, beside that we have no data for the past years about the number of diagnostic X-ray examinations per year, it seems that there is an urgent need for the reduction of the annual genetically significant dose from X-ray examinations, although very few people are being exposed.

#### References

1. Ministry of Health, Iraq-Unpublished Observations.
2. Recommendations of the ICRP. ICRP publication No 1 Pergamon Press, Oxford (1959).
3. Recommendations of the ICRP. ICRP-publication No. 8 Pergamon Press (1962).
4. Joel C. Lubenau, et. al. Results of the Pennsylvania department of health dental X-ray survey program. IL. Phys. vol. 19, 151 (1968).
5. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation, UN, No. 16 (4/5216), New York (1962).
6. Koso Ueda-Report on revised projections of population in Iraq by sex and age group from 1957-1980. Baghdad 1970.
7. Mousa S. Abbas-Approximately calculation to the genetically significant dose of the Iraqi population in 1972 from the medical X-ray diagnostic examinations. To be published.
8. Kunz, B, et. al-Studies of the exposure of the population in Gzechozlov a Mia to ionizing radiation. Peaceful uses of atomic energy No II. IAEA-vienna 1972.
9. KashiUmE, Y, et al: Genetically significant dose from diagnostic medical X-ray examinations in Japan, 1969. Health Physics, Volume 23 No 6, 1972.

## RADIOBIOLOGICAL SIGNIFICANCE OF $^{125}\text{I}$ MICRODOSIMETRY

A.R. Reddy and K.K. Nagpal  
Institute of Nuclear Medicine & Allied Sciences  
Probyn Road, Delhi-110007(India).

### Abstract

Significant difference in the microscopic dose distribution due to  $^{125}\text{I}$  and  $^{131}\text{I}$  in the thyroid gland is reported. Its radiobiological significance is studied using varying doses of these two radionuclides in one month old rats. The total body weight increase, thyroidal retentivity of radioiodine, 24 hour uptake and the thyroid weight at the end of the treatment are measured in both the treated and control groups. Thyroid weight is taken as an index of thyroid cell population and any reduction in its increase with age compared to that in controls(which is taken as 100% cell survival)is considered as due to the impairment of proliferative capacity of the thyroid cells. Average gland doses due to  $^{125}\text{I}$  and  $^{131}\text{I}$  for 50% cell survival are 40500 and 18500 rad respectively. The 24 hour uptake is significantly reduced in 71 uCi  $^{131}\text{I}$  treated rats whereas in all other treated ones there is no change compared to controls. Body weight increase is impaired in both the treatments, more so with  $^{125}\text{I}$ . These differences are explained in terms of differences in dose distributions across a thyroid follicle due to  $^{125}\text{I}$  and  $^{131}\text{I}$ .

### Introduction

For the same activity administered the mean dose to the thyroid gland is 2-3 times less with  $^{125}\text{I}$  compared to  $^{131}\text{I}$ . Microscopic dose distribution across a thyroid follicle, which is a basic unit of the thyroid gland, shows that with  $^{125}\text{I}$  the dose at the colloid-cell interphase is 2-6 times and 4-10 times that over the nucleus and that at the basal membrane respectively depending upon the colloid content in the gland whereas with  $^{131}\text{I}$  it is practically uniform<sup>1-3</sup>.

The radiobiological significance of this difference in microscopic dose distribution was studied in the adult rat thyroid by different workers<sup>4-6</sup>. However, the normal adult rat thyroid has a closed, well differentiated cell system with little proliferation and upon irradiation(with about 500 rads of X-rays) shows little change as judged by weight, cell counts or DNA and RNA content<sup>5</sup>. So to see the effects of irradiation on thyroid cell population, particularly on its proliferative capacity, it is necessary to promote the cell multiplication artificially, say by administration of methyl thiouracil.

Instead, young rats of one month age were chosen for the present investigation. At this age their thyroid cells are rapidly proliferating<sup>7</sup> and the thyroid weight increases by a factor of 2-3 within a span of 2 months. So a study of the radiation effects is possible without the administration of a drug.

### Materials and Methods

One month old Holtzman strain male rats were used for the study. They were fed with standard dry pellet Hindustan Liver rat food and tap water ad libitum. 5 rats were taken for each dosage. One of the following doses in a volume of 0.2 ml was injected intraperitoneally to each group: 100, 50, 25, 12.5 and 2.5 uCi of  $\text{Na}^{125}\text{I}$ ; 71, 24.2 and 4.8 uCi of  $\text{Na}^{131}\text{I}$ . Two groups of 5 rats each served as respective controls for the two radionuclides. The total body weight and thyroid retention of radioiodine(by in vivo counting with an end window



G.M. Counter over the thyroid) were measured periodically.

When the rats attain 3 months age they were injected intraperitoneally a tracer dose of  $^{125}\text{I}$  for measuring the thyroid uptake at the end of the treatment. 24 hours after administration of the tracer the rats were sacrificed. The two lobes of the thyroid from each rat were taken out and weighed correct to 0.2 mg. The thyroidal activity was obtained using a Nuclear Chicago Autogamma well counter and was expressed as the percentage of the injected activity per unit thyroid weight.

### Results and Discussion

#### Variation of Thyroid Weight and Radiiodine Uptake with Age:

Fig.1 shows the variation of thyroid weight and 24 hour radioiodine uptake as a function of age. The mean thyroid weight of 30 d old rats is  $6.8 \pm 2.3$  mg for a mean body weight of  $45.2 \pm 7.3$  g while that of 100 d old ones is  $24.2 \pm 2.6$  mg for a mean body weight of  $210 \pm 18.1$  g corresponding to an increase in the thyroid weight by a factor of 3.5. However, the variation in thyroid weight expressed as a fraction of body weight is only from  $0.154 \pm 0.006$  at 30 d to  $0.115 \pm 0.006$  at 100 d. The 24 hour thyroid radioiodine uptake expressed as a percentage of the injected dose per unit thyroid weight in mg varies from  $0.059 \pm 0.0049$  to  $0.016 \pm 0.0049$  with the age of the rat. Higher uptake per unit thyroid weight in the one month old rats indicates the hyperactivity of their thyroids.

#### Biological Half-life of Radiiodine in Thyroid:

The in vivo thyroid activity is monitored periodically for all the animals and Fig.2 gives the retention pattern in arbitrary units as a function of time. In case of  $^{125}\text{I}$ , the retention could be expressed as a single exponential with a half-life of 10-13 d, irrespective of the dose administered. So only the data corresponding to 100 uCi group is presented in Fig.2. The pattern of retention of  $^{131}\text{I}$  was more complex, consisting of at least two components. The size and half-life of the two components depended on the dose administered. The half-life of the fast component increased and that of the slow component decreased with the decrease of dose.

#### Estimation of Average Gland Dose:

The average gland dose, D, is estimated for each treated rat from the knowledge of its 24 hour uptake, U, the thyroid weight, m, in g and the effective half-life, T, in h using the well known equation

$$D = 1.44 \text{ AUT } \sum \Delta_i \phi_i / m \quad \text{rad}$$

where A is the activity administered in uCi,  $\Sigma$  the summation sign,  $\Delta_i$ , the equilibrium absorbed dose constant in g-rad/uCi-h, and  $\phi_i$ , the absorbed fraction in the thyroid gland for i th component. The value of  $\Sigma \Delta_i \phi_i$  is calculated to be 0.0506 for  $^{125}\text{I}$  using the values of  $\Delta_i$  and  $\phi_i$  listed in MIRD pamphlets<sup>9-11</sup> and includes both penetrating and non-penetrating components. For  $^{131}\text{I}$  the contribution due to photons is neglected. Considering the thyroid as a sphere the absorbed fraction for  $^{131}\text{I}$  beta radiation is obtained using Berger's tabulation of  $\phi$  for different sphere sizes. The value of  $\phi$  is about 0.8 for the rat thyroid<sup>10</sup>.

The mean gland dose corresponding to 100 uCi  $^{125}\text{I}$  treated group is  $34322 \pm 9560$  rad. The value for 71 uCi  $^{131}\text{I}$  treated group is  $21820 \pm 4034$  rad.

### Body Weight vs. Days after Administration of Activity

Figs. 3 and 4 give the ratio, Y, of the body weight on the day of observation to that on the day of the administration of activity, plotted against the time, X, elapsed from the day of administration of activity for the  $^{125}\text{I}$  and  $^{131}\text{I}$  treated groups respectively. The data in each case is statistically analysed and the respective regression equations, correlation coefficients, r, and the standard errors,  $S_{\text{MY}}$ , are given in the same figures.

The data shows that there is a reduction in the body weight ratio of the treated groups in comparison to the control groups and the reduction is more pronounced in the case of  $^{125}\text{I}$ . The reduction in the body weight indicates that there is probably a disturbance in the release of the right amount and right type of iodinated compounds which affect body growth and that the disturbance is pronounced with  $^{125}\text{I}$ .

### 24 hour Radioiodine Uptake at the end of Treatment:

The 24 hour uptake was obtained for all the treated and control groups by administration of a tracer dose of  $^{125}\text{I}$ , just one day prior to sacrifice. The results are given in Table 1. The uptake value for all the treated groups (except that of 71 uCi  $^{131}\text{I}$ ) is the same as that of the corresponding control group (C). The lower uptake of 71 uCi  $^{131}\text{I}$  group may be due to a damage of the iodine trapping mechanism and/or due to a decrease in the number of follicular cells per unit thyroid weight.

Table 1: Radioiodine Uptake at the end of  $^{125}\text{I}$  and  $^{131}\text{I}$  treatments

$^{125}\text{I}$ treatment		$^{131}\text{I}$ treatment	
Mean Gland Dose (rad)	24 hour uptake	Mean Gland Dose (rad)	24 hour uptake
34322 (100 uCi)	0.0284 ± 0.0059	21800 (71 uCi)	0.020 ± 0.0094
24867 (50 uCi)	0.033 ± 0.0092	15793 (24.2 uCi)	0.044 ± 0.0099
12604 (25 uCi)	0.030 ± 0.0078	2500 (4.8 uCi)	0.044 ± 0.0047
6182 (12.5 uCi)	0.025 ± 0.0073	C	0.037 ± 0.0047
1299 (2.5 uCi)	0.032 ± 0.011		
C	0.029 ± 0.0065		

### Percentage Cell Survival vs. Average Radiation Dose to the Thyroid:

In Fig. 5 the percentage cell survival is plotted against the average gland dose for both  $^{125}\text{I}$  and  $^{131}\text{I}$  treated groups. The thyroid weight at the end of the treatment normalised to the then body weight is taken as an index of cell survival. The percentage cell survival is calculated by the following equation:

$$\% \text{ cell survival} = (m/M)_j \times 100 / (m/M)_0$$

$$\text{where } (m/M)_j = \frac{\sum_{i=1}^{n_j} (m_{ij}/M_{ij})}{n_j} \text{ and } (m/M)_0 = \frac{\sum_{i=1}^{n_0} (m_{i0}/M_{i0})}{n_0}$$

Here  $m_{ij}$  and  $M_{ij}$  are the thyroid and body weights of the  $i$ th rat in  $j$ th treated group consisting of  $n_j$  number of rats.  $m_{i0}$  and  $M_{i0}$  are the thyroid and body weights of  $i$ th rat in the control group consisting of  $n_0$  rats.

The values of  $D_{50}$  (the gland dose for 50% cell survival) for  $^{131}\text{I}$  and  $^{125}\text{I}$  respectively are 18500 and 40500 rad. Their ratio, 0.46, suggests that RBE for  $^{125}\text{I}$  compared to  $^{131}\text{I}$  is less than one which is contrary to expectation. This may be explained in terms of the differences in microscopic dose distribution across a thyroid follicle due to  $^{125}\text{I}$  and  $^{131}\text{I}$ .

Using Berger's scaled absorbed dose distributions<sup>12</sup>, the dose distribution across a thyroid follicle was computed assuming colloid mass as 50% of the gland mass. In each follicle it is considered to be in the form of a sphere of 25  $\mu\text{m}$  radius surrounded by follicular cells of length 9  $\mu\text{m}$  with their nuclei 3  $\mu\text{m}$  away from the apical membrane. In Fig.6, the ratio of dose rate at any point  $R_s$ , to that at the center of the colloid,  $R_0$ , is plotted against the distance  $s$ . The values of  $R_s/R_0$  at the colloid cell interphase, over the nucleus and at the basal membrane are 0.49, around 0.25 and 0.14 respectively. The average gland dose corresponds to that at colloid-cell interphase as the colloid content is assumed as 50% of the total gland mass with all the thyroid iodine concentrated in it<sup>2</sup>.

So in case of  $^{125}\text{I}$  treatment, the nuclei of the follicular cells at 50% cell survival level received only about 20000 rad as against the gland dose of 40500 rad. For  $^{131}\text{I}$ , however, dose distribution across the follicle is more or less uniform and so the gland dose (18500 rad for 50% cell survival) itself is the dose to the nucleus. Thus, if the dose to the nucleus is considered the 50% survival dose is same with both the nuclides.

The same microdosimetric considerations explain the differences found in 24 hour uptake at the end of  $^{131}\text{I}$  and  $^{125}\text{I}$  treatments and their respective controls. The percentage cell survival at which the significant reduction in 24 hour uptake was observed with  $^{131}\text{I}$  is 44.3%. The corresponding gland doses with  $^{131}\text{I}$  and  $^{125}\text{I}$  are 21800 and 46500 rad respectively. The dose at the basal membrane (which is considered as the site for trapping of iodine) of the follicular cell of 9  $\mu\text{m}$  length is about one fourth that at the colloid-cell interphase, that is, about 12000 rad in case of  $^{125}\text{I}$ . So the absence of reduction in 24 hour uptake in  $^{125}\text{I}$  treated groups is due to the inhomogeneity in dose distribution resulting in only about 12000 rad at the site of trapping compared to the gland dose of 46500 rad. Since in case of  $^{131}\text{I}$  treated group the gland dose (21800 rad) itself is the dose to the basal membrane also the reduction in 24 hour uptake observed can be expected.

#### Acknowledgement

Authors are indebted to M/s A.Nagaratnam, and H.S. Yadav for many helpful discussions and to Col S.K. Mazumdar, Director, INMAS, for permission to present the paper.

#### References

1. F.C.Gillespie, J.S.Orr and W.R.Greig, Brit.J. Radiol., 43,40,1970.
2. A.R.Reddy, K.G.K.Sastry, M.M.Gupta and A.Nagaratnam, Proc.Symp. on Basic Mechanisms in Radiation Biology and Medicine, New Delhi, 711, 1971.
3. Y.Feige, A. Gavron, E. Lubin, Z.Lewitus, M. Ben-Porath, J.Gross, E.Loewinger, Proc. Symp. on Biophysical Aspects of Radiation Quality, Lucas Heights, 383, 1971.
4. J.Gross, M.Ben-Porath, A. Rosin and M.Eloch, Thyroid Neoplasia (Eds. S. Young and D.R.Inman), Academic Press, New York, 291, 1968.
5. W.R.Greig, J.F.E. Smith, W.P. Duguid, C.J. Foster, and J.S. Orr, Int. J. Radiat. Biol., 16, 211, 1969.
6. W.R.Greig, J.F.E. Smith, J.S. Orr and C.J. Foster, Brit.J. Radiol. 43, 542, 1970.
7. J.Logothetopoulos, Endocrinology, 73, 349, 1963.
8. I.Doniach, Brit. J. Cancer, 11, 263, 1957.
9. L.T.Dilmann, MIRd pamphlet No.4, J. Nuc.Med. Suppl. 1, 1968.
10. M.J.Berger, Proc. Symp. Medical Radionuclides, Radiation Dose and Effects, USAEC, 85, 1970.
11. M.J.Berger, MIRd pamphlet No.2, J. Nuc. Med. Suppl. 1, 1968.
12. M.J. Berger, MIRd pamphlet No.7, J Nuc. Med. Suppl. 5, 1971.

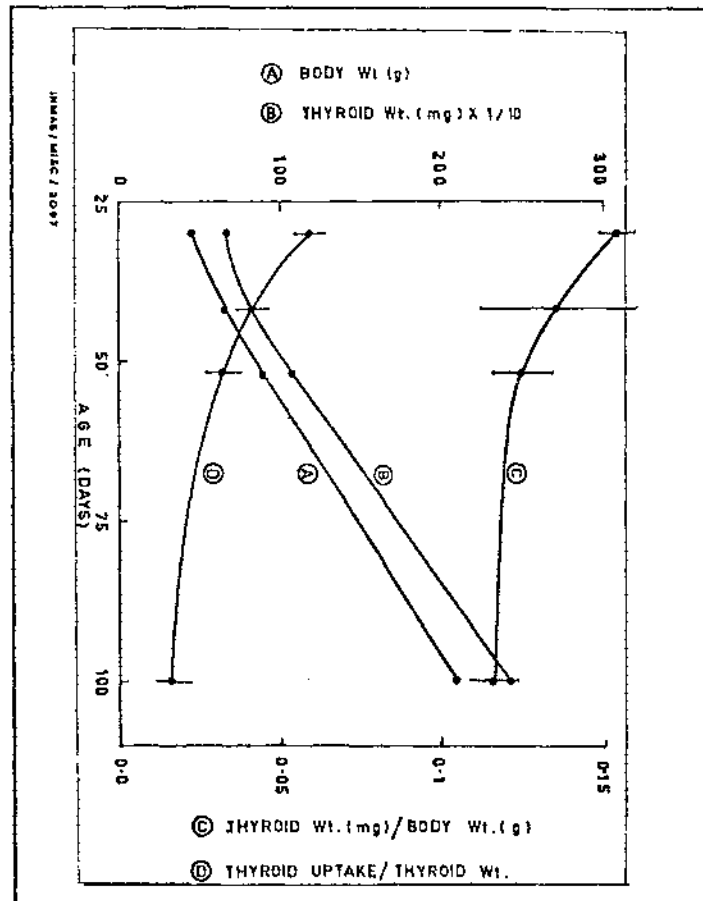


FIG. 1. Variation of body weight (A), thyroid weight (B), thyroid weight expressed as a fraction of the body (C), and 24 hour thyroid radioiodine uptake per unit thyroid weight (D) with age of rats.

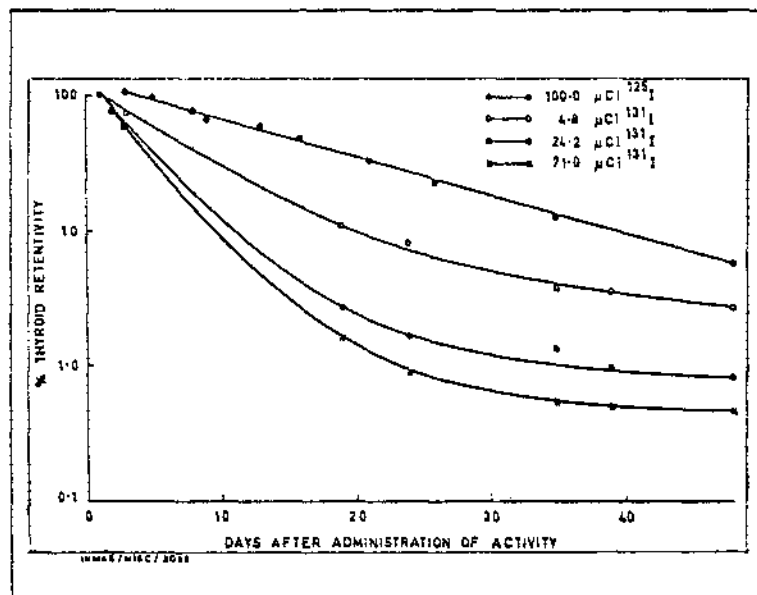


FIG. 2. Thyroid retentivity of radioiodine as a function of time in days after the administration of the activity.

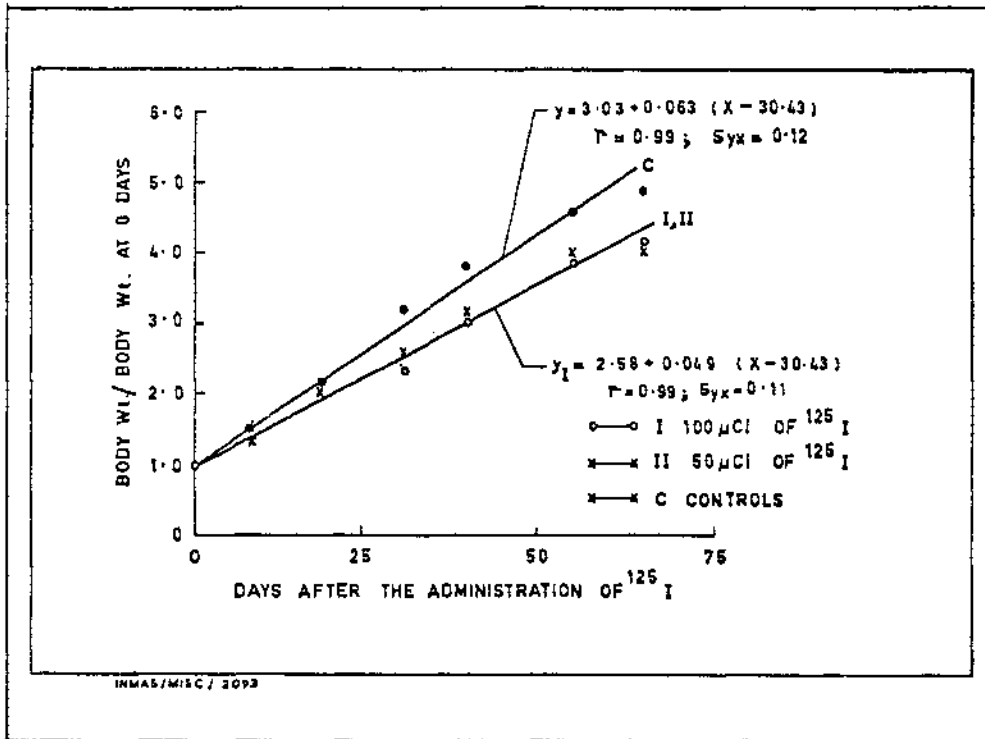


FIG. 3. Variation of the body weight ratio of  $^{125}\text{I}$  treated groups and the respective controls as a function of time after the administration of the activity.

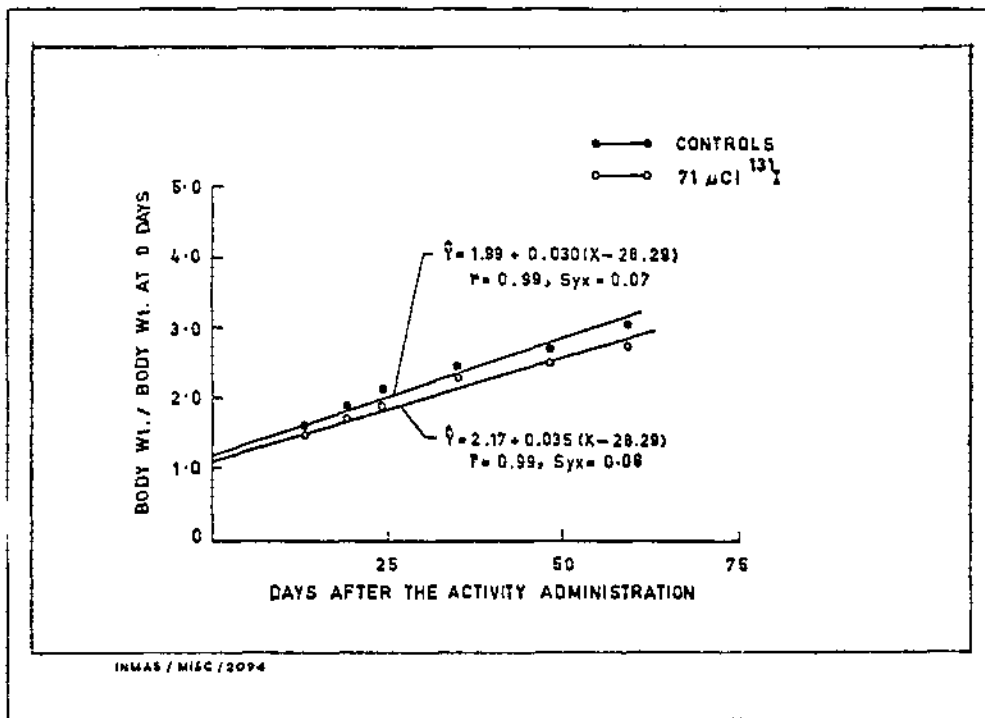


FIG. 4. Variation of the body weight ratio of  $^{131}\text{I}$  treated groups and the respective controls as a function of time after the administration of the activity.

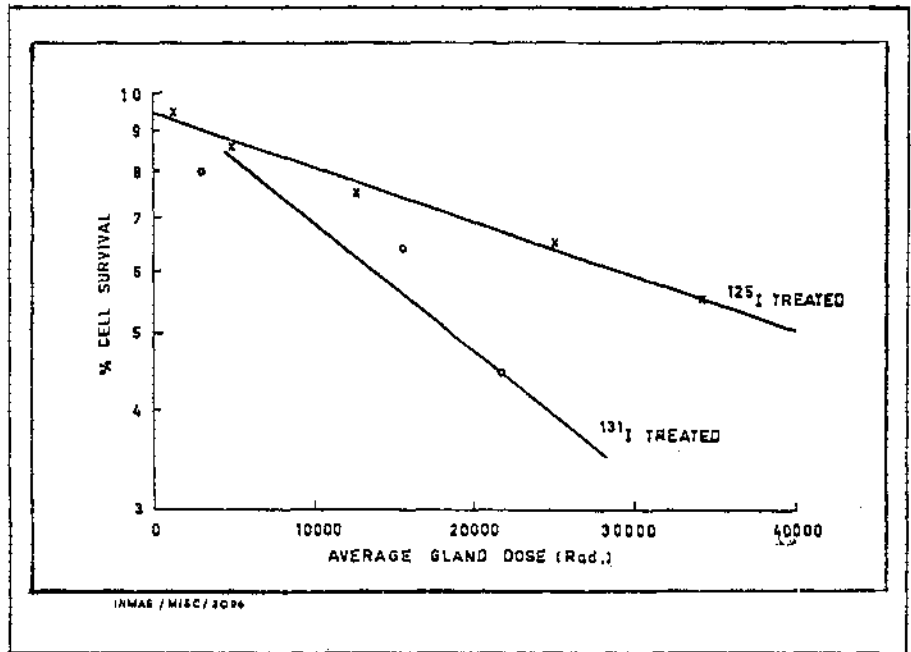


FIG. 5. Cell survival curves for  $^{125}\text{I}$  and  $^{131}\text{I}$  treated groups.

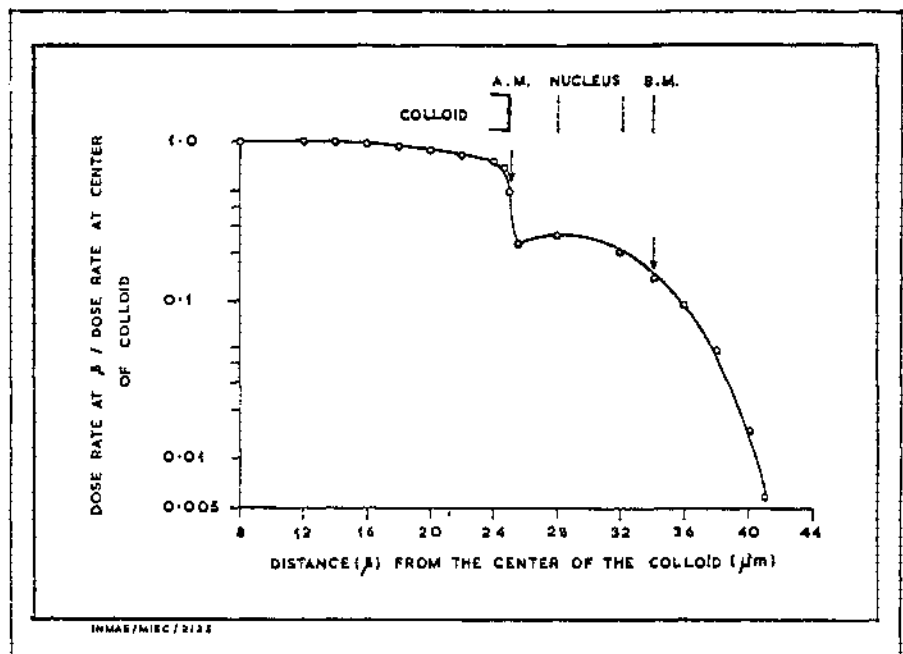


FIG. 6. Dose rate ratio,  $R/R_0$ , for  $^{125}\text{I}$  as a function of distance from the center of the colloid. At distances below  $8\ \mu\text{m}$  the ratio is 1.0. The two arrows point to the positions of Apical Membrane (A.M.) and Basal Membrane (B.M.) of the follicular cell. Position of the nucleus is also shown.

DOSIMETRY OF INTERNAL EMITTERS IN NUCLEAR MEDICINE  
AND RADIATION PROTECTION : AT WHAT LEVEL

A. Nagarathnam and A.R. Reddy  
Institute of Nuclear Medicine & Allied Sciences  
Probyn Road, Delhi 110007(India)

Abstract

The inadequacy of conventional dosimetry at the organ level assuming a uniform activity distribution has been brought out in specific examples. In each case dosimetry at different anatomical levels is presented bringing out the probable understanding and lacuna in the radiobiological consequences of such dosimetry. Bone dosimetry of alpha and beta emitters is presented. Dose to whole kidney and differential doses to medulla and cortex from  $^{203}\text{Hg}$ -neohydrin are described. The mean dose to lung from  $^{131}\text{I}$ -MAA, the mean local dose to capillary bed and to capillary epithelium adjacent to an MAA particle are reviewed. The importance of Auger electron emission and the consequent transmutational effects is discussed with the examples of  $^{125}\text{I}$  in thyroid,  $^{125}\text{I}$ -UDR in proliferating cells,  $^{59}\text{Fe}$  in erythrocytes. High doses to limited portions of fingers while handling  $^{113\text{m}}\text{In}$  and  $^{99\text{m}}\text{Tc}$  labelled pharmaceuticals is presented.

Introduction

In nuclear medicine the clinician wants to know what are the potential hazards to a patient if he undergoes a particular investigative procedure which is expected to yield diagnostic information of clinical value. In the case of radiation protection the similar question is: what are the potential hazards to the worker under given exposure situation. The focus of interest is the biological effect which is the end-point in a complicated chain of events at the physical, chemical and physiological levels. Conventional dosimetry is mainly concerned with the primary event, viz. physical step, and the absorbed dose is an indicator only of physical events that happen at the macroscopic level. But we are far from understanding the biological significance of the absorbed dose. Partial consideration to the secondary events is attempted to be given by assigning a somewhat arbitrary value for the RBE or QF and quoting a rem dose.

For a proper evaluation of the biological effect, we should know the microscopic spatial and temporal distribution of the primary and secondary events. This leads to a detailed consideration of several factors, some associated with the radiation alone like radiation quantity and dose rate, others associated with the target as well, like location of radionuclide in the cell, the biochemistry of the labelled compound, bond rupture resulting from nuclear recoil after beta emission, chemical effect of nuclear transmutation on functional integrity of molecule, effect of sudden changes of charge on daughter nuclide (particularly important for isotopes decaying by electron capture), oxygen tension, cells at risk, radiosensitivity of cells in question, etc.

Further, concepts like LET and absorbed dose are macroscopic quantities or 'expectation values'. As the volume over which the absorbed energy is computed is reduced, the fluctuations associated with the stochastic nature of the interaction process assume increasing importance; concepts like 'event size', 'local energy density' and 'event size spectrum' have then to be introduced. This approach has not yet been made in practical situations of concern in internal dosimetry.

Very often we are not quite clear as to what is the biological end-point that is of relevance, although it is generally accepted that for comparatively

low doses as are encountered in routine occupational exposure or diagnostic nuclear medicine procedures, the effects may be classified into two categories, viz (i) those leading to the impairment of the functional integrity of the organ (this may be due to reduction in number of functioning cells or fibrosis with scarring) and (ii) induction of malignancies<sup>1</sup>.

With all these complications, the question arises: At what level should dosimetry be done? Can we be satisfied with the conventional calculation of absorbed dose at the organ level assuming a uniform concentration of radionuclide? Or should we go down to the tissue, cell and even subcellular level? Should we consider the stochastic nature of the interaction process and enter into details of the microdosimetric concepts? What degree of sophistication is necessary and what degree sufficient?

The problem is discussed in terms of some well-known examples of practical interest.

### Bone Dosimetry

Bone dosimetry is a classical example of the inadequacy of the conventional organ dose computation for an assessment of the potential risks. The inhomogeneous structure of bone and bone cavities (where the linear dimensions of the inhomogeneities are frequently of the same order as the range of the ionizing particles), the varieties of cells at risk and the non-uniformity of distribution of the radioisotope make the situation complex. A good deal of effort and ingenuity have gone into the solution of the problem from both the theoretical and experimental sides. From a consideration of the critical tissues for radiation damage, it is usual to calculate the following separately<sup>2</sup>:

- i) Dose to a very small tissue-filled cavity in the bone matrix,  $D_0$  (to evaluate risk to osteocytes, cells lining Haversian canals and blood vessels in Haversian systems which are concerned with maintaining the functional integrity of bone as a living tissue).
- ii) Mean dose to endosteal cells near the surface of bone trabeculae in the marrow cavities,  $D_s$  (osteogenic sarcoma risk).
- iii) Mean dose to active marrow in trabecular cavity  $D_m$  (leukaemogenesis risk).

Typical results for radium and strontium-90 are shown in Table 1<sup>2</sup>.

Table 1  
Dose rates in rad/year from skeletal burden of 1 uCi

	Radiation	$D_0$	$\bar{D}_m$	$\bar{D}_s/\bar{D}_m$
$^{226}\text{Ra}$ series	$\alpha$	36	10.5	7
$^{90}\text{Sr} + ^{90}\text{Y}$	$\beta$	2.7	1.1	0.5

It is only by a detailed consideration of the cells at risk that we are able to perceive a major difference between the alpha and beta emitters. In view of the limited range of alpha rays, the bone marrow dose is only a small fraction of the endosteal dose in the case of alpha emitters. This is borne out by experience where we find that the incidence of leukaemia in radium



poisoning cases has been negligibly low and osteogenic sarcomas (and cancers of paranasal sinuses) are more common. With  $^{90}\text{Sr}$  both leukaemia and osteogenic sarcomas have been induced in animals.

We shall next consider the question of non-uniformity of distribution of radium (and strontium) in bone. In addition to a diffuse distribution in the bone matrix, hot spots also occur where the local concentration may be 30-40 times the average concentration<sup>3</sup>. For  $^{226}\text{Ra}$ , the range of variation of concentration is faithfully reflected in a corresponding range of variation of dose rates between the different concentration sites (factor of 16). Due to the longer range of beta rays from  $^{90}\text{Sr}$ , the local dose rate variations are not that marked (factor of 3). We still do not know whether the hot spots play a role in radiation damage to bone and what the biological significance of the non-uniformity of damage is.

#### Neohydrin Dosimetry

Controversy was intense a few years ago whether  $^{203}\text{Hg}$ -neohydrin should not be banned as a radiopharmaceutical for kidney and brain scanning in view of the high kidney doses. Neohydrin concentrates primarily in the cortex from where it is eliminated only very slowly. Since the cortical mass is about half that of the kidney, the dose to cortex would be twice that to the kidney as a whole had the isotope been uniformly concentrated. The cortex dose can be taken as 146 rads and the medulla dose as 77 rads per millicurie of  $^{203}\text{Hg}$ -neohydrin<sup>4</sup>.

We may discuss the question a little further. Is there firm evidence to show that 100 rads to the cortex is necessarily more harmful than 50 rads to the kidney as a whole? What is the biological end-point we are looking for? The natural incidence of malignant tumours of the kidney is quite small; also there is as yet no established case of radiation-induced kidney tumour. If malignancy is not the critical end effect, we have next to consider impairment of functional integrity. At the levels used in diagnostic procedures, gross impairment like acute or chronic nephritis is ruled out and much milder damage, which cannot be unambiguously pinpointed, must be considered. It appears that the fine vasculature is the histological site of damage of primary importance in the pathogenesis of radiation induced nephrosclerosis; the renal epithelium is relatively resistant but it may degenerate as a result of damage to the fine vasculature<sup>5</sup>. Since the proximal and distal parts of the tubules lie mainly in the cortex, it is not inconceivable that secondary tubular damage may be somewhat more intense from 100 rads to cortex than 50 rads to total kidney. On the other hand, it has been pointed out<sup>6</sup> that the effective surface area of the renal cortex is about 4 times larger than the surface area of the kidney. Hence the escape of the beta radiations from the cortex will reduce this dose variation factor of 2 by an amount which has not yet been computed. The uncertainty remains.

#### $^{131}\text{I}$ -Human Serum Albumin Macroaggregates for Lung Scanning

Uncertainty in the effective tissue mass to be considered in the dose computation can be illustrated by  $^{131}\text{I}$  HSA macroaggregates in lung scanning. If a homogenous distribution of MAA in lungs is assumed, average dose to lung is about 1.5 rads for 30  $\mu\text{Ci}$ <sup>7</sup>. If a more realistic volume distribution is assumed, viz. the capillary bed of the lungs with a mean diameter of 10  $\mu\text{m}$ , the average local absorbed dose is nearly 5.5 rad. At the cellular level, absorbed dose to capillary epithelium adjacent to an MAA particle is several orders of magnitude higher than the average local absorbed dose, although this extreme dose is received only over a distance of one or two cell thicknesses. The problem is the anatomical level at which the dose is to be evaluated. If the induction of malignancy is the end-point of interest, the integral dose is

probably a valid indicator of the potential hazard. We do not yet know the biological significance of the very high level of local absorbed dose.

A somewhat similar situation exists in the field of radiation protection for assessing hazards from plutonium inhalation.

#### Significance of Auger Effect

Radionuclides decaying by electron capture and isomeric transitions are attractive in in vivo applications since they do not emit particulate radiations which give a radiation dose to the organ but do not contribute any diagnostic information. Several such radionuclides are now in common use, e.g.  $^{51}\text{Cr}$ ,  $^{55}\text{Fe}$ ,  $^{57}\text{Co}$ ,  $^{58}\text{Co}$ ,  $^{67}\text{Ga}$ ,  $^{75}\text{Se}$ ,  $^{85}\text{Sr}$ ,  $^{123}\text{I}$ ,  $^{197}\text{Hg}$  and  $^{99\text{m}}\text{Tc}$ ,  $^{113\text{m}}\text{In}$ . Certain special features of the electron capture process and the process of internal conversion consequent on gamma photon emission are of great relevance. After these two processes, the K or L shell vacancy initiates orbital excitation and electrons fall down from outer orbits successively into the vacancies in inner orbits. Excess energy is lost by X-ray emission in part but a large part of de-excitation occurs through the emission by the Auger effect of several electrons of low energy with a range in tissue of less than a micron. They, therefore, give a very high local dose to the tissue over a micron length. In addition their LET is very high and hence a correspondingly high RBE/QF will have to be postulated, leading to an intense local rem dose. We are still far from understanding the precise biological significance of this peculiar feature but some indications are available in the case of  $^{125}\text{I}$  dosimetry which we shall touch upon subsequently.

Another consequence of Auger electron emission needs attention. As a result of the release of several Auger electrons, the daughter atom is left with a strong positive charge. If this charged nuclide is bound within a molecule it attracts electrons from various molecular positions and the positive charges are distributed throughout the molecule. The various positively charged atoms within the molecule strongly repel each other, which may lead to a virtual 'Coulombic explosion' of the molecule.

#### $^{125}\text{I}$ Dosimetry

Conventional macroscopic dosimetry of  $^{125}\text{I}$  in thyroid has been shown to be entirely inadequate in view of the special characteristics of the radiations from the radionuclide, and one has to go down to subcellular microdosimetry for obtaining a better understanding of the possible biological effects of  $^{125}\text{I}$ . Since the range of Auger electrons is small compared to the dimensions of the thyroid cell, the cell-colloid interface or the apical membrane, which is the seat of thyroid hormone biosynthesis, receives a high dose. On the other hand the nucleus which is farther away gets only about one-fourth the dose to apical membrane. The variation between the nuclear and apical membrane doses is accentuated in thyrotoxic conditions in view of the greater distance of the nucleus from the apical membrane of the columnar cell of the thyrotoxic gland. The clinical significance of these observations has led to intense interest in the use of  $^{125}\text{I}$  for therapy of thyrotoxicosis in preference to  $^{131}\text{I}$ . However the clinical experience as well as the follow-up periods are as yet too small for an unambiguous conclusion to be drawn<sup>8</sup>.

The biological significance of the high LET of the Auger electrons and the possibility of Coulombic explosion mechanisms have not yet been clarified. Perhaps since the  $^{125}\text{I}$  decay takes place mainly in the colloidal gel outside the apical membrane the latter effect may not be critical<sup>9</sup>.

On the contrary if  $^{125}\text{I}$  is attached to essential structures such as DNA the influence of the charge transfer processes and the associated Coulombic explosion effects may be comparable to if not outweigh the radiation effects in producing the biological effect. This has been demonstrated<sup>10,11</sup> while studying the relative effect of varying doses of  $^{125}\text{I}$ -UdR,  $^{131}\text{I}$ -UdR and  $^3\text{H}$ -TdR on proliferating mouse cells *in vivo*. The toxicity of  $^{125}\text{I}$ -UdR was reported to be 10 times greater than that with  $^3\text{H}$ -TdR when these specific precursors of DNA were utilised by either bone marrow cells, or proliferating cells in the whole body in general. A similar finding was reported with ascites tumor cells also. The greater radiotoxicity of  $^{125}\text{I}$ -UdR has been explained as due to a variety of factors including (i) differences in energy deposition in the cell nucleus per disintegrating atom (ii) greater inhomogeneity in the distribution of energy around the site of decay in the case of  $^{125}\text{I}$  (iii) transmutional effects of  $^{125}\text{I}$ , specially the consequences of molecular explosion.

### $^{55}\text{Fe}$ Dosimetry<sup>12</sup>

$^{55}\text{Fe}$ , an electron capture radionuclide, is an important neutron activation product found in fallout. Levels as high as 3 pCi/mg blood have been recorded in New York residents. It has been computed that the dose to erythrocytes is about 10 times that to whole blood since the Auger electrons deposit their energy entirely within the erythrocyte itself where the  $^{55}\text{Fe}$  is tagged. Dose to aggregates of ferritin molecules in which highest concentration of iron is found has been calculated to be about 200 times that to erythrocyte. However, the critical tissue in this case is perhaps the erythrocyte precursor cells in the bone marrow wherein a concentration around one-third of that in erythrocytes has been found, leading to a dose around 3 times the blood dose.

### Skin Dosimetry

Skin dosimetry has acquired some urgency in view of the increasing use of short-lived isotopes like  $^{113\text{m}}\text{In}$  and  $^{99\text{m}}\text{Tc}$  in nuclear medicine. In the milking of the generator, the preparation of the radiopharmaceuticals and injection to the patient, levels of several tens of millicuries have to be handled at a time. The tips of two or three fingers, in particular, get exposed to significantly high doses (around 10 mrem/mCi-min). If the ICRP dose limit of 75 rem/year to hands is not to be exceeded, we would be severely curtailed in the scope of work; not more than two or three brain scanning preparations can be handled per person per week.

In this connection the health physicist turns to ICRP for guidance. A report of an ICRP Task Group<sup>1</sup> has something to say on skin dosimetry. The report recognises that the end-point of relevance here is not carcinogenesis, since the skin is relatively highly radioresistant, but radiation dermatitis. According to the report, in the case of irradiation of part of a tissue, the significant parameter is the mean dose to the entire tissue. If only a fraction of tissue is exposed, the dose allowed can be 1/f of dose limit for the whole organ. On this basis, if the dose limit for entire skin with an area of about 2 square meters is 30 rem/year, the dose to 1 cm<sup>2</sup> (of the order of high exposure areas of finger tips) could go as high as 60 kilorems. So why need we worry?

But the situation is not that simple. There is a limit beyond which the above concept cannot be extended. A vital consideration is the range of dose rate over which effective linearity of dose response can be assumed to hold. The point at which departure from linearity occurs will depend on the precise cellular mechanisms involved and the extent to which abscopal effects come into play. It may happen that at high doses the response may be higher than

would be predicted from a linear hypothesis since a number of contiguous cells are affected and irreparable physiological damage occurs. The Report says that linear response can be assumed up to hundred rems per year and possibly several hundred rems per year, and recommends that 'the present limit of 30 rem/year averaged over 1 cm<sup>2</sup> of skin be increased to at least 100 rems in a year with a proviso that irradiation of the same area year after year should be avoided if possible'. The health physicist wishes that the ICRP Task Group had categorically set a specific limit, say 500 rems per year, instead of vaguely leaving it at, 'at least 100 rems per year', so that he could ask the technicians, with some authoritative sanction behind him, to accept a higher work load with the generators. Note again the words, 'if possible'. In the present case of working with generators the irradiation is going to be received by the same area year after year unless a right handed person could be persuaded to become left-handed. Of course there is no sanctity about the limit of 1 cm<sup>2</sup> which is taken as the 'significant area' for averaging and the Report says '1 cm<sup>2</sup> seems reasonable on grounds of operational convenience'.

#### Conclusion

In this present paper dosimetry at different anatomical levels has been presented with the help of specific examples. In each case probable biological significance of such dose estimation also has been brought out. However, still there are several uncertainties in the biological significance of such detailed dose estimates and the importance of the possible transmutational effects with electron capture radionuclides.

#### Acknowledgement

Authors' thanks are due to Col. S.K. Mazumdar, Director, INMAS, for permission to present this paper.

#### References

1. ICRP Publication No. 14, Pergamon Press, London, 1969.
2. ICRP Publication No. 11, Pergamon Press, London, 1968.
3. F.W. Spiers, *Radioisotopes in the Human Body: Physical and Biological Aspects*, Ch. V, Academic Press, New York, 1968.
4. W.S. Snyder and M.R. Ford, personal communication, 1970.
5. P. Rubin and G.W. Casarett, *Clinical Radiation Pathology*, Vol. I, Ch. 8, W.B. Saunders, Philadelphia, 1968.
6. E.M. Smith, Proc. Symp. on Radioactive Pharmaceuticals, USAEC, 649, 1966.
7. D.E. Tow et al., *Am. J. Roent.*, 96, 664, 1966.
8. A.R. Reddy, INMAS/HP/10-72, 1972.
9. H.H. Ertl, et al., *Phys. Med. Biol.*, 15, 447, 1970.
10. L.E. Feinendegen et al., Proc. Symp. On Biophysical Aspects of Radiation Quality, IAEA, 419, 1971.
11. K.G. Hofer and W.L. Hughes, *Rad. Res.*, 47, 94, 1971.
12. M.E. Wrenn, Proc. First Congress of IRPA, Pergamon Press, 843, 1968.

## CONTAMINATION OF AIR AND SURROUNDINGS BY PATIENTS TREATED WITH LARGE QUANTITIES OF IODINE 131 FOR THYROID CARCINOMA

C.H. Marshall, Ph.D., R. Chandra, Ph.D., Manfred Blum, M.D.

New York University Medical Center  
560 First Avenue  
New York, New York 10016

### Abstract

On six occasions patients were treated with radioiodine for thyroid carcinoma and were isolated. The administered doses were in the range of 100 - 200 mCi of  $^{131}\text{I}$ . All items from the room were collected, to determine the typical contamination deposited during the therapy. Continuous air monitoring was also performed. The total contamination collected on such items as bedding, towels and eating utensils was typically of the order of 1 mCi. Contamination of these items varied erratically during the first three days of therapy. Abnormally high levels of contamination were associated with certain medical disorders. Air burdens were in the range of 10 - 200 pCi per litre during the first day and usually declined progressively during the course of therapy.

### Introduction

In radioiodine therapy of thyroid carcinoma, 100 mCi or more of  $^{131}\text{I}$  is administered to the patient orally. This presents a unique radiation protection problem in the hospital since the patient is both a radiation source and a source of environmental contamination. The problem of managing the patient as a source of radiation is one which is common to other forms of therapy where radioactive material is administered or implanted.<sup>1</sup> The magnitude of the contamination problem with  $^{131}\text{I}$  is not generally recognized. Most of the material is excreted in the urine and feces, but a few percent is excreted in saliva and perspiration.<sup>2</sup> During the initial hours after administration the saliva is especially rich in  $^{131}\text{I}$ . This leads to contamination of the air and items in contact with the patient and other items in the room. Previous studies have attempted to determine the resulting  $^{131}\text{I}$  burdens in medical and para-medical personnel caring for such patients,<sup>3,4</sup> and the dose to relatives exposed to outpatients treated with relatively small quantities for hyperthyroidism.<sup>5,6</sup> We have described preliminary results of a study in which patients were isolated for a few days following administration of material for carcinoma therapy, during which time, room air contamination was determined.<sup>7</sup> This paper is an extension of this latter study.

## Experimental Details

This experiment has been in progress since May 1970. Five patients were treated and followed in this study, and one received two treatments during this interval, and was thus studied twice. The administered doses were in the range of 100 - 200 mCi as shown in table 1. Each patient was located in a private room and isolation precautions were observed. The nurses were supplied with over-gowns and gloves which they wore each time they entered the patient's room. Routine room cleaning operations were suspended during the days in which the patients were isolated. Food was supplied on disposable dishes and with disposable utensils. Each day the bedding and towels, and the disposable dishes and utensils and remaining food items were sealed into plastic bags and labeled accordingly. Some areas of the floor and particularly the bathroom floor, were covered with heavy brown paper which remained in place until the end of the period of observation. Air was sampled by drawing room air through two tubes in series filled with silver nitrate solution, using a pump of approximately two litres per minute capacity. This trap was placed about 1 meter from the bedside. The patients were allowed to move about within the room and were not unduly distressed by these special precautions, which were continued until the quantity of material in the patient had fallen below approximately 30 mCi. The rooms were well ventilated and the air changed approximately once per hour.

After the patient had been discharged, the bags of potentially contaminated items were counted in the whole body facility at New York University Medical Center. This consists of a single 8 inch sodium iodide detector which was placed sufficiently far from each of the bags of contaminated items to give less than 20% variation in detection sensitivity over the volume of each bag.

The air sampler flow rate was determined with the full impedance of the silver nitrate traps present. The quantity of iodine trapped in the silver nitrate during each sampling interval was determined by counting in a standard well-type scintillation counter. Comparison of the quantity trapped in the first and second tube indicated that this trap could be considered essentially 100% efficient for this source of air-borne radioiodine.

## Results

The contamination collected on bedding, towels, dishes, utensils and other items is summarized in table 1. These values were all corrected for decay of the radioactive material between the time of collection and the time of measurement. The quantity of contamination found on bedding and utensils varied erratically during the several days of the therapy as shown. The average total of  $^{131}\text{I}$  contamination on bedding, dishes and utensils averaged over all patients and normalized to 150 mCi administered dose is shown in table 2 for successive days of treatment. The concentration of radioiodine in the air is summarized in table 3, for the same five patients. Air sampling was not performed during the first treatment of patient A. Average air concentrations (normalized to 150 mCi administered dose) are also shown in table 3.

Patient	Administered Activity (mCi)	Contamination on Bedding & Towels (uCi)				Contamination on Dishes & Utensils (uCi)				Contamination on Bathroom Floor (uCi)
		Day 1	2	3	4	1	2	3	4	
A1	200	370				20	180	-	-	-
B	200	31	25	25	3	66	27	-	-	12
C	200	127	37	39	396	570	105	110	490	-
A2	150	69	320	250	-	8	6	0.5	-	14
D	150	2	40	17	6	2	9	77	3	9
E	100	340	76	98	150	328				200

TABLE 1. Contamination measured on Bedding, Towels, Dishes, Utensils, and Bathroom Floor

Day	1	2	3	4	TOTAL 1-4
Bedding & Towels	140 (2-500)	700 (20-300)	90 (20-250)	130 (2-300)	400 (60-1000)
Dishes & Utensils	100 (2-400)	50 (6-150)	50 (.5-100)	180 (3-400)	300 (15-950)
TOTAL					700 (150-1500)

TABLE 2. Mean and (Range) of Contamination Measurements Normalized to 150 mCi Administered Activity

Patient	Concentration in Air (pCi/litre)		
	Day 1	Day 2	Day 3
B	59	-	-
C	26	124	77
A2	178	73	48
D	15	7	5
E	10	8	-
Normalized Mean	55	47	37

TABLE 3. Measured Concentration of <sup>131</sup>I in Air and Mean Concentration Normalized to 150 mCi Administered Activity

Items	Activity (uCi)
Nurses Gown	1
Thermometer	60
Telephone Mouthpiece	0.3
Nightgown	71
Water Jug	3.7

TABLE 4a. Contamination Measured on Representative Items

Items	Activity (uCi)
Paper Tissues	3400
Respirator	252

TABLE 4b. Exceptional Contamination on Individual Items

Contamination found on some individual items of special interest is listed in table 4. The figure of approximately 1 microcurie total contamination on the nurses isolation gowns was typical for all patients except where contamination by vomitus, stool or urine occurred. Following accidental contamination by stool, about 100 uCi was detected on an isolation gown. Thermometers, toothbrushes and containers for dentures were always contaminated. The telephone mouthpiece was always slightly contaminated.

### Discussion

The air burdens and the levels of contamination were related to the behavior of the patients and to their medical problems. For example, levels of contamination on bedding were clearly related to the radioactivity in perspiration, to the time each patient spent in bed, and to medical problems such as incontinence. In general, contamination was either due to contact with saliva, perspiration, urine, feces or vomitus, or to exposure to iodine in air. The latter was assumed to be due to saliva spray but could also involve oxidation and volatilization of iodine on contaminated items.

The pathways of iodine metabolism, the time interval after ingestion of the isotope and the thyroid functional status of the patient influence the excretion and dissemination of radioiodine. Early on, the material is largely in inorganic form and extra-thyroid in location. It is cleared from the blood efficiently by the salivary glands and kidneys leading to abundant contamination of saliva and urine and some radioactivity in sweat. After 24 hours radioiodine is mainly present in the form of thyroxine, a small proportion of which is excreted in stool. In hypothyroid individuals hormone synthesis is reduced and a relatively larger proportion of the  $^{131}\text{I}$  is excreted in the saliva and urine than in euthyroid or hyperthyroid patients. Furthermore, in the former patients the isotope persists in these biologic fluids for prolonged periods.

In general, iodine levels in air declined progressively after the day the radioiodine was administered (table 3) with the exception of patient C. This patient had copious salivation during periods of nausea. Since this patient had large metastases to the liver and subsequently developed granulocytopenia, the nausea and salivation were attributed to symptoms of mild radiation sickness. As a result of the excess salivation approximately 3mCi of  $^{131}\text{I}$  was deposited in disposable paper tissues during the first four days of therapy. The air burden also differed from the general case since the concentration increased during the first two days before declining. This is shown in detail in figure 1. Figure 1 also shows the pattern observed for patient A2 who was also associated with a high air burden.



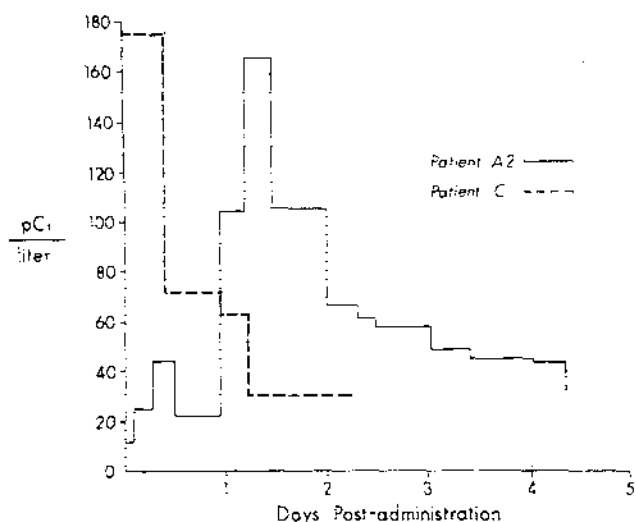


Figure 1. Concentration of  $^{131}\text{I}$  in Air After Administration of Radioiodine to Patients A2,C.

Total contamination on bedding, towels, dishes and utensils was greatest for patients C and E. The high value for patient C was due to exceptionally high contamination on dishes and utensils and this was consistent with the excess salivation problem. Patient E had an enlarged prostate and suffered from hesitancy in voiding and dribbling after completion of the urinary stream. Abnormally high levels of contamination on the bathroom floor and on towels and bedding were associated with these problems. One patient suffered a transient cerebral ischemic episode and required the use of a respirator, which was consequently contaminated as shown in table 4.

The measurements of air burdens have been used to estimate the potential burdens to other persons exposed to this source of radioactive material. Assuming that there is constant occupancy at the point of air sampling, that the concentration in air falls with a 1.5 day effective half life in the presence of the patient, the subject breathes  $2 \times 10^4$  litres of air per day, 23 percent of inhaled iodine enters the thyroid and decays with a 7.6 day effective half life,<sup>8</sup> and the initial concentration in air is in the range shown on day 1 in table 3, then the maximum thyroid burden is in the range 0.1 - 1  $\mu\text{Ci}$  and is reached in 4 days. This represents the situation which might be approached if two patients shared the same room or if relatives were exposed to an outpatient over long periods. The situation would presumably be worse if the room air were not changed by a central air conditioning system and were either static or recirculated by a single air conditioner. These potential burdens are reasonably consistent with burdens predicted for relatives of thyroid carcinoma patients by normalizing results obtained for hyperthyroid outpatients to doses used for carcinoma therapy.<sup>6</sup> Medical and paramedical personnel are only exposed to air contamination for a relatively short time if the situation is well controlled and nursing duties are rotated. Such a person exposed to the levels observed on day 1 for a period of 10 minutes would potentially accumulate 0.5 - 8  $\text{nCi}$ . This is reasonably consistent with

thyroid burdens in the range of 0.1 - 1.8 nCi reported for medical and paramedical personnel from this institution following administration of 100 mCi doses of radioiodine to patients for thyroid carcinoma.<sup>3</sup> Thyroid burdens in paramedical personnel of approximately 0.1 uCi reported for other institutions<sup>4</sup> are consistent with our measurements of air burdens only if long periods of exposure are assumed, or if the rate of air change is less than in this institution.

### Conclusions

The levels of contamination following radioiodine therapy for thyroid carcinoma are much larger than those normally tolerated in an open laboratory. Air burdens exceeded the MPCa values for controlled areas (9 pCi/litre, 40 hour week and 3 pCi/litre, 168 hour week<sup>8</sup>). However, thyroid measurements on medical and paramedical personnel and predictions based on our observations of air burdens indicate that such personnel will not accumulate excessive thyroid burdens if the patient management is well controlled. The problem of managing these patients is discussed in NCRP 37<sup>1</sup>, where it is stated that patients may contaminate dishes, utensils and bedding, whereas we have found this to be invariably the case. The problem of air contamination is not discussed in the report. We consider that it is important to recognize that the contamination problem continues to be significant for 3 or 4 days following administration of radioiodine and that significant air burdens are present during this time. Radioiodine therapy is of proven value and importance but we consider that patients should generally be hospitalized for 3 or 4 days and should be managed with attention to the contamination and air burden problems. The considerations for radiation safety are closely related to the nature of the patient's medical problems, the pathways of iodine metabolism and the routes of iodine excretion.

### References

1. NCRP Report 37, Washington, D.C. (1970)
2. Silver, S., Radioactive Nuclides in Medicine and Biology. Lea & Febiger, Philadelphia (1968)
3. Blum, M., Liuzzi, A., Thyroid <sup>131</sup>I Burdens in Medical and Paramedical personnel. JAMA, 200, 992-994 (1967).
4. Sear, R., Radiation Hazards Resulting from Clinical Use of Radio-Iodine. Acta. radiol. (Phys., Biol., Ther.) 2, 263-272 (1964)
5. Buchan, R.C.T., Brindie, J.M., Radioiodine Therapy to Out-Patients - the Contamination Hazard. Brit. J. Radiol. 43, 479-482 (1970)
6. Chandra, R., Marshall, C.H., Radioiodine Therapy to Out-Patients The Contamination Hazard. Brit. J. Radiol. 44, 557 (1971)
7. Blum, M., Chandra, R., Marshall, C.H., Environmental Contamination with Iodine <sup>131</sup> Related to Treatment of Hyperthyroidism and Carcinoma of the Thyroid Gland. IEEE Transactions on Nuclear Science, NS-18 No. 1, 57-59 (1971).
8. I.C.R.P. Publication 2. Pergamon Press, New York (1959).

A COMPREHENSIVE APPROACH FOR THE EVALUATION OF COMPARATIVE  
DOSIMETRY OF INTERNALLY ADMINISTERED RADIOPHARMACEUTICALS

Parvathi Hosain and Fazle Hosain  
Department of Radiological Science  
The Johns Hopkins Medical Institutions  
Baltimore, Maryland, USA

Abstract

Recent innovations both in instrumentation and radiopharmaceuticals are helping nuclear medicine to develop as a discipline in medical practice primarily for diagnosis with an emphasis on scanning. Radiation safety of a radiopharmaceutical is based on the assessment of the radiation exposure to the critical organ from a tracer dose. It is also important to consider relative tracer doses necessary for an optimal result for selecting one of the several radiopharmaceuticals available for a similar investigation. A comprehensive formulation of an index has been attempted for the relative dosimetry integrating mainly (i) conventional method of dose calculation, (ii) physical properties of radionuclide, (iii) available nuclear instrumentation, (iv) metabolic fate of radiopharmaceutical, and (v) gross considerations for radio-sensitivity of organs and dose rate. This approach has been illustrated considering the situation of brain scanning using Hg-197 chlormerodrin, Tc-99m pertechnetate, Tc-99m DTPA, short-lived In-113m DTPA and long-lived Yb-169 DTPA.

Introduction

Nuclear medicine is appearing as a discipline in medical practice which involves primarily the use of radiopharmaceuticals for diagnosis. Recently scintigraph has attained tremendous importance due to availability of advanced nuclear instruments and short-lived radiopharmaceuticals. Agents with high photon yield with low radiation dose are considered most desirable for clinical use.

Large number of radiopharmaceuticals are being developed in order to scan various organs and diseased conditions, and to improve the existing methods. Acceptance of any new product for clinical use depends on its cost, efficacy, specificity, toxicity and dosimetry. Although radiation exposure is an important consideration, it is difficult to control the medical applications by any formal method due to the fact that the primary interest lies in the immediate benefit to the patient.

The present exposition has been aimed to provide a guideline for the evaluation of relative dosimetry while choosing one of the several similar agents.

### Comparative Dosimetry

Conventional method of dose calculation has been improved and standardized greatly<sup>1</sup>. It is based on the utilization of physical properties of radionuclides, absorbed fraction of the radiation, and the metabolic fate of the radiopharmaceutical. It provides a practical value for the concept of safety with a recommended tracer dose for a clinical study. It could be expressed by

$$D \text{ (rads/mCi)} = 1000 \text{ c/m} \cdot 1.44 \cdot T_e \cdot \Sigma \Delta \phi = R \cdot A \quad \dots\dots\dots(1)$$

where, R is the maximum dose rate (rads/mCi/hr) to an organ of mass  $\pi$  gm ( $=1000 \Sigma \Delta \phi / \pi$ ), and A is the area under time-concentration curve (approximately equal to  $1.44 \text{ c} \cdot T_e$  where c is the maximum fractional concentration and  $T_e$  is the effective half-life in hours).

Comparative dosimetry becomes important when several similar agents are available for a particular investigation. Usually the tracer dose is adjusted to keep the radiation dose within a safe limit. However, an appropriate approach should be to find out the relative amounts of tracer doses that would give similar measurable count rate at the time of study. Occasionally this method has been adopted<sup>2</sup>. The relative tracer dose could be expressed by

$$T \text{ (mCi)} = 1/(f \cdot e \cdot d) \quad \dots\dots\dots(2)$$

where, f is the fractional yield of the photons used for detection, e is the detection efficiency of the instrument or at least the photo-peak interaction coefficient, and d is the activity remaining after decay or elimination by the mean time of the study. It is assumed that the target to non-target ratio for the different agents remain approximately the same.

The variations in radiosensitivity of critical and other organs have always raised questions in mind, but it is difficult to assign any quantitative value. However, it is well known that bone marrow is most radiosensitive. It would be reasonable to ascribe an empirical sensitivity factor ( $S_0$ ) of 3, 2, and 1 respectively for bone marrow (and gonads during reproductive age), gastrointestinal, and the rest of the organs. Further, it is also desirable to incorporate a sensitivity factor for increased dose rate ( $S_r$ ). Empirically,  $D/R (= 1.44 T_e) = 10^{n+1}$  may be used for a normalization of dose-rate sensitivity: assuming  $n=0$  for normal condition ( $S_r = 1$ ), one could arrive factors like  $S_r = 1/(n+1)$  or  $(n+1)$  for  $-n$  or  $+n$  values, the intermediate values could be obtained graphically. As an example,  $S_r = 2$  if 10 rads are delivered at the rate of 10 rads/hr instead of 1 rad/hr.

$$\text{Then, } S = S_0 \cdot S_r \quad \dots\dots\dots(3)$$

In summary, the normalized radiation dose (NRD) for different radiopharmaceuticals used for similar investigations can be expressed by

$$\text{NRD} = R \cdot A \cdot T \cdot S \quad \dots\dots\dots(4)$$

It is assumed that the radiopharmaceuticals are pure, otherwise the contributions of any radionuclide impurity and any radiochemical impurity should be taken into consideration.

### Example of Brain Scanning

Mercury-197 labeled chlormerodrin has been and is being used for brain scanning<sup>3</sup>, although technetium-99m pertechnetate has become the most preferred agent<sup>4</sup>. Chelates (DTPA) labeled with Tc-99m<sup>5</sup>, In-113m<sup>6</sup> and Yb-169<sup>7</sup> can be used for brain scanning. Kidneys, upper large intestine and bladder could be taken as the critical organs for chlormerodrin, pertechnetate and chelates, respectively.

The radiation doses for the different agents were calculated by taking values of nuclear parameters (except for ytterbium-169<sup>9</sup>) and absorbed fractions of energy for different organs from MIRD pamphlets<sup>1</sup>. Biological factors (such as, concentrations in organs, biological half-lives, time of study after administration of tracer dose, and usual tracer doses) were assumed. However, these assumptions were based on various publications (such as the summary in a text book<sup>9</sup>). Estimation of relative tracer dose was based on the useful photon yield, photo-peak interaction coefficient of useful photon energy in 2-inch NaI crystal (thickness), and the fractional activity remaining after effective loss by the mean time of study. Relative tracer doses were then normalized to 10 mCi of Tc-99m pertechnetate. Further, it was assumed that a 100% of the dose was initially uniformly distributed in the total body and remained uniform although the fractional concentrations in different organs were different. The empirical sensitivity factor for dose rate was obtained using a semi-log plot of  $10^{T_{1/2}+1}$  (for 1.44 T<sub>1/2</sub> values) against  $\mu_n$ .

Table 1 and Table 2 represent the basic physical and biological data. Table 3 shows the aspects of dosimetry for the total body. Table 4 summarizes the results for the critical organs.

Table 1: Basic physical data for the radiopharmaceutical

Labeling nuclide	Chemical agent	Physical half-life (hr)	Useful photon energy (keV)	Useful photon yield (%)	Usual tracer dose (mCi)
Hg-197	Chlormerodrin	65.0	67-81	90.8	0.75
Tc-99m	Pertechnetate	6.0	140.5	88.3	10.0
Tc-99m	Chelate (DTPA)	6.0	140.5	88.3	10.0
In-113m	Chelate (DTPA)	1.67	393.0	65.4	15.0
Yb-169	Chelate (DTPA)	763.2	177 & 198.0	55.3	10.0

Table 2: Basic biological data for the radiopharmaceutical

Agent	Photo-peak interaction coefficient (2-in NaI)	Biological half-life (hr)	Time gap of study (hr)	Relative tracer dose (mCi)	$\Sigma\Delta\phi$ for total body
Hg-197 Chlor.	1.00	6	3.0	13.6	0.225
Tc-99m Pert.	0.98	48	1.0	10.0	0.130
Tc-99m DTPA	0.98	2	0.5	11.6	0.130
In-113m DTPA	0.77	2	0.5	24.7	0.474
Yb-169 DTPA	0.97	2	0.5	17.2	0.354

Table 3: Calculation of dosimetry for the total body

Agent	Dose rate rads/mCi/hr	Area under conc.-time curve (1.44 cT <sub>e</sub> )	Radiation dose (rads/mCi)	Rads per usual tracer dose	Normalized rads/test
Hg-197 Chlor.	0.0032	7.91	0.0253	0.018	0.38
Tc-99m Pert.	0.0019	7.68	0.0146	0.146	0.16
Tc-99m DTPA	0.0019	2.16	0.0041	0.041	0.08
In-113m DTPA	0.0068	1.31	0.0089	0.089	0.41
Yb-169 DTPA	0.0079	2.87	0.0227	0.227	0.60

Table 4: Calculation of dosimetry for the critical organ

Agent	Concerned organ	Fractional conc. (c)	Biological half-life (hr)	Rads per usual tracer dose	Normalized rads/test
Hg-197 Chlor.	Kidneys	0.2	1704	8.2	75.9
Tc-99m Pert.	UL intest.	0.1	12	0.8	2.0
Tc-99m DTPA	Bladder	0.5	2	1.5	2.8
In-113m DTPA	Bladder	0.5	2	4.5	20.8
Yb-169 DTPA	Bladder	0.5	2	10.6	28.1

#### Discussion

Radiation dose rate (R) can be calculated with a high degree of accuracy except when the mass of the concerned organ is variable or uncertain (such as bladder with urine<sup>10</sup>). Considerable uncertainty could be inherent with 'A' due to difficulties in obtaining distribution patterns of the radiopharmaceuticals in human organs under normal and diseased conditions. Relative tracer doses could be calculated with sufficient accuracy if instrumental sensitivity for different agents (radionuclides) are determined experimentally. Quantitative assessment for radiosensitivity would remain a radiobiological problem particularly with reference to dose rate. However, it does not seem very unreasonable, at present, if a factor of 2 is used to increase the index of radiation dose when 10 rads are delivered at the rate of 10 rads/hr instead of 1 rad/hr.

Reduction of radiation doses in diagnostic uses of radionuclides has remained an important consideration from the point of view of exposure to patient and population. In earlier days, usually low level counting techniques have been considered to reduce the tracer dose<sup>11</sup>. In recent days, it appears that the diagnostic values are being enhanced by quantitation of scans with computers in studies with multi-millicuries of short-lived radiopharmaceuticals<sup>12</sup>. In practice, one has to compromise to certain extent the radiation dose with the specificity, efficacy or the cost of the radiopharmaceutical. In the growing phase of the development of radiopharmaceuticals, it is hoped that the present consideration would help in the selection of the agent to reduce the radiation dose in an investigation under optimal condition.

### Acknowledgment

The authors wish to thank Prof. Henry N. Wagner, Jr., and the United States Public Health Service (for partial support with GM-10548).

### References

1. MIRD Pamphlets: J. Nucl. Med. Suppl. No. 1-3, 1968 & 1969.
2. Hosain, F., Hosain, F., Iqbal, Q.M., Carulli, N., and Wagner, H.N., Jr.: Measurement of plasma volume using  $^{99m}\text{Tc}$  and  $^{113m}\text{In}$  labeled proteins. Br. J. Radiol. 42: 627, 1969.
3. Wagner H.N., Jr.: Principles of Nuclear Medicine. W.B. Saunders Co. 1968, p. 663.
4. Blahd, W.H.: Nuclear Medicine. McGraw-Hill Co., 1971, p. 243.
5. Hauser, W., Atkins, H.L., Nelson, K.S., and Richards, P.: Technetium-99m DTPA: A new radiopharmaceutical for brain and kidney scanning. Radiology 94: 679, 1970.
6. Clements, J.P., Wagner, H.N., Jr., Stern, H.S., and Goodwin, D.A.: Indium  $^{113m}$  diethyltriaminopentacetic acid (DTPA): A new radiopharmaceutical for brain scanning. Am. J. Roentgenol. 104: 139, 1968.
7. Gilday, D.L., Reba, R.C., Hosain, F., Longo, R., and Wagner, H.N., Jr.: Evaluation of ytterbium-169 diethylenetriaminopentacetic acid as a brain-scanning agent. Radiology 93: 1129, 1969.
8. Syed, I.B., and Hosain, F.: The basic dosimetry for ytterbium-169 chelate. (To be published).
9. Powsner, E.R., and Raeside, D.E.: Diagnostic Nuclear Medicine. Grune and Stratton, Inc., 1971, pp. 184-185.
10. Hosain, F., Syed, I.B., and Hosain, F.: Radiation dose to bladder from radioactive glomerular agents (abst.). Phys. Biol. Med. 17: 866, 1972.
11. Hosain, F.: Reduction of doses in diagnostic uses of radioisotopes. Ind. J. Med. Res. 48: 250, 1960.
12. Wagner, H.N., Jr., and Katarajan, T.K.: Computers in nuclear medicine. Hospital Practice 7: 121, 1972.

REDUCTION OF RADIATION EXPOSURE TO NUCLEAR MEDICINE PERSONNEL  
BY THE USE OF NEW "INSTANT" TECHNIQUES FOR PREPARATION  
OF TECHNETIUM RADIOPHARMACEUTICALS

Philip J. Robbins\*, Byron M. Branson and Richard J. Grant\*\*  
\*Nuclear Medicine Laboratory, Bureau of Radiological Health, FDA  
and \*\*Radioisotope Laboratory  
at University of Cincinnati Medical Center  
Cincinnati, Ohio 45229

Abstract

Radiation exposure to the hands from the preparation of  $^{99m}\text{Tc-Sn-MAA}$  from an instant kit and  $^{99m}\text{Tc-HAM}$  from a non-instant kit has been measured by thermoluminescent dosimetry (TLD). The exposure to the fingers from the preparation of 100 mCi of  $^{99m}\text{Tc-HAM}$  exceeded the exposure from preparation of the same amount of  $^{99m}\text{Tc-Sn-MAA}$ . An exposure of 153 mR/100 mCi of prepared radiopharmaceutical to the left middle finger was the highest value recorded in the study.

Introduction

Use of short half-lived radiopharmaceuticals such as  $^{99m}\text{Tc}$  and  $^{113m}\text{In}$  compounds has greatly reduced patient exposure in nuclear medicine while requests for diagnostic procedures using these radionuclides have increased. Because of the short half-lives of  $^{99m}\text{Tc}$  and  $^{113m}\text{In}$ , these radiopharmaceuticals must be prepared on the day of use. Pre-tested vials of sterile, pyrogen-free reagents (kits) have streamlined the preparation of various radiopharmaceuticals but personnel contact with the radionuclide extends 10 to 90 min beyond the normal time for generator elution and transfer of patient dose to a syringe.

This study was designed to measure the radiation exposure to experienced nuclear medicine personnel preparing the lung scanning agents, technetium-99m macroaggregated albumin ( $^{99m}\text{Tc-Sn-MAA}$ ) using an instant kit and technetium-99m human albumin microspheres ( $^{99m}\text{Tc-HAM}$ ) using a conventional kit. An instant kit is one which requires a minimum number of manipulations and can be prepared quickly. Our kit for  $^{99m}\text{Tc-Sn-MAA}$  meets these requirements. Technetium-99m human albumin microspheres is prepared from a conventional kit requiring many manipulations and 25 min elapsed time. The methods of preparing  $^{99m}\text{Tc-Sn-MAA}$  and  $^{99m}\text{Tc-HAM}$  will be described to illustrate the various steps in which unnecessary exposure to personnel may occur.

Table 1 lists the various  $^{99m}\text{Tc}$  radiopharmaceuticals prepared in nuclear medicine laboratories and the typical maximum quantities of  $^{99m}\text{Tc}$  used. The exposure time is the period during which personnel handle the radionuclide directly or remotely on a daily basis.

If one person prepared all the radiopharmaceuticals without shielding, on a typical day he could be exposed to 200-300 mCi of



$^{99m}\text{Tc}$  for 30-40 min. This, in fact, is not the case since an L-block shield<sup>1</sup> and lead generator eluate shields (cylindrical shields)<sup>2</sup> are used to minimize the exposure. Handling the vials restricts the exposure to the fingertips, wrists, and forearms.

Table 1 Quantity of  $^{99m}\text{Tc}$  used and daily exposure times for typical radiopharmaceuticals

	Quantity (mCi)	Exposure Time (min)
$^{99m}\text{Tc}$ -sulfur colloid	50	5
$^{99m}\text{Tc}$ -human serum albumin	30	1
$^{99m}\text{Tc}$ -macroaggregated albumin ( $^{99m}\text{Tc}$ -MAA)	120	10
$^{99m}\text{Tc}$ -macroaggregated albumin ( $^{99m}\text{Tc}$ -Sn-MAA)	120	2
$^{99m}\text{Tc}$ -DTPA	15	1
$^{99m}\text{Tc}$ -diphosphonates	70	1
$^{99m}\text{Tc}$ -human albumin microspheres ( $^{99m}\text{Tc}$ -HAM)	140	14

It can be seen from Table 1 that the exposure from  $^{99m}\text{Tc}$ -MAA or  $^{99m}\text{Tc}$ -HAM would exceed the exposure from other radiopharmaceuticals if only the quantity and the exposure time are considered. The technique for preparation of  $^{99m}\text{Tc}$ -Sn-MAA, developed by this laboratory<sup>3</sup> requires less than 5 min elapsed time, and involves an exposure time of only 2 min. It was thought that this shortened exposure time would be of advantage in terms of personnel effort and radiation exposure.

#### Methods and Materials

The modification of the method of Robbins et al.<sup>3</sup> outlined in Table 2 was used for preparing  $^{99m}\text{Tc}$ -Sn-MAA. Fifty to one hundred twenty mCi of  $^{99m}\text{Tc}$  were handled in its preparation.

Table 2 Preparation of  $^{99m}\text{Tc}$ -Sn-MAA

1. Add 1 ml tin stock solution to a vial containing albumin macroaggregates
2. Swirl
3. Add  $^{99m}\text{TcO}_4$  ( 5 ml)
4. Swirl
5. Centrifuge and withdraw supernatant solution
6. Reconstitute with 6 ml saline
7. Assay in dose calibrator

Technetium- $^{99m}$  human albumin microspheres were prepared according to the method described in the 3M Company package insert using the

3M brand  $^{99m}\text{Tc}$ -HAM kit.<sup>4</sup> An outline of this procedure is given in Table 3. Fifty to one hundred forty mCi of  $^{99m}\text{Tc}$  were handled in this procedure.

Table 3 Preparation of  $^{99m}\text{Tc}$ -HAM

---

To the labeling vial, containing microspheres and a thiosulfate tablet:

1. Add  $^{99m}\text{TcO}_4^-$  (10 ml)
  2. Sonicate 2 min
  3. Agitate in boiling water 6 min
  4. Cool in water bath 1 min
  5. Withdraw all liquid from labeling vial
  6. Add 10 ml saline suspending solution
  7. Sonicate 1 min
  8. Withdraw liquid from labeling vial
  9. Add 10 ml saline suspending solution
  10. Assay in dose calibrator
- 

The 3M brand albumin microsphere labeling system, composed of the labeling and suspending unit, and rinsing unit and a shield with lead-glass window for the tagging vial were also employed throughout the  $^{99m}\text{Tc}$ -HAM preparation.

Exposure data were obtained from duplicate sets of TLD's placed on the distal medial aspect of the thumb, the distal lateral aspect of the middle finger, the distal medial aspect of the ring finger, and the anterior wrist surface of each hand. The TLD's were worn during five or more preparations of each radiopharmaceutical so that the exposure was accumulated while handling more than 500 mCi of  $^{99m}\text{Tc}$ . An average exposure was calculated from the two measurements at each point. The exposure is expressed as mR per 100 mCi prepared. LiF chips consistent to within  $\pm 5\%$  were used in these studies (TLD 100's,  $1/8 \times 1/8 \times 0.035$  in). Six TLD's, arranged two each at 3, 5 and 8 cm from the central point source, were exposed to 17 mCi  $^{99m}\text{Tc}$  for 20 min for use as standards. The TLD's were readout in a TLR-5 Eberline Reader using standard techniques.

### Results

Table 4 contains the results of measuring hand exposure during preparation of  $^{99m}\text{Tc}$ -Sn-MAA and  $^{99m}\text{Tc}$ -HAM. While handling a total of 523 mCi of  $^{99m}\text{Tc}$  for nine batches of  $^{99m}\text{Tc}$ -Sn-MAA, the total quantity prepared was 497 mCi with an average tagging yield of 95%. For  $^{99m}\text{Tc}$ -HAM, a total of 518 mCi of  $^{99m}\text{Tc}$  handled in six runs provided 362 mCi with an average tagging yield of 70%. The results in Table 4 show that  $^{99m}\text{Tc}$ -HAM contributes more radiation exposure to the fingers than does the  $^{99m}\text{Tc}$ -Sn-MAA. The unusually high exposures to various fingers are indicative of the preparation techniques. In both procedures transferral of the  $^{99m}\text{TcO}_4^-$  from its storage vial to the syringe for addition to the reaction vial is considered the cause for high exposures. Due to

the technique in performing this step, the exposure to the left ring finger (for  $^{99m}\text{Tc-Sn-MAA}$ ) was higher than the other fingers. This technique is also considered one of the causes of high exposure to the left thumb (for  $^{99m}\text{Tc-HAM}$ ). Use of a screw-cap cylindrical shield, which exposes only the rubber septum of the pharmaceutical vial, is thus recommended for each of these procedures.

Table 4 Average exposure to the hands during preparation of  $^{99m}\text{Tc-Sn-MAA}$  and  $^{99m}\text{Tc-HAM}$

	mR/100 mCi prepared	
	$^{99m}\text{Tc-Sn-MAA}$	$^{99m}\text{Tc-HAM}$
<u>Right Hand</u>		
Wrist	9	7
Thumb	47	86
Middle Finger	55	109
Ring Finger	45	46
<u>Left Hand</u>		
Wrist	10	19
Thumb	53	151
Middle Finger	60	153
Ring Finger	94	76

In preparing  $^{99m}\text{Tc-Sn-MAA}$ , the left ring finger received additional exposure when the reaction vial was held in the left hand to withdraw the supernatant solution (Step 5, Table 2). Holding the vial with tongs in the left hand is recommended for this step of the procedure. The left thumb and middle finger received added exposure from the microspheres in Step 4, Table 3 wherein the tagging vial was wiped with a paper towel held in the left hand. This step is necessary to prevent rusting of the interior of the special shield for the tagging vial. Use of remote wiping technique is also recommended in this step. Monitoring of the exposure to the face, chest and body trunk from both of these procedures was less than detectable by the TLD techniques employed (<10 mR total cumulated exposure).

#### Conclusions

The exposure from instant kits is less than with other methods because the simplicity of the instant procedures permits shorter exposure time and less contact with the pharmaceutical vial. The use of instant kits is to be encouraged.

Furthermore, while the radiation exposure to the fingers from preparation of  $^{99m}\text{Tc-HAM}$  is greater than that received from  $^{99m}\text{Tc-Sn-MAA}$ , the exposure level from both procedures could be reduced by adherence to procedural recommendations in the discussion. Additionally, all procedures should periodically be evaluated in terms of exposure, especially to the hands. The techniques monitored in this study are just two from the various  $^{99m}\text{Tc}$  radiopharmaceuticals prepared daily by most nuclear medicine laboratories. Based on the highest exposure recorded in this study (153 mR/100 mCi to the left middle finger), a person

preparing 200 batches of 80 mCi each of  $^{99m}\text{Tc}$ -HAM during the year would receive about 24 R/year to the hand. While this is within the recommended limits of NCRP (75 rems/year to the hands)<sup>5</sup>, it represents the exposure from  $^{99m}\text{Tc}$ -HAM preparation only. The use of instant kits, such as  $^{99m}\text{Tc}$ -Sn-MAA and  $^{99m}\text{Tc}$ -DTPA<sup>6</sup> along with an adequate knowledge of the procedural steps will contribute to a lower total exposure from all phases of nuclear medicine.

#### Acknowledgments

The authors appreciate the assistance of Mr. Douglas Weitzel, Chief Technician of the Nuclear Medicine Laboratory, Bethesda Hospital, Cincinnati, Ohio in obtaining the radiation exposure values for  $^{99m}\text{Tc}$ -HAM.

#### References

1. Barnett, H.D. and Harris, C.C.: "A Means of Reducing Radiation Exposure from Technetium Generators," J. Nucl. Med., 11, 180-181 (1970).
2. Branson, B.M., Hoops, R.G. and Grant, R.J.: "Reducing Personnel Exposure in Nuclear Medicine," J. Nucl. Med., 14, 382 (1973).
3. Robbins, P.J., Fortman, D.L. and Lewis, J.T.: "A New Lung Scanning Agent:  $^{99m}\text{Tc}$ (Sn)MAA," Int. J. Appl. Radiat. Isotop., 24, 481 (1973).
4. 3M Company, Nuclear Products, "Labeling instructions for 3M brand albumin microsphere -  $^{99m}\text{Tc}$  labeling kit," Nuclear Products Division, 3M Company, St. Paul, Minnesota.
5. National Council on Radiation Protection and Measurements, "Basic Radiation Protection Criteria," (NCRP Report No. 39) National Council on Radiation Protection and Measurements, Washington, D.C., 1971.
6. Eckleman, W. and Richards, P.: "Instant DTPA," J. Nucl. Med., 11, 761 (1970).

## REDUCING PERSONNEL EXPOSURE IN NUCLEAR MEDICINE

Byron M. Branson\*, Richard G. Hoops\*, and Richard J. Grant\*\*

\*Nuclear Medicine Laboratory, Bureau of Radiological Health, FDA  
and \*\*Radioisotope Laboratory, University of Cincinnati College of Medicine  
Cincinnati, Ohio 45229

### ABSTRACT

Nuclear medicine procedures are growing at a rate greater than 15%/yr and the radiopharmaceutical industry over 25%/yr. A nuclear medicine facility in a medium-large hospital routinely handles 300-700 mCi of  $^{99m}\text{Tc}$ /day plus prepares  $^{99m}\text{Tc}$  radiopharmaceuticals and handles millicurie amounts of other isotopes. As this specialty grows, so must exposure reduction techniques. Periodic assessment of personnel handling the molybdenum-technetium generators and  $^{129}\text{Cs}$  were made using TLD's. These data are reviewed with comments about potentials for personnel exposures and exposure reducing methods. Additional shielding and new handling techniques have been incorporated and are discussed.

### INTRODUCTION AND BACKGROUND

The Nuclear Medicine Laboratory is located and operated in collaboration with the Radioisotope Laboratory. In these laboratory facilities, the following contribute primarily to personnel radiation exposure:

1. daily handling of 500 mCi to 1.4 curies of  $^{99m}\text{Tc}$ ,
2. weekly processing of mCi amounts of  $^{129}\text{Cs}$ ,
3. preparing throughout the week a variety of  $^{99m}\text{Tc}$  labeled radiopharmaceuticals from kits, and
4. weekly handling of mCi amounts of  $^{131}\text{I}$ ,  $^{133}\text{Xe}$  and  $^{113m}\text{In}$ .

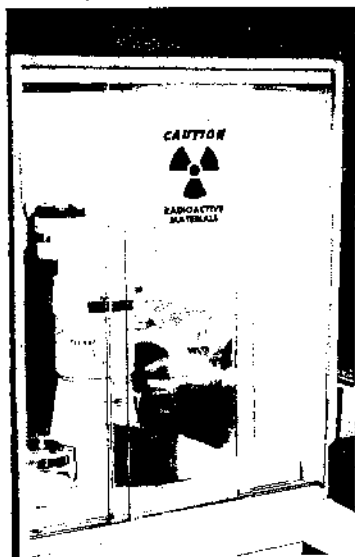
Each of these can increase the daily exposure to laboratory personnel and result in exposure levels that exceed what we would have predicted just a few years ago.

Max Lombardi<sup>1</sup> reported on a 12 month survey of 69 hospitals that nuclear medicine procedures grew at a rate of 16% per year and that the radiopharmaceutical industry grew 25-26% in a similar 12 month period. Accompanying this growth in radiopharmaceutical usage and new nuclear medicine laboratories in many hospitals, the problem of personnel protection has been recognized by some but unrecognized by many.

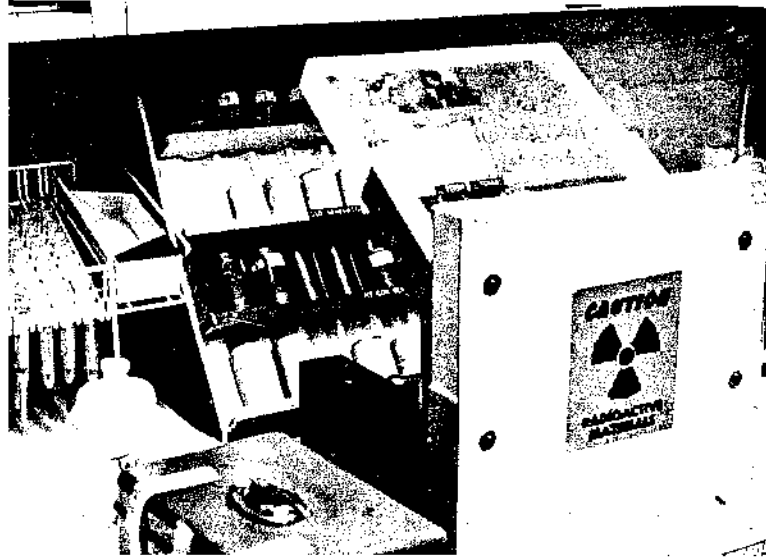
The National Council on Radiation Protection and Measurements (NCRP)<sup>2</sup> recommends 75 rems/year, 25 rems/qtr., as the permissible dose equivalent to the hands. They have further characterized this as an "interim concession" in "comment" to this section and have also indicated that "all reasonable efforts should be made to keep exposure of the hands and forearms within the general limit for skin, 15 rems/year".

A study 4 years ago by Neil<sup>3</sup> on the radiation exposure to the hands from handling  $^{99m}\text{Tc}$  showed a maximum dose equivalent of 10 rems/curie/minute for the index finger and thumb with lesser dose equivalents for other parts of the hands. Using Neil's data, a physician that gives 400 injections/year of 10 mCi each and in only 30 seconds handling time would receive 30 rems for that year for that portion of his hand alone, if handling of any other radioisotopes is neglected. ( $^{131}\text{I}$ ,  $^{133}\text{Xe}$ ,  $^{18}\text{F}$ ,  $^{129}\text{Cs}$ , etc.)

The need for additional personnel protection for all radioisotopes in the laboratory resulted in a complete rearrangement of our "hot" lab area. Lead-lined housings with sliding lead glass doors were specially built\* for two molybdenum-technetium generators, figure 1. The two end doors must be moved to the middle where there is a third fixed plate of lead glass. These form a body shield while the hands may be inserted through the openings on either side of the lead glass. We have also built  $^{99m}\text{Tc}$  eluate organizer racks, figure 2, that provide  $\frac{1}{2}$ " lead shields for each eluate bottle. Each shield has a lid with a hole for the head of the eluate bottle. The bottles are placed on a slant pointed away from the operator. Each tier of eluate shields is color coded to identify the eluate by the parent generator. Added positions are available for  $^{99m}\text{Tc}$  radiopharmaceuticals, prepared from kits. An additional tier permits storage of other radiopharmaceuticals in use that day. All the  $^{99m}\text{Tc}$  and most radioiodine doses are prepared behind the face and body shield<sup>4</sup> seen just in front of the organizer rack.



**Figure 1.** Mo-Tc house built of plywood, Pb lined epoxy painted, fluorescent light and 3 Pb glass doors. Base made as a tray to contain spills.



**Figure 2.** Eluate organizer rack for  $^{99m}\text{Tc}$  placed immediately behind a face and body shield. Most radiopharmaceutical doses are prepared at this work station.

Most of the diagnostic administrations of radioisotopes in nuclear medicine are given at some location other than the hot laboratory. For injections, a transporting tray, with a lead syringe holder anchored to the side of the tray is used and is shown in figure 3. In addition, the tray is equipped with other items the physician will need for injection. This special tray has been helpful in reducing exposure and contamination and has proven very easy and convenient to laboratory personnel.

We conservatively estimated that the generator housing reduces major body exposure 2 times during the elution procedure. The eluate shields probably reduce personnel exposure by a factor of 5, and the face and body shield reduces the technicians total body exposure an estimated factor of 5. The personnel exposure reduction realized from the dose tray is difficult to calculate, but it is our conservative guess that the hand exposure is reduced a factor of 5 to 15 depending on the distance the dose syringe must be carried, etc.

\* These were designed for one specific company's generator and might require modification for a different brand generator.

In order to assess the personnel exposures more specifically, we have studied some hand exposures during routine handling procedures using thermoluminescent dosimeters (TLD's). For  $^{99m}\text{Tc}$  and  $^{129}\text{Cs}$  the procedure used was to place TLD\* dosimeters in the anterior and posterior positions on one or two fingers, the thumb, and wrist of each hand. Additionally, TLD's were placed on the forehead or glasses frame, chest and gonadal area of the body. Unless otherwise stated the TLD's on the fingers were in the finger ring position. We assumed that all radiation absorbed by the lithium fluoride chip was from either  $^{99m}\text{Tc}$  or  $^{129}\text{Cs}$ . This actually is not the case; for instance, the aluminum holder for the target material in the cesium production was emitting high energy gamma rays from  $^{22}\text{Na}$  and  $^{24}\text{Na}$  and likewise for  $^{99m}\text{Tc}$  the 740 and 780 keV  $^{99}\text{Mo}$  radiation is emitted through the generator shielding. For each study two lithium fluoride chips were placed at each dosimetry point; each was measured separately and the count data averaged.

At the time of this study, two molybdenum technetium generators were received each week; each rated at 400 mCi 5 days after receipt. One technician had the responsibility for removing the old generator, installing the new one, and obtaining the daily elutions required (about 5 curies/week). For this study, the TLD's were worn only during these procedures. The resultant TLD data, Table I, reflects the expected higher exposure on the anterior surface of each hand and higher exposure to the right hand. Data from the forehead, chest and gonadal area of the body were all under 10 mR/week. The highest finger exposures averaged 131 mR/week which over 50 weeks would amount to 6.5 R maximum cumulative exposure. In some other laboratory with only one 400 mCi generator and using only the shielding provided with the generator, the annual exposure could be as much as 20 R for the same person performing this task.

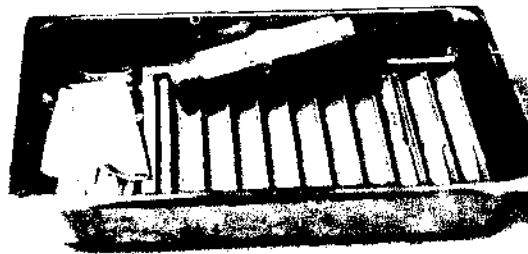
Table I. TLD PERSONNEL DATA FOR  $^{99m}\text{Tc}$  IN mR  
- Handling Generators Only -

		Weekly Average	
Fingers - Anterior Surface		61	109
		36	94
Thumb - Anterior Surface	R	46	198
	L	54	124
Wrist - Anterior Surface	R	19	33
	L	10	19
Totals - Fingers		49	131
- Hands		37	109

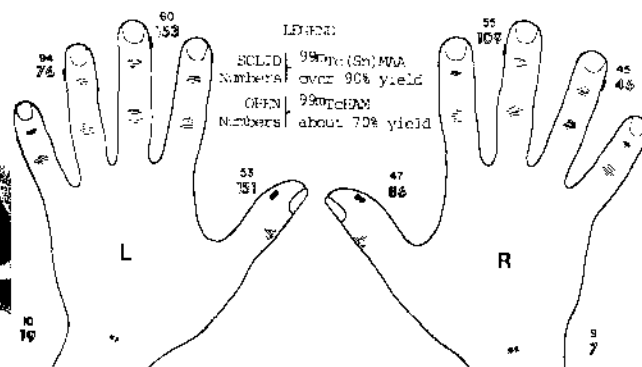
50 Week Total - Fingers (131 x 50) = 6550 mR

A more recent study has compared the preparation of  $^{99m}\text{Tc}$  macroaggregated albumin prepared according to the method of Robbins<sup>5</sup> and  $^{99m}\text{Tc}$  human albumin microspheres labeled according to the method described by the manufacturer (3M Company) using their equipment. For this latter study TLD's were placed as shown in figure 4. The TLD's were on the lateral and medial borders of the finger tips so as not to interfere with operator's finger tip sensitivities. The dosimeters were worn during five or more preparations of each radiopharmaceutical, each study involving the handling of more than 500 mCi of  $^{99m}\text{Tc}$ . The exposure expressed is in mR/100 mCi of the prepared product. An analysis of this exposure data is given in Mr. Robbins' paper<sup>6</sup>. The higher exposure levels for  $^{99m}\text{Tc}$ -HAM are due to the higher levels of  $^{99m}\text{Tc}$  handled for the resulting

\*The chips used in these studies, TLD-100 (1/8 x 1/8 x .0035 inch) were selected to have sensitivities within  $\pm 5$  percent of each other. In some  $^{99m}\text{Tc}$  studies, two sets of TLD's were exposed, one at twice the activity level of the other. These became the calibration standards for the two isotopes.



**Figure 3.** Transporting tray with lead holder for dose syringe plus items needed at injection site.



**Figure 4.**  $^{99m}\text{Tc}$  Exposure in mR/100mCi  $^{99m}\text{Tc}$  Prepared. Wrist TLDs anterior side only.

yield, and difference in handling procedures required. Unusual exposure data can be accounted for in specific handling operations. Yet, based on the left hand finger exposures recorded in this study, the preparation of 200 batches of 80 mCi each of  $^{99m}\text{Tc}$  during a year would result in approximately 17 R/year to the hand. While this is within the 75 rem/year NCRP limit, (it exceeds the 15 rem/year skin exposure recommendations) it only accounts for exposure from the preparation of one radiopharmaceutical.

A new radioisotope for tumor and heart scanning,  $^{129}\text{Cs}$  is produced by cyclotron\* irradiation and is air shipped to Cincinnati where the target material is chemically processed to extract the cesium as cesium chloride. The chemical extraction takes about 60 minutes. In the first few production runs, the  $^{129}\text{Cs}$  yield from each bombardment was 2 to 4 millicuries total. As the procedure was refined and the cesium yield increased, more shielding was incorporated throughout the extraction-purification process. Handling devices were likewise incorporated into the process. Figure 5 shows a pair of vice-grip pliers used to hold the aluminum target. A long bolt has been substituted for the adjustment screw on the pliers to add distance between the target and hands, yet maintain reasonable operation.

Figure 6 shows the special tools assembled for this procedure; tweezers permanently attached to tongs for handling the target cover, a long handle with a funnel stopcock in the slotted right end of the handle, a flexible pick-up tool and an allen wrench built into a long handle for removing the target cover. At the time of the cover removal the target was emitting more than 500 R/hour at 1 centimeter. One other device, figure 7, is a remote hydraulically operated syringe. By coupling two syringes tip to tip with small plastic tubing, one acts as a piston controlled by the other. The piston can push or pull the primary syringe plunger to deliver or take up liquid. The beauty of this system is that the personnel radiation exposure is essentially eliminated for this part of the procedure. Refinements on the procedure occurred over a period of 3 months and with each succeeding week we noted decreases in radiation exposure. Table II summarizes this exposure for the hands. Immediately after run #1 techniques to reduce the dose were incorporated. As will be noted, the effect was dramatic on run #2, one week later. With succeeding weeks, and with the employment of new shielding and tools, the average exposure dropped. For brevity of data presentation runs #3 and #6 are eliminated but fit as expected in the step-by-step exposure reduction - a 6-8 times reduction. If the exposures recorded in run #1 were received over a 50 week period, it would result in more than 8 rem exclusive of exposures in other duties with other radioisotopes. However, employing the techniques and shielding described, the  $^{129}\text{Cs}$  exposure is about 1 R/year.

\* In cooperation with the Naval Research Laboratory cyclotron, Washington, D.C.



Table II. TLD HAND DATA FOR  $^{129}\text{Cs}$  EXTRACTION

		Run	1	2	4	5	7
FINGERS AND WRIST	Right						
	Average(mR)		149	46	35	28	20
	Reduction		← x3 →		← x2 →		
	Left						
Average(mR)		147	35	22	21	16	
Reduction		← x4 →		← x2 →			
FINGERS ONLY	Right						
	Average(mR)		167	48.3	31.8	31.2	18.8
	For 50 Weeks(R)		8.4	2.4			1.0
	Left						
	Average(mR)		162	40	27	26	17
	For 50 Weeks(R)		8.1	2.0			0.9
Week Total(R)		8.2	2.2			1.0	

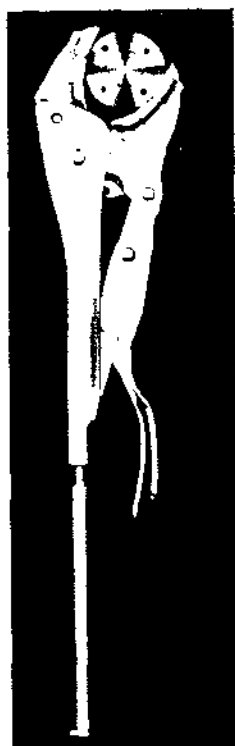


Figure 5. Modified vise grip pliers for holding irradiated cyclotron target secure and at a safe working distance.

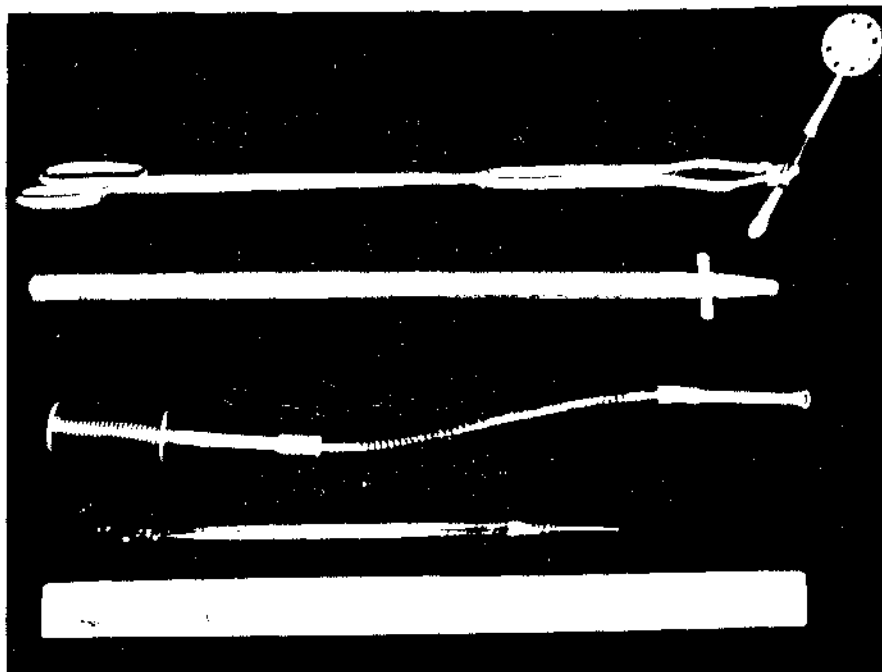


Figure 6. Special tools for  $^{129}\text{Cs}$  extraction. Top to bottom: tongs and tweezers, long handle with funnel stop-cock (right end), pick-up tool, and allen wrench in a long handle.

In summary, we have indicated some of the ways we have assessed and reduced personnel exposure during routine radioisotope handling in our laboratory. The results are within or close to the recommended guideline of 15 rems/year for the specific function analyzed. From our experience, personnel exposures in nuclear medicine laboratories can and must be reduced by modifying current techniques and practices. In a growing clinical field radiation

exposure assessments should be made every few months to determine where improvements can be made for the protection and exposure reduction of the laboratory personnel.

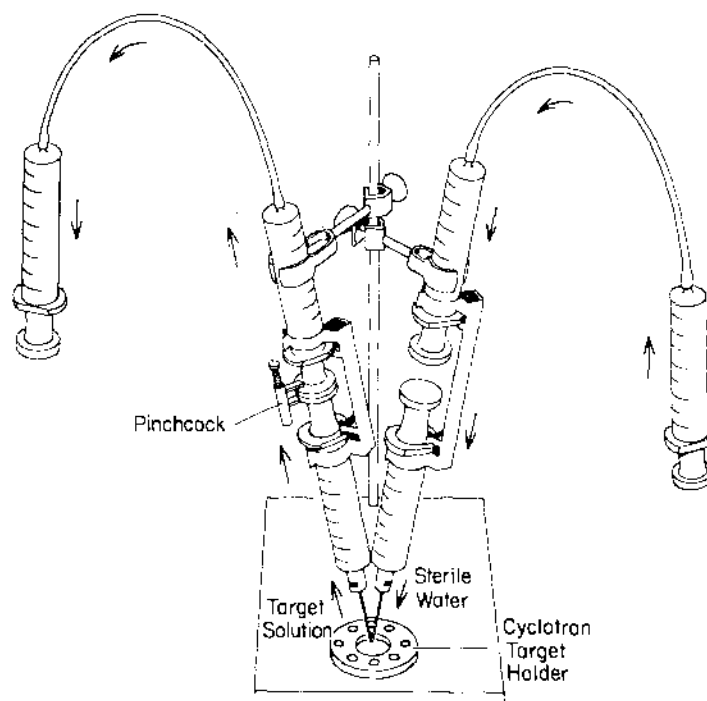


Figure 7. Remote control syringes used for target dissolution. The system on the right is used to wash the target, the one on the left is used to pick-up the solution.

#### REFERENCES

1. Lombardi, M.H., Beck, W.L., and Cloutier, R.J.: "Survey of Radiopharmaceutical Safety in Sixty-Nine Hospitals," Proceedings of Seventh Midyear Topical Symposium of the Health Physics Society, San Juan, Puerto Rico, December 1972, in press.
2. National Council on Radiation Protection and Measurements: "Basic Radiation Protection Criteria (NCRP Report No. 39)," National Council on Radiation Protection and Measurements, Washington, D.C., 1970, p. 91.
3. Neil, C.M.: "The Question of Radiation Exposure to the Hands from Handling  $^{99m}\text{Tc}$ ," *J. Nucl. Med.*, 10, 732-734 (1969).
4. Barnett, H.D. and Harris, C.C.: "A Means of Reducing Radiation Exposure from Technetium Generators," *J. Nucl. Med.*, 11, 180-181 (1970).
5. Robbins, P.J., Fortman, D.L., and Lewis, J.T.: "A New Lung Scanning Agent:  $^{99m}\text{Tc}(\text{Sn})\text{MAA}$ ," *Int. J. Appl. Radiat. Isot.*, 24, 481 (1973).
6. Robbins, P.J., Branson, B.M., and Grant, R.J.: "Reduction of Radiation Exposure to Personnel by the Use of New 'Instant' Techniques for Preparation of Technetium Radiopharmaceuticals," these Proceedings.

---

This paper supported in part by USPHS-FDA Contract #PH-86-67-212.

## RADIOPROTECTION IN A NUCLEAR MEDICINE UNIT

A. Favino <sup>+</sup>, G.F. Besseghini <sup>°</sup>  
Occupational Diseases Institute  
Pavia University- Italy

### Abstract

The main aspects of the radioprotection program of a Nuclear Medicine unit organically included in the services of an Occupational Diseases Institute are described and discussed. Analysis of the records of the environmental and personal dosimetry points out to the opportunity for monitoring particularly of the internal contamination. The need for employing usual diagnostic equipment for regular monitoring of the working staff is stressed. Due to the increasing diffusion of WBC for diagnostic applications, scheduling of this tool for monitoring of the personnel is recommended.

A transportable WBC of the shadow type susceptible to be transferred from the laboratory to a mobile unit in less than half an hour is described for clinical and radioprotection applications including monitoring of operators handling gamma emitters in medical units and in industry when no facilities for internal dosimetry are available.

Monitoring of the excreta of patients submitted to radiometabolic therapy has been one of the aims of the health physics programs in this Nuclear Medicine unit. An original system for monitoring liquid radioactive waste is described as a first step for ecologic control of spreading of radioactive contamination.

### Introduction

Problems related to safe handling of radioactive isotopes in medical practice are well known and have been widely discussed in a number of reports <sup>1-5</sup>. When applied to single medical institutions general regulations have to be tailored for the particular situation, the problem being often to meet requirements for high standards of safety as necessary in nuclear technology with the not unlimited resources of small units.

Starting a Nuclear Section in an Occupational Diseases Institute we were faced with a number of problems related to diagnostic, therapeutic and research applications of isotopes as well as to radioprotection of workers handling radionuclides.

<sup>+</sup>Physician - <sup>°</sup> Consultant physicist.

In this paper some of the main features of the radioprotection program of a Nuclear Medicine unit will be reported and discussed based on the observations made during first 3 years of practice. Present stage of development of the unit can be evaluated as comparable to a medium sized diagnostic laboratory, therapeutic applications being still limited to some experience with radioiodine treatment.

I.-Remarks after 3 years experience with a radioprotection program in a Nuclear Medicine unit .

General points of the radioprotection program are:

- I) a stressing accent on autoradioprotection as far as collection, registration and first evaluation of data are concerned : weekly check for white coats, fortnightly surveys for detection of laboratory contamination, monthly registration of film dosimetry data for environmental and personal monitoring, quarterly whole body examinations. The bulk of this job is committed to the physician of the operating staff ( 1 physician and 2 technicians).
- II) a decision to avoid heavy shielding structures in the laboratory. Location of the unit in a partly underground floor made preferable to keep windows which opened over a street of the town.
- III) a rigorous organization of the unit with a clear definition of the controlled zone and of compulsory pathway with a monitoring unit at the end. Part of the controlled zone are rooms for metabolic therapy and for waste disposal.

Planimetric organization of the laboratory, consumption of radionuclides, dosimetry of the surveilled zone and summary of data of the external irradiation and external and internal contamination of the working personnel are reported in fig.I and tables I,II,III. The following concluding considerations are drawn:

- I) environmental monitoring is worthwhile in a Nuclear Medicine unit if special shielding of walls and ceilings are not available because safe limits can be overcome if the amount of handled isotopes rises enough.
- II) personal monitoring is important especially as far as internal contamination is concerned. Notwithstanding observation of safety rules, internal contamination can occur:  $^{75}\text{Se}$  and  $^{131}\text{I}$  were mostly found. Uptakes and scanning instruments available in each nuclear medicine laboratory are of course useful for detecting contaminations of the order of a fraction of 1 microcurie of the used gamma emitters: periodical checks with a WRC as frequently as possible for satisfactory internal dosimetry however is suggested.

II.- Presentation of a transportable WRC of the shadow type.

In the country efficient WRC are available but a need is felt for monitoring of workers in organizations where WRC facilities are not provided as in some Nuclear Medicine units and in industry. Many examples of transportable WRC have been described<sup>6-8</sup>. A main feature of the present model is that it can be used both in the laboratory set up as in a mobile unit and that in 30 minutes its

t 1.5, damped, can be transferred, by an elevator, from the laboratory to the car, ready for emergency. At least 1% of most restrictive MPBB for gamma emitters with an accuracy of  $\pm 20\%$  can be appreciated in 20 minutes. This WBC is composed by 2 blocks of comparable weight: a 5 in. x 4 in. NaI(Tl) crystal shadowed by a 10 cm thick Pb shield elliptically collimated and a bed plane shielded by 5 cm Pb. Remote devices can move electrically the detector vertically from 66 cm to 114 cm from ground level and can adjust the segments of a chair geometry. The apparatus is equipped with a 800 channels analyzer with computation facilities, analogue display, digital data printing and magnetic tape recording. In the laboratory as well as in the mobile unit are provided a room for external decontamination, air filtering and conditioning; a whole body scanning bed is in progress.

### III.- Control of radioactive waste disposal

For gaseous waste a ventilation circuit is operating in the laboratory from the over-pressure WRC room to the filter of the laboratory cupboard ( fig. 7). At present the only gaseous waste is  $^{133}\text{Xe}$  expired in perfusion studies in a closed circuit and partly adsorbed at room temperature on a shielded activated charcoal, which is then deposited in a plastic closed bag to decay in the waste room, whereas the not absorbed fraction is ventilated through a short stack on the roof of a 3 floor building. Absorption procedure is not practical but it is suggested by the very closed proximity of other houses.

Solid waste, collected in plastic bags, are stored for some months until activity is less than  $1 \mu\text{Ci}/10 \text{ kg}$ ;  $^{75}\text{Se}$  contaminated material is kept distinct and stored for longer time.

For liquid radioactive waste we assumed that effluents from the laboratory can be evaluated, on the consideration of  $\text{MPC}_w$ , around 10% of the effluents from a proportionate radiometabolic therapy unit using  $^{131}\text{I}$ . So, if a treatment of radioactive waste is decided, the main point is to remove the activity released from treated patients. It has been calculated that, if 75 litres a day are released from the WC of the patient, 1 clarifier and 3 tanks in series with an individual dilution volume of 1500 litres, can provide a 37% reduction each of the concentration of the radioactivity so that only a fraction of 2% can reach the well. Two electrovalves allow discharge into the well from the laboratory and from the patient line at a concentration lower than  $10^{-3} \mu\text{Ci}/\text{ml}$ . Two G.M. detectors shielded by 9 cm Pb are operating to remove the radioactivity released from the laboratory and to measure the total discharged activity which is given by the product of the known volume voided by the pump ( 1240 litres) by its measured radioactive concentration. Due to further dilutions ( 700  $\text{m}^3/\text{day}$  in the sewerage collector and 4200  $\text{m}^3/\text{min}$  in river Ticino) present release is less than 1/10000 of the  $^{131}\text{I}$   $\text{MPC}_w$  for the population. The counterpart is the care which is requested for looking after the system ( as for repair of valves, pumps, cleaning of detectors ).

Table I

Isotopes ( mCi) handled in first 3 years of laboratory activity

Year	<sup>131</sup> I	<sup>99m</sup> Tc	<sup>198</sup> Au	<sup>197</sup> Hg	<sup>75</sup> Se	<sup>87m</sup> Sr	<sup>67</sup> Ga	<sup>169</sup> Yb	<sup>133</sup> Xe	<sup>125</sup> I	<sup>3</sup> H
1	43	80	16	5	11	-	-	-	-	0.04	0.02
2	64	1340	39	6	21	68	-	-	-	0.06	0.02
3	101	4546	67	7	55	80	40	3	475	0.22	0.03

Table II

Summary of dosimetry records of operators ( subjects A,B,C)

Year	µCi/yr internal contamination			µCi/yr white coat contamination			mrem/month external irradiation (film)				
	A	B	C	A	B	C	A	B	C		
1	0.01	-	-	0.2	0.3	0.08	20	20	20		
	( <sup>75</sup> Se )										
2	-	0.01	-	0.4	0.5	0.09	20	20	20		
	( <sup>131</sup> I )										
3	0.33	-	0.01	3.3	0.5	0.9	20	20	20		
	( <sup>131</sup> I )			( <sup>131</sup> I )							

A= physician : examines the patient - administers diagnostic and therapeutic doses ; B= technician : cleans glassware, stocks radioactive waste ; C= technician : performs radiochemical work. Every one does in vivo investigations ( scanning, fast dynamic studies).

Table III

External dosimetry of surveilled zone in the laboratory (mrem/month)

Year	door of the room for waste disposal	<u>room of sources deposit</u>			all other spots
		wall	window	unstairs	
1	15	37	6	0	20
2	29	50	12	20	20
3	44	89	49	20	20

The following spots have been monitored: uptake room, scanning room. in vivo dynamic studies room, radiochemical laboratory, animals rooms.

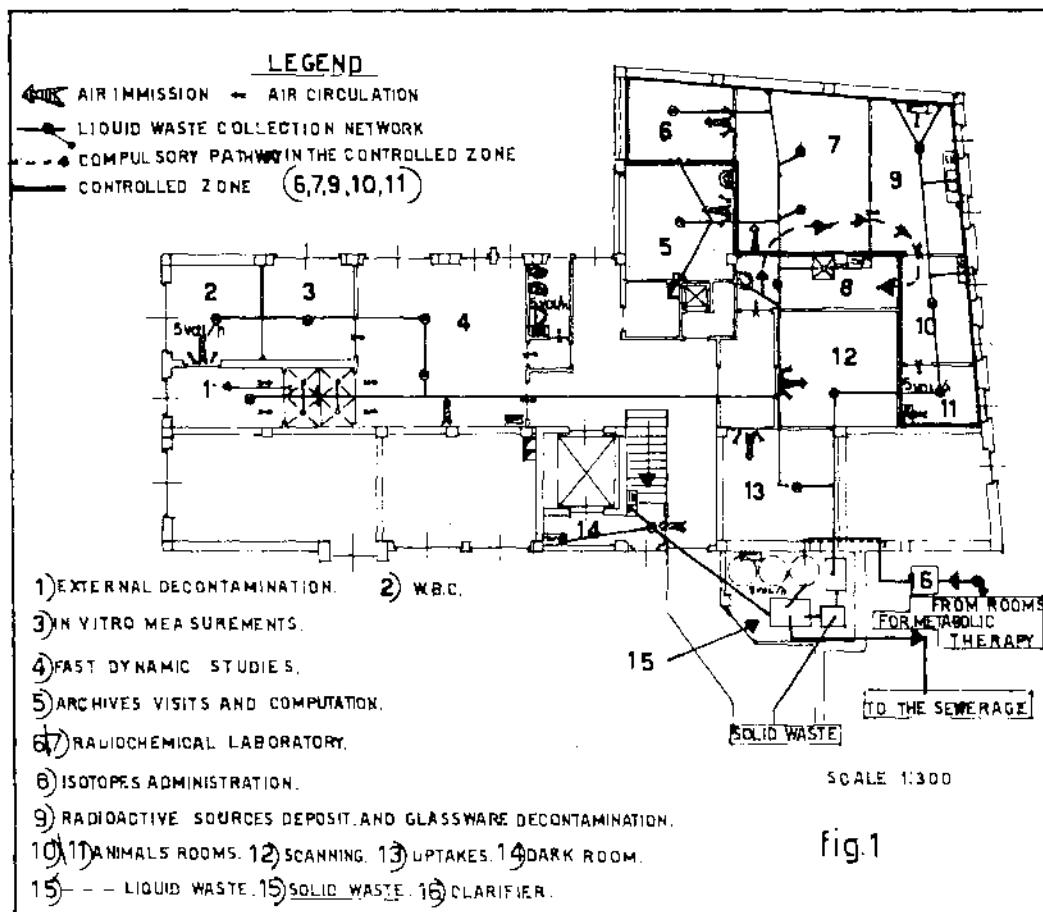
Table IV

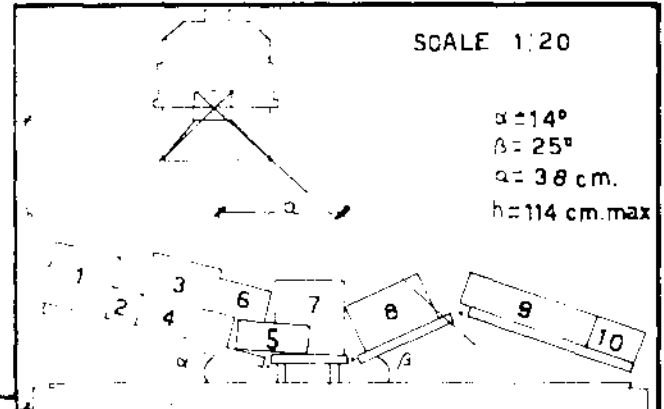
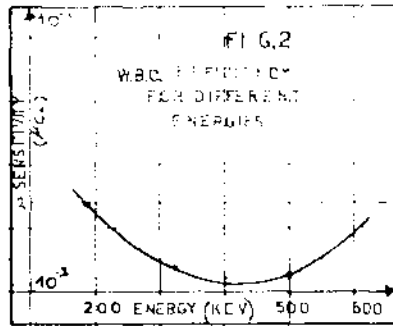
Percentage of frequency of detection of contamination in absorbing coverings higher than  $1 \times 10^{-4}$  µCi/cm<sup>2</sup> (fortnightly surveys).

Year	percentage
1	21.2
2	14.1
3	19.4

### References

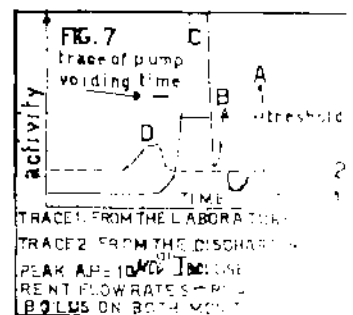
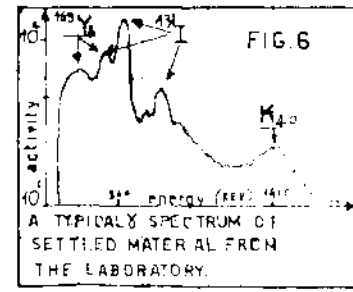
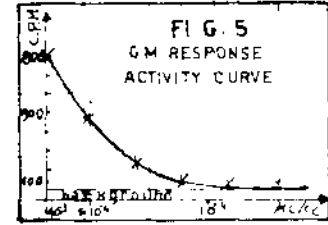
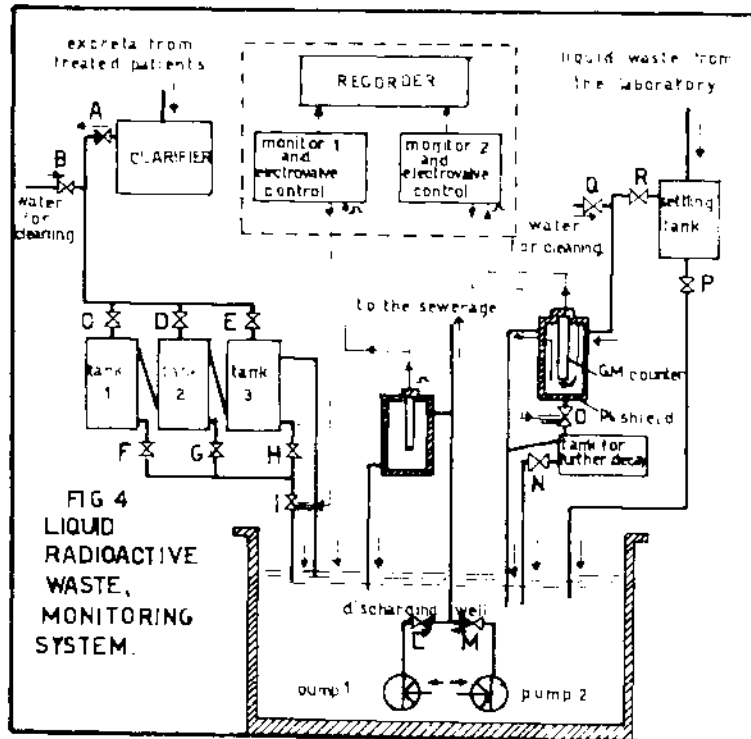
- 1.- IAEA : The management of radioactive waste produced by radioisotopes users- Safety series n. T2, T965
- 2.- IAEA : The management of radioactive waste produced by radioisotopes users: Technical addendum-Safety series n. T9, T966
- 3.- ICRP : Report of Committee V on the handling and disposal of radioactive materials in hospitals and medical research establishments-ICRP Publication 5, Oxford, Pergamon ,T964
- 4.- Vennart J. : Radiation hazard in work with radioisotopes .p. 188 in : Radioisotopes in medical diagnosis -Belcher E.H., Wetter H. ( Eds. ) , London , Butterworths . T971
- 5.- Blake K.C.H., Rapley L.F. : Disposal of radioactivity into a city sewerage system- Wlth. Phys. , 23 , 86T, T972
- 6.- Parker D. , Anderson J.J. : A portable whole body counter- Wlth. Phys. , 13 , T4T, T967
- 7.- Scott L.M., Abele W.M., Brvant E.H., Cromwell H.W., West C.M. : Design and development of a mobile in vivo radiation monitoring laboratory - Am. Ind. Hyg. Assoc. J. , 30 , T67, T969
- 8.- Howard L.E. , Spickard J.H. , Wilhelmsen M. : A human radioactivity counter and medical van -Wlth. Phys. , 21 , 417, T971.





**FIG. 3**  
WBC SHADOW IN TPE A GEOMETRY - EFFICIENCY FOR DIFFERENT BODY SEGMENTS AND ENERGIES

BODY SEGMENTS	I-131 364-kev	I-131 637-kev	FE-59 1095-kev	FE-59 1292-kev
1 HEAD	0.027%	0.027%	0.018%	0.017%
2 NECK	0.136%	0.147%	0.045%	0.026%
3 UPPER TRUNK	0.051%	0.059%	0.029%	0.021%
4 ARM	0.050%	0.047%	0.027%	0.024%
5 FORE ARM	0.023%	0.056%	0.007%	0.007%
6 MIDDLE TRUNK	0.056%	0.060%	0.032%	0.032%
7 LOWER TRUNK	0.032%	0.036%	0.021%	0.014%
8 LIMB	0.019%	0.019%	0.018%	0.014%
9 LEG	0.001%	0.001%	0.001%	0.001%





## RADIATION PROTECTION IN A NUCLEAR PHARMACY

Rodney D. Ice and Kenneth R. Hetzel

College of Pharmacy and Nuclear Medicine Section  
The University of Michigan  
Ann Arbor, Michigan 48104

### Abstract

Regional nuclear pharmacies are emerging throughout the world to meet the increased demands for radiopharmaceuticals. A radiation protection program for a nuclear pharmacy encompasses facility design, quality control, dispensing and documentation, and provides for increased utilization of radiopharmaceuticals. The resultant radiation protection program is synergistic between health physics and pharmacy principles.

### Introduction

Regional nuclear pharmacies provide radiopharmaceutical services for multiple hospitals located over wide geographic areas. Such pharmacies provide the necessary pharmaceutical expertise for preparing radioactive chemicals into pharmaceuticals and also provide an increased assemblage of radiopharmaceuticals at reduced costs to many hospitals that cannot individually afford nuclear pharmacy services.

A regional nuclear pharmacy may have inventories of 5-10 Curies of radioactive materials, with greater than Curie quantities of Iodine-131, Xenon-133 and Molybdenum-99/Techneium-99m.

The University of Michigan Hospital Regional Nuclear Pharmacy, over the past two years, has evaluated radiation protection as applied to nuclear pharmacy practice. Facility Design, administered Doses, product Dispensing and quality control Documentation are considered to be the principle axioms for the development of a radiation protection program in nuclear pharmacy.

### Nuclear Pharmacy Design

Nuclear pharmacies must always incorporate health physics principles associated with "wet" radiochemistry laboratories. In addition, special consideration must be given to pharmaceutical techniques, i.e. aseptic preparation of parenteral products, synthesis of radiolabeled organic compounds, dispensing of radioactive gases, repeated handling of syringes containing radioactive materials, maintenance of product quality and potential contamination of non-radioactive pharmaceuticals.

Traffic flow patterns within a nuclear pharmacy must be defined for efficient utilization, as well as radiation protection planning. A general consideration of functional separation of activities within the pharmacy aids in radiation protection and pharmaceutical quality. The package receiving and shipping area should be a separate room to minimize potential radioactive contamination and to reduce airborne dust and particulate matter from entering the compounding area. A dispensing area, separate from the compounding area, will minimize traffic where bulk quantities of radioactive material are used and parenteral products formulated. In addition, a dispensing window will reduce the access of unauthorized personnel. The quality control laboratory should be housed in a separate room, as it is predominately an instrumentation facility, and considered only to contain tracer quantities of radioactive materials. Finally, because adjunctive (non-radio-

active) pharmaceuticals are often stocked and dispensed with radiopharmaceuticals, a separate room for pharmaceuticals again provides radiological and pharmaceutical quality assurance.

An adequate floor plan for efficient nuclear pharmacy design is shown in Figure 1. The compounding and dispensing rooms form an integral, limited access area for the storage, preparation and dispensing of radiopharmaceuticals. Low traffic flow in the compounding area reduces potential spread of contamination. Dispensing of unit-dose radiopharmaceuticals to medical personnel is conducted through the dispensing window; a procedure which guarantees limited access only to authorized individuals and further reduces the potential spread of radioactive contamination. The non-radioactive pharmaceutical dispensing area and quality control laboratory are located across the hall from the compounding and dispensing rooms. Adjacent to the nuclear pharmacy is a separate radiochemical laboratory where packages are received and shipped. Included in the radiochemical laboratory is an absolute filtered radiochemical hood for the storage and dispensing of iodine-131 and xenon-133.

Too often, one segment of a laboratory bench is chosen for all radiopharmaceutical preparation. Such a design can offer satisfactory health physics considerations, but poses a serious potential risk of product cross contamination and erroneous product selection and dispensing. Nuclear pharmacy design should provide separate work areas for the compounding and dispensing of radiopharmaceuticals. To facilitate both health physics and pharmacy requirements, the University of Michigan Nuclear Pharmacy has designed a lead laminated plywood ( $\frac{1}{2}$  inch lead) compounding enclosure shown in Figure 2.

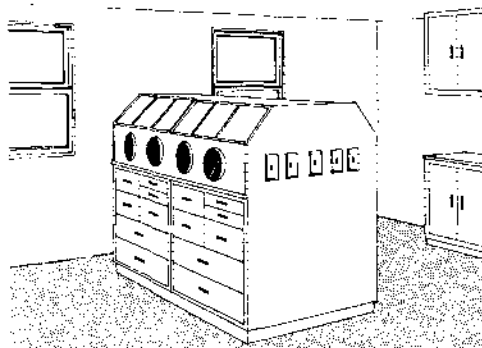


Figure 2  
Compounding Enclosure

for aseptic subdivision of parenteral products and not to vent volatile radioactivity. Laminar flow hoods are manufactured to provide either horizontal or vertical air flow. Since horizontal air flow is directed towards the operator and could lead to a severe personnel contamination hazard, it must be stressed that only vertical laminar air flow hoods should be chosen for use in nuclear pharmacy. Sterile air is provided in a laminar air flow hood by a series of HEPA filters which are designed to remove all

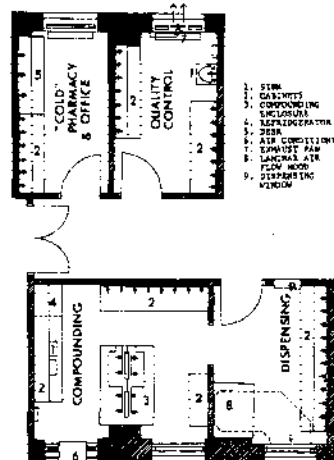


Figure 1  
Floor Plan

This installation is specific for technetium-99m compounding and contains sliding lead glass (1.7 mm lead equivalent) doors across the top. The primary design consideration of this enclosure was consolidated shielding with separation of compounding involved with technetium radiopharmaceuticals: a union of radiation protection with pharmaceutical quality assurance.

Within a nuclear pharmacy, laminar air flow hoods are used to provide a sterile work area

dust and bacteria from the air before entering the working space. To insure the sterility of the working compartment, articles should be autoclaved or suitably disinfected before entering the hood.

Several problems associated with shielding are evident with a nuclear pharmacy. Too often personnel without health physics training fail to consider the energy of associated radiation and assume that shielding designed for technetium-99m is suitable for fluorine-18, iodine-131 or phosphorous-32. The designation of separate work areas for different radionuclides allows for optimal shielding design associated with each product. Such considerations, however, should be flexible to allow for adaptation of new procedures and new radionuclides. Nuclear pharmacies should be cautioned against stacking heavy, free-standing, lead blocks, since they may present serious hazards, i.e. traumatic injury to legs and feet from accidental falling blocks, radiation exposure through non-interlocking joints, and excessive weight induced structural damage to cabinetry. Inexpensive and highly effective shielding for low energy radionuclides can be obtained with lead perchlorate shields suggested by Barnett and Harris.<sup>1</sup>

#### Radiopharmaceutical Dispensing

The proper dispensing of radiopharmaceuticals will affect the absorbed radiation dose to the patient and the pharmacist. Maximal patient protection is achieved by the utilization of unit dose radiopharmaceuticals. All radiopharmaceuticals are dispensed from the nuclear pharmacy on prescription. The prescription indicates the requested study, the preliminary diagnosis, patient name, height, weight, age and the time that the patient is to receive the dose. With this information, the pharmacist can correlate the radiopharmaceuticals with the proposed study and patient information in such a manner that the patient will receive the optimum dose. Each product is dispensed precalibrated to the time of administration, and receives duplicate assays of the radioactive contents. The radiopharmaceutical is dispensed with a label indicating the patient's name, time and route of administration, date, prescription number, physician's name and pharmacist's initials. Unit dose dispensing has reduced the potential for dispensing and administration errors associated with major drug delivery systems.<sup>2</sup>

It is believed that the concept of unit dose dispensing of radiopharmaceuticals is an efficient and effective method of reducing errors associated with the administration of radioactive pharmaceuticals.

As an additional radiation protection device for the pharmacist, as well as maintaining the pharmaceutical quality of the products, our pharmacy utilizes a shielded syringe-valve dispensing system as modified from Hoar.<sup>3</sup> A schematic diagram is given in Figure 3. The solution to be unit dose dispensed is drawn from the shielded large volume syringe (B) into the shielded large volume syringe (B) via a 3-way valve (C). A unit dose is subsequently dispensed by attaching a small volume syringe (D) to the dispensing part of the 3-way valve, drawing off the desired volume, attaching a needle, assaying the contents and affix-

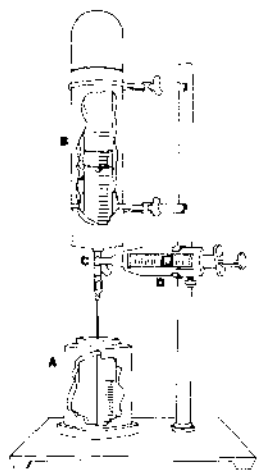


Figure 3  
Dispensing System

ing the label. This dispensing process is conducted within the laminar air flow hood to insure sterility of the product.

The use of the 3-way valve dispensing unit not only insures pharmaceutical quality of the product, but decreases the hand and finger doses to the pharmacist with an average dispensing time of 25 seconds. Several reports 4,5,6 indicate the potential problem hand and finger doses to medical personnel. Implementation of a 3-way valve dispensing system significantly reduces exposure problems since the pharmacist need not handle the barrel of the syringe.

#### Administered Radiation Dose

The reduction of medical exposures and patient protection have always been the goals of nuclear medicine personnel. Tremendous gains in reducing medical exposures have been accomplished by using shorter half-life radiopharmaceuticals, by increasing biological turnover times, and by selection of the best radionuclide for labeling purposes. However, little consideration has been given to the administered dose. The relative assessment of benefit versus risk can only be accomplished if the administered dose is tailored to the individual patient. Too high an administered dose results in increased absorbed dose. Too low an administered dose may result in a missed diagnosis or a readministration of the drug. The Nuclear Pharmacy has evaluated the administered dose regarding the genetically significant population, the method of dose determination and the pharmacists role in product selection.

An evaluation of 200 randomly selected patients receiving a radiopharmaceutical indicated that 12.5% were less than 18 years old; 34.5% were between 18 and 45, and 53% were greater than 45 years old. The genetically significant population (less than 45) represents a 13% increase as compared to the national average of 1970. Leblanc and Johnson<sup>7</sup> have also indicated that exposures from nuclear medicine procedures increased from 5% gonadal exposure/admission in 1964 to 11% in 1968.

Administered radiopharmaceutical doses vary from clinician to clinician. The selection of an administered dose may be from a table of doses determined from past experience,<sup>8</sup> the application of body weight, e.g. mCi/Kg or from a series of rules, e.g. Young's rule, Clark's rule or surface area.<sup>9,10,11</sup>

While adult administered doses are fairly well established, large variations in pediatric doses are apparent. Administered doses based upon age show great limitation when one considers the variability of a given age. For example, the 3rd percentile of a 10 year old girl is 53.2 lbs., while the 97th percentile is 101.9 lbs.<sup>12</sup> Administered doses determined by weight, while better than doses determined by age, usually underestimate the requisite clinical dose. The underestimated dose is due to 1) weight changes as a function of the cube of linear dimensions while the necessary photon fluence for adequate lesion localization varies with the square of linear dimensions, and 2) ratios of organ/body weights in infants are greater than those observed in adults.

To provide a uniform and reliable method of computing administered radiopharmaceutical doses, for the broad spectrum of patients seen in our clinic, Nuclear Pharmacy employs body surface area as modified by height and weight. Administered doses, in mCi/m<sup>2</sup> are usable for all patients regardless of variations in weight, height, age or sex.

As a further consideration of the absorbed radiation dose, our pharmacists participate in the selection of the radiopharmaceutical for the patient. Qualified nuclear pharmacists have the

necessary training in radiopharmaceuticals, biopharmaceutics and metabolism to guide the physician in selecting the optimum drug to obtain the maximum diagnostic information. In addition, the nuclear pharmacist has played a vital role in identifying patients that may have drug interactions which prevent the meaningless use of a radiopharmaceutical. For example, a request for a red cell survival test one week after a gallium-67 scan.

Radiopharmaceutical Documentation

Good radiation protection principles require adequate record keeping to evaluate personnel methods and product control. Pharmacy requires substantial record keeping to validate a product's suitability for human use. To meet the requirements of both health physics and pharmacy, a product quality control system has been developed. A schematic diagram of the quality control program is shown in Figure 4.

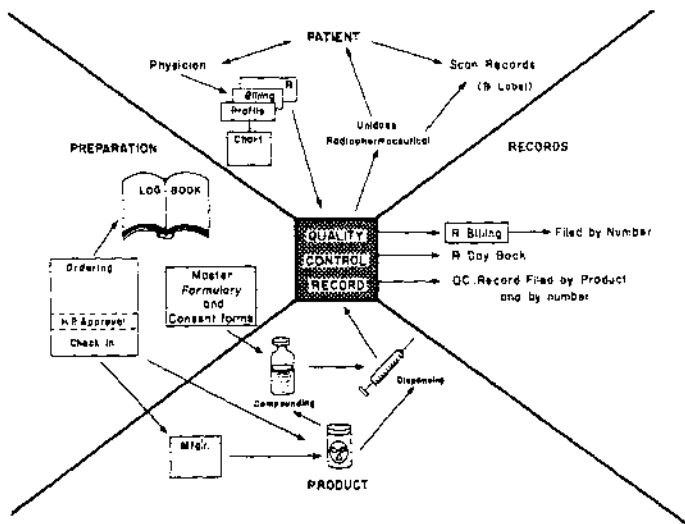


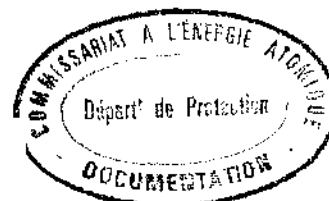
Figure 4  
Quality Control Program

Each product received into or compounded by the nuclear pharmacy has an assigned quality control card (center). Each product is further designated by a number indicating the date received or compounded. All compounded radiopharmaceuticals are prepared in accordance with a master formulary. All records of tests, e.g. sterility, apyrogenicity, particle size, radiochemical purity, chemical purity, radionuclidic purity, pH, etc. are indicated on the quality control card. All unidoses dispensed from the product are also recorded with the

quality control record. In this manner, a complete history of any radiopharmaceutical can be ascertained with a brief glance at the quality control record card.

As part of our quality control program, we require that used syringes be returned to the nuclear pharmacy. This alleviates the hazard of loose contaminated syringes and also allows the syringe to be reassayed. In this manner, complete assessment of the administered dose can be determined. LeBlanc and Johnson<sup>13</sup> and Abdel-Dayem<sup>14</sup> have reported on retained activity of Xenon-133 within syringes. Freedman<sup>15</sup> has described a similar deposition of technetium-99m sulfur colloid in the rubber plunger of disposable syringes. Our reassay procedure indicates that many radiopharmaceuticals have residual activity in syringes, especially radio-labeled proteins.

As part of the documentation concern for radiopharmaceuticals, our pharmacy has assumed the responsibility for the maintenance of all records necessary for clinical trials of new radiopharmaceuticals, as well as validating drug interactions or adverse reactions. Within our hospital, the patients chart is available to the pharmacist during the prescription preparation. A review of previous or existing drug therapy by the pharmacist can offer lead to the



explanation of modulation in drug distribution. When an adverse reaction occurs, our nuclear pharmacy coordinates the compilation of associated data and the reporting of the reaction to the professional societies and concerned authorities.

#### Conclusions

1. The objectives of pharmacy and health physics are synergistically compatible in reducing personnel and patient radiation exposure.

2. Methods for reducing radiation exposures within regional nuclear pharmacies while maintaining large inventories and increased utilization of radiopharmaceuticals requires adequate facility design, consideration of administered dose, unidose dispensing and quality control documentation.

3. Surface area measurements used in consideration of administered doses have aided in providing uniform and reliable scans between patients, especially in pediatric nuclear medicine.

#### References

1. H.D. Barnett and C.C. Harris, "A Means of Reducing Radiation Exposure from Technetium Generator Operations", J. Nucl. Med., 11, 180-181 (1970).
2. C. Timmer, "Our Unit Dose System Cuts Medication Errors", Pharm. Times, 50-58 (April 1972).
3. M.E. Hoar, "A Study of Methods Involved in Extemporaneous Syringe Filling and Degree of Contamination", Drug Intel. Clin. Pharm., 7, 132-137 (1973).
4. C.M. Neil, "Question of Radiation Exposure to Hand from Handling  $^{99m}\text{Tc}$ ", J. Nucl. Med., 10, 732-734 (1969).
5. R.S. Clayton, J.E. White, M. Brieden and A. Rodriguez, "Skin Exposure from Handling Syringes Containing Radioactive Isotopes", Amer. J. Roentgenol. Radium Ther. Nucl. Med., 105, 897-899 (1969).
6. Y. Takaku and T. Kida, "Radiation Dose to the Skin and Bone of the Fingers from Handling Radioisotopes in a Syringe", Health Phys., 22, 295-297 (1972).
7. A. LeBlanc and P.C. Johnson, "Medical Radiation Exposure Survey in a Hospital with Nuclear Medicine Laboratory", Health Phys., 19, 433-437 (1970).
8. J.J. Conway, "Considerations for the Performance of Radionuclide Procedures in Children", Semin. Nucl. Med., 2, 305-315 (1970).
9. J.G. Kereiakes, H.N. Wellman, G. Simmons and E.L. Saenger, "Radiopharmaceutical Dosimetry in Pediatrics", Semin. Nucl. Med., 2, 316-327 (1972).
10. M.J. Reilly, Ed., "Pediatric Drug Dosage", Amer. J. Hosp. Pharm., 29, 699-700 (1972).
11. H.C. Shirkey, "Usual Doses for Infants and Children", Dosage-Posology Handbook, Am. Pharm. Assn., Wash. D.C. (1965) pp 5-10
12. "Documenta Geigy, Scientific Tables", 6th ed., Geigy Pharmaceuticals, Ardsley, New York, (1962) pp 613-622.
13. A.D. LeBlanc and P.C. Johnson, "The Handling of Xenon-133 in Clinical Studies", Phys. Med. Biol., 16, 105-109 (1971).
14. H.M. Abdel-Dayem, "Handling of Radioactive Xenon-133 Dissolved in Saline", J. Nucl. Med., 13, 231 (1972).
15. G.S. Freedman, "Sulfur Colloid- $^{99m}\text{Tc}$  Losses in a Disposable Syringe", Radiology, 99, 197-198 (1971).

## AEROSOLS AND LUNG MODELS

### РАДИОАКТИВНЫЕ АЭРОЗОЛИ КАК ФАКТОР ВНУТРЕННЕГО ОБЛУЧЕНИЯ ПРИ ИСПОЛЬЗОВАНИИ РАДИОАКТИВНЫХ ВЕЩЕСТВ

БАДЬИН В.И., БАТОВА Э.Г., ПАРХОМЕНКО Г.М.

Институт биофизики Министерства здравоохранения СССР, Москва  
Союз Советских Социалистических Республик

In this report we consider the problem of determining the probable accumulation of the isotopes in the organism. These questions are solved on the basis of values of mean air contamination.

The experimental results were applied to the regulation of the air contamination.

Authors have proposed to use the term "the standard work conditions" in radiation protection practice. These "conditions" are characterized by the hygienical safety factor equal to 5, which should be included in the values of regulated PMS.

Общеизвестные принципы организации производства позволяют обеспечить безопасные условия труда для персонала только на базе норм радиационной безопасности, в основе которых лежит лимитирование дозовых нагрузок (первичные дозиметрические характеристики), определяющих степень опасности непосредственно для индивидуума.

Одновременно действует статус среднегодовых допустимых концентраций (вторичных дозиметрических характеристик), относящихся к параметрам среды, в которой работает персонал, и характеризующих безопасные условия труда.

Именно здесь, по мнению авторов, существуют принципиальные трудности как в подходах к определению этих норм РБ, так и в оценках лучевых нагрузок по показателям загрязненности воздушной среды.

В первую очередь в проблеме "Радиоактивные аэрозоли как возможный фактор внутреннего облучения" возникает задача перехода от величины экспериментально определяемого среднего загрязнения воздуха к вероятному накоплению изотопа в организме на основе данных по статистическим закономерностям распределения концентраций и различным дозиметрическим характеристикам радиоактивных аэрозолей.

Во всех случаях при качественной характеристике вдыхаемого радиоактивного вещества фактически ведется речь о модели аэрозолей, в той или иной степени аргументированной. Например, некоторая модель аэрозолей может представляться как суперпози-

ция в том или ином соотношении газообразных соединений изотопа, мелкодисперсной и грубодисперсной аэрозольной компоненты различной растворимости.

В качестве примера остановимся на влиянии дисперсности пыли на величину отложения в дыхательном тракте. Радиоактивные аэрозоли дезинтеграции "горячих" лабораторий, как правило, относятся к классу грубодисперсных, а распределение размеров аэрозольных частиц с достаточной точностью описывается логарифмически нормальным законом, характеризующимся в случае плутония следующими параметрами:  $\sigma_g = 1,6 \pm 3,4$  мкм,  $\beta_g = 2,0 \pm 2,7$  аэродинамический  $\sigma_{ae} = 2,4 \pm 5$  мкм и активным среднегеометрическим аэродинамическим радиусом  $\sigma_{ag}$  аэр. =  $20 \pm 50$  мкм. Учитывая вышеприведенную дисперсность, можно считать, что мгновенное распределение активности по дыхательной системе характеризуется следующими величинами: для носоглотки мгновенный общий коэффициент задержки активности равен  $K_{a_1}$  общ = 90%, для области трахеи - бронхиальное дерево  $K_{a_2}$  общ = 4%, для собственно легких (от дыхательных бронхиол и ниже)  $K_{a_3}$  общ = 2% /1/.

Таким образом, первоначальное отложение активности в собственно легких в этом примере будет значительно в 5-10 раз ниже, чем по принятой в настоящее время модели расчета СДК.

Весьма важным для ряда изотопов, которые могут находиться как в дисперсной фазе, так и в дисперсионной среде (например, тритий, углерод, ртуть, иод и др.), является вопрос об относительной роли перкутанного поступления в организм из воздуха.

На примере паров ртути сравним перкутанный и ингаляционный пути поступления. Авторами совместно с Ю.М. КОВАЛЕНКО было показано, что равновесное содержание ртути в организме в мкюри для первого случая выражается формулой  $A_1 = 33 \left(\frac{c}{c_0}\right)^2 \times \frac{S \cdot c}{\lambda}$  (1) а для второго  $A_2 = K_a^{отв} \times \frac{S \cdot c}{\lambda}$ , где  $S$  - концентрация паров ртути в мкюри/см<sup>3</sup>,  $c/c_0$  - относительная упругость паров ртути,  $S$  - поверхность тела человека в см<sup>2</sup>,  $\lambda$  - постоянная выведения ртути из организма в мин<sup>-1</sup>,  $\nu$  - скорость дыхания в см<sup>3</sup>/мин. Отсюда  $\frac{A_2}{A_1 + A_2} = \left[ 33 \frac{S}{\nu K_a^{отв}} \left(\frac{c}{c_0}\right)^2 + 1 \right]^{-1}$  (2) Анализ показывает, что для стандартного человека через кожу поступает от 2% до 65% всей ртути при изменении относительной упругости от 1% до 10%.

Анализ радиационной обстановки при загрязнении воздушной среды радиоактивными аэрозолями показал, что распределение их концентраций во времени и пространстве подчиняется логарифмически нормальному закону. Последнее представляется объяснимым, если, в частности, предположить, что распределение интенсивности источников загрязнения описывается усеченным гиперболическим законом, когда вероятность появления концентрации "с" обратно пропорциональна ее величине. Тогда распределение логарифмов концентраций будет равновероятным, а композиция таких распределений приводит к логарифмически нормальному закону.

Величина такого важного параметра, как стандартное геометрическое отклонение концентраций ( $\beta_g \geq 1$ ) является при прочих равных условиях хорошим показателем степени радиационной надежности технологического процесса, а также определяет и вероятную величину дозовой нагрузки и ее разброс. Значение  $\beta_g$  для эмпирических распределений, как видно из таблицы лежит в пределах 2-13. Поскольку дозу внутреннего облучения можно приближенно представить как произведение трех случайных величин загрязнение воздушной среды помещения, времени пребывания человека в них



и проскока аэрозолей через средства индивидуальной защиты, то при условии, что каждая из них независима и подчиняется лог-нормальному закону с дисперсиями  $\lg^2 \beta_{\text{зс}}$ ,  $\lg^2 \beta_{\text{зг}}$ ,  $\lg^2 \beta_{\text{зк}}$ , соответственно, дисперсия дозы будет равна:

$$\lg^2 \beta_{\text{зд}} = \lg^2 \beta_{\text{зс}} + \lg^2 \beta_{\text{зг}} + \lg^2 \beta_{\text{зк}}. \quad (3)$$

Если величина загрязнения воздушной среды помещения и время пребывания в них человека скоррелированы (коэффициент корреляции  $\Gamma$ ), то

$$\lg^2 \beta_{\text{зд}} = \lg^2 \beta_{\text{зс}} + \lg^2 \beta_{\text{зг}} + 2 \lg \beta_{\text{зс}} \cdot \lg \beta_{\text{зг}} \Gamma. \quad (4)$$

Поскольку обычно  $\beta_{\text{зд}} < \beta_{\text{зс}}$ , то  $\Gamma < 0$ , что отражает известное положение: чем больше загрязненность, тем обычно меньше время пребывания в ней человека.

Реальные дисперсии первичных дозиметрических характеристик приведены в таблице.

Таблица  
Стандартное геометрическое отклонение радиационных характеристик ( $\beta_{\text{з}}$ ).

№№	$\beta_{\text{з}}$ внеш. для индивидуальных доз гамма-нейтронного облучения		$\beta_{\text{зн}} \approx \beta_{\text{зд}}$ для величин накопления радиоактивных веществ в организме	$\beta_{\text{зг}}$ для величин поступления радиоактивных веществ в организм	$\beta_{\text{зс}}$ для концентраций радиоактивных аэрозолей	
	$\gamma$	$\pi$			обычная	при рем. эксплуат. работах
1	1,8	1,9	2,8	2,8	5,7	3,9
2	2,5	1,7	2,3	2,3	4,2	2,8
3	2,3	2,4	2,3	1,8	2,5	4,1
4	2,3	2,1	2,3	5,1	6,6	3,2
5	2,1		2,3	1,7	2,4	10,8
6	2,6		5,0	2,1	2,0	12,9
7	2,3		1,8			
8	1,6					
9	1,8					
10	1,7					
Среднее значение	$2,1 \pm 0,3$	$2,0 \pm 0,2$	$2,7 \pm 0,7$	$2,4 \pm 0,8$	$3,9 \pm 1,6$	$6,2 \pm 3,7$

Таким образом, специфика работы заключается в том, что существует разброс показателей загрязнения воздуха и соответствующее ему размытие первичных дозиметрических характеристик.

Кроме того, величины накопления изотопов в организме людей, работающих в "одинаковых" условиях, имеют значительно больший разброс, чем биологические константы.

Этот факт требует введения допустимых рабочих пределов загрязнения воздуха, абсолютная величина которых меняется каждый раз в зависимости от дисперсии величин накопления изотопов в организме и отличается от СДК на коэффициент запаса.

В качестве примера рассмотрим вариант, когда распределение величин первичных дозиметрических характеристик подчиняется логарифмически нормальному закону с  $\beta_{\text{з}} = 5$ . В этом случае 21%

работающих будут иметь нагрузки значительно выше средней. Это означает, что при среднем загрязнении воздуха в пределах СДК и обычном режиме труда у 1/5 работающих дозовая нагрузка на организм будет превышать СДН, что нельзя считать удовлетворительным. Поэтому для оценки радиационной обстановки по усредненным значениям дозиметрических характеристик необходимо ввести новый критерий, восполняющий отсутствие у средней величины индивидуальных черт. Этот критерий может быть сформулирован следующим образом: безопасной средней величиной дозиметрической характеристики можно считать такую величину, при которой вероятность появления радиационной нагрузки, превышающей допустимую, будет приемлемо мала.

Исходя из всего изложенного, для определения допустимого рабочего предела коллективных показателей радиационной обстановки в численное значение среднегодовой допустимой величины (СДВ) или СДК в случае аэрозолей следует ввести коэффициент запаса для индивидуума  $K_{\text{и}}$ .

Очевидно,  $K_{\text{и}}$  определяется величиной  $\beta_{\text{гд}}$  и принимаемым коэффициентом риска  $\beta = \frac{100}{a}$  ( $a$  - % людей, имеющих радиационную нагрузку выше допустимой).

Выражение для вычисления  $K_{\text{и}}$  может быть получено следующим образом.

Доля людей ( $\frac{a}{100} = \frac{1}{\beta}$ ) с радиационной нагрузкой больше какой-либо величины и имеющей интегральное распределение, подчиняющееся лог-нормальному закону, равна

$$1/\beta = 1 - F(m) = 0,5 [1 - \Phi(\xi)],$$

где  $\Phi(\xi)$  - интеграл вероятности.

Отсюда  $1 - 2/\beta = \Phi(\xi)$  или выражая  $\xi$  через обратную функцию,  $\xi = \Phi^{-1}(1 - 2/\beta)$ .

Коэффициент запаса  $K_{\text{и}}$  определяется нами как отношение двух значений  $m$   $K_{\text{и}} = \frac{m_1}{m_2}$ , одно из которых является среднеарифметической величиной ( $m_1$ ) наблюдаемого распределения первичной дозиметрической характеристики  $F(m_1)$  с параметрами  $m_{1g}$  и  $\beta_g$ , равной среднегодовой допустимой величине ( $m_1 = \text{СДВ}$ ), определенная по старому критерию безопасности, делитель же соответствует среднеарифметической величине распределения  $F(m_2)$  со старым стандартным геометрическим отклонением  $\beta_g$ , но сдвинутым относительно  $F(m_1)$  таким образом, что  $F(m_2 = m_1) = 1 - \frac{1}{\beta}$ , т.е. доля людей с радиационной нагрузкой больше предельно допустимой величины ( $\text{ПДВ} = m_1$ ), становится равной  $a$ . Вспомогая, что

$$\xi = \frac{\lg m - \lg m_g}{\lg \beta_g} \quad \text{получим} \quad \Phi^{-1}(1 - \frac{2}{\beta}) = \frac{\lg m_1 - \lg m_{2g}}{\lg \beta_g}$$

$$\text{или} \quad m_{2g} = m_1 \beta_g^{-\Phi^{-1}(1 - 2/\beta)}, \quad \text{но} \quad m_2 = m_{2g} \beta_g^{0,5 \lg \beta_g}$$

$$\text{и окончательно} \quad m_2 = m_1 \beta_g^{0,5 \lg \beta_g - \Phi^{-1}(1 - 2/\beta)}$$

соответствует по определению коэффициента запаса  $K_{\text{и}} = \frac{m_1}{m_2}$ , что уравнению

$$K_{\text{и}} = \frac{\beta_{\text{гд}}}{K} \quad (5)$$

\*) ВТОРИЧНЫЕ ДОЗ. ХАРАКТЕРИСТИКИ.

В настоящем докладе авторы предлагают по аналогии с существующим термином "стандартный человек" ввести понятие "стандартных" условий труда, характеризуемых коэффициентом запаса равным 5. Тогда допустимый рабочий предел загрязнения воздуха, соответствующий "стандартным" условиям, может трактоваться как новая СДЛ, которая будет жестче общепринятой в 5 раз.

#### ЛИТЕРАТУРА

1. Бадьин В.И., Саяпина Р.И. "Оценка радиационной опасности короткоживущих аэрозолей, неравновесных эманаций и грубодисперсной радиоактивной пыли". Доклад, Брюссель, 19-22/IV-71 г.
2. Алферов М.И., Бадьин В.И., Батова Э.Г., Пархоменко Г.М., Саяпина Р.Я., Саяпин Н.И., Суровежин Н.Н. "Оценка радиационной обстановки при работе с радиоактивными веществами", в кн. Радиационная медицина, Атомиздат, М., 1972, стр. 210-214.
3. Бадьин В.И., Саяпина Р.Я. "Принципы установления допустимых рабочих пределов показателей радиационной обстановки в профессиональных условиях". Доклад, Брюссель, 19-22/IV-71 г.
4. Бадьин В.И., Ермилов А.И., Маргулис У.Я., Хрущ В.Т. "О статистическом подходе при разработке математических моделей для оценки внутреннего облучения". Тезисы доклада, Международный конгресс по радиационной защите, Вашингтон, 1972 г.

ОБОСНОВАНИЕ ОСНОВНЫХ РАДИОБИОЛОГИЧЕСКИХ ХАРАКТЕРИСТИК  
ТОРИЯ-232 И ПРОДУКТОВ ЕГО РАСПАДА И ГИГИЕНИЧЕСКИХ НОРМ  
(СДН и ПДП) В ВОЗДУХЕ ПРОИЗВОДСТВЕННЫХ ПОМЕЩЕНИЙ.

П.П.Ляровский, Н.А.Павловская, Ю.Т.Капитанов,  
Л.Т.Еловская, Л.Г.Манеева.

Институт гигиены труда и профзаболеваний АМН СССР  
Москва  
С С С Р

Summary

Rats were administered, intratracheally or by inhalation, with soluble ( $^{228}\text{ThCl}_4$ ), insoluble ( $^{232}\text{ThO}_2$ ) thorium compounds together with its decay products as well as thorium-232 present in ores. Distribution and retention of Th-232 and its daughters were established for lung, lymph nodes, liver, bone, spleen, kidney and others.

The maximum admissible dose (MAD) was determined for small quantities of thorium inserted in ores.

The hygienic norms of thorium-232 and natural thorium (maximum admissible concentration) in atmospheres of workrooms were estimated.

Известие.

Радиотоксическое действие тория тесно связано с особенностями распределения радионуклидов ряда тория-232, уровнями их накопления в органах и тканях, скоростями выведения из организма.

Данные о метаболизме дочерних радиоэлементов ряда тория-232 в организме при поступлении их в органы дыхания совместно с материнским элементом или возникновении в результате распада материнского радионуклида в месте локализации последнего крайне ограничены. 1, 2, 3

Сведений о выведении тория-232 из легких при поступлении радионуклида в кристаллической решетке руд и концентратов в литературе не найдено. Данные о сочетанном действии микроколичеств тория и элементов, входящих в состав редкометаллических руд, отсутствуют.

Материалы и методы.

Исследования проведены на белых крысах, которым однократно интратрахеально вводили нерастворимое ( $^{232}\text{ThO}_2$ ) и растворимое ( $^{228}\text{ThCl}_4$ ) соединения тория совместно с дочерними радионуклидами

Оксид тория метили радиоторием.  $^{228}\text{Th}$  находился в равновесии с продуктами распада. Вводили  $4,8 \cdot 10^{-6}$  кюри  $^{232}\text{ThO}_2$  и  $3,4 \cdot 10^{-7}$  кюри  $^{228}\text{ThCl}_4$  на миллиграмм веса.

Счетный медианный диаметр (СМД)  $^{232}\text{ThO}_2$  был равен 1,03 мкм, массмедианный диаметр (ММД) - 4,9 мкм.

Хлорид радиотория вводили в изотоническом растворе при pH=7. 35% тория-228 находилось в мономерной форме.

Мышь забивали через 3, 12, 24, 72 часа и 30, 90, 180, 270 суток после введения.

В органах и тканях количественно определяли  $^{232}\text{Th}$ ,  $^{228}\text{Th}$ ,  $^{224}\text{Ra}$ ,  $^{212}\text{Pb}$ ,  $^{212}\text{Bi}$ .

### Результаты.

В результате проведенных последовательных установлено, что распределения тория-232 и его дочерних продуктов в основном зависит от растворимости соединений.

В почки и печень поступает преимущественно свинец-212.

Радий-224 накапливается в 3-4 раза меньших количествах.

При введении растворимого соединения тория в равновесии с дочерними радионуклидами, радий-224, свинец-212, висмут-212 находятся, в основном, в скелете. Максимальное количество радия-224 и свинца-212 накапливается в скелете через 24-72 часа после введения и достигает 78 и 75% соответственно от общего содержания их в организме.

Торий-228 в ранние сроки после введения находится преимущественно в легких. Постепенно количество тория-228 в легких снижается и через 90 суток содержание радионуклида в скелете становится выше, чем в легких. Спустя 270 суток после введения количество тория-228 в скелете достигает 65% и в легких - 22,8% от общего содержания радионуклида в организме. Максимальное содержание свинца-212 в печени и почках достигает 6,7 и 8,6% соответственно. Содержание радионуклидов ряда тория-232 в почках, печени, селезенке значительно ниже, чем в скелете. Наибольшее количество радия-224, находящегося в почках, равно 4,1% и в печени - 2,76% от содержания в организме. В перибронхиальных лимфатических узлах находится не более 2,68% тория-228 и 1,29% радия-224 и практически отсутствует свинец-212.

При расчетах поглощенных доз, установлении токсического действия и выборе критического органа существенное значение имеет не только общее содержание радионуклида в органе, но и его концентрация. При однократном интратрахеальном введении окиси тория наиболее высокая концентрация тория-232 и -228, радия-224 и свинца-212 почти во все сроки наблюдения отмечается в легких. Лишь спустя 3 месяцев после поступления препарата концентрация радионуклидов ряда тория в перибронхиальных лимфатических узлах становится выше, чем в легких. При поступлении в органы дыхания хлорида тория наиболее высокая удельная активность тория-228 и радия-224 почти в течение всего времени наблюдения находится в легких. Лишь через 90 суток после введения препарата удельная активность тория-228 и радия-224 в перибронхиальных лимфоузлах становится в 2,5 раза выше, чем в легких. Удельная активность свинца-212 в легких выше, чем в скелете лишь в течение первых суток после введения. Уже через 3 суток удельная активность этого изотопа в почках, печени, костной ткани становится выше, чем в легких.

Скорости выведения радионуклидов ряда тория из легких зависят от их растворимости.

При поступлении двуокиси тория совместно с дочерними радионуклидами выведение тория-232, тория-228, радия-224, свинца-212,

висмута-212 осуществляется в две стадии. Около 40% поступивших в органы дыхания радионуклидов выводится с эффективным периодом полувыведения ( $T_{эфф.}$ ) около 3-х часов, эффективный период полувыведения остальной части тория-232 и 228 равен 180-200 суток. Изменения содержания радия-224, свинца-212, висмута-212 в легких при введении двуокиси тория определяется биологическим выведением, радиоактивным распадом и накоплением в результате распада материнского радионуклида и характеризуется следующими значениями полупериодов удержания:  $T_{224Ra} = 160$  суток,  $T_{212Pb} = 160$  суток,  $T_{212Bi} = 116$  суток. (Рис. 1).

При введении в органы дыхания хлорида радиотория динамика выведения радионуклидов ряда тория-228 существенно отлична от наблюдаемой при поступлении двуокиси тория (рис. 2). Выведение тория-228 из легких осуществляется с 3-мя периодами полувыведения:  $T_{эфф.} \leq 3$  часа,  $T''_{эфф.} = 2,25$  суток и  $T'''_{эфф.} = 60$  суток. Изменения содержания радия-224 и свинца-212 в легких в ранние сроки после введения осуществляется быстрее, чем тория-228. Уже через 3 часа после введения хлорида тория содержание тория-228, радия-224, свинца-212 в легких составляет 65,7, 22,4 и 46,7% от введенного количества. Изменения содержания радия-224 спустя первые 3 часа после введения хлорида радиотория осуществляется с двумя периодами полувыведения:  $T_{1/2} = 1,18$  суток и  $T''_{1/2} = 60$  суток. Проведенные нами расчеты показали, что периоды биологического выведения радия при этом равны  $T_b = 0,93$  суток /от 3 до 72 часов/ и  $T''_b = 1,19$  суток /от 3-х до 270 суток/.

Активность легких, обусловленная свинцом-212 и висмутом-212, снижается очень быстро и через 72 часа после введения составляет всего 0,2% от введенного количества. Очень быстрый спад активности свинца-212 постепенно прекращается и в период от 3 до 90 суток активность его остается почти без изменений. Затем активность свинца-212 и висмута-212 в легких вновь начинает снижаться. В результате разных скоростей выведения радионуклидов ряда тория при поступлении хлорида тория в легких наблюдается значительное нарушение радиоактивного равновесия в ряду тория-228.

Тенденция к более быстрому выведению дочерних радионуклидов по сравнению с материнским может быть обусловлена большей скоростью перехода их в кровь. Доли радионуклидов, переходящих во внутренние органы и ткани из мест поступления соединений тория, определяются величиной всасывания их в кровь и тропностью к определенному органу.

При поступлении соединений тория в органы дыхания скорости резорбции радионуклидов ряда тория-232 в кровь зависят от химической природы элементов, растворимости соединений тория и времени с момента его поступления (табл. 1).

Таблица 1  
Скорости резорбции в кровь крыс тория-228, радия-224, свинца-212 в % от содержания в организме /сутки при интратрахеальном введении соединений тория.

Радионуклид	Время после введения					
	3ч.	24ч.	72ч.	30 сут.	90 сут.	180 сут.
	вводили двуокись тория					
Торий-232	0,1	0,01	0,001	0,001	0	0
Радий-224	2,2	15,9	40,3	4,2	4,3	3,4
Свинец-212	-	11,0	10,3	5,6	6,3	4,3
	вводили хлорид тория					
Торий-228	0,3	0,22	0,24	0,36	0,02	0
Радий-224	-	85	27	16,2	11,3	22,5
Свинец-212	-	24	27,6	29,6	31	24,6

Как следует из табл. 1 абсорбция радия-224 и свинца-212 в кровь при введении в органы дыхания как растворимых, так и нерастворимых соединений тория выше, чем материнского радионуклида. Незначительный переход тория в кровь при введении двуокиси тория может быть обусловлен крайне плохой растворимостью его в воде и тканевых жидкостях. Более высокий переход радия-224 и свинца-212, находящихся в кристаллической решетке практически нерастворимого в воде соединения двуокиси тория, по-видимому, обусловлен повышенной выщелачиваемостью этих радионуклидов.

Из крови радионуклиды ряда тория переходят преимущественно в костную ткань. При поступлении  $\text{ThO}_2$  в органы дыхания в скелет переходит до 0,12% тория-232, 17% - радия-224, 13% - свинца-212 и 7% - висмута-212. В печень и почки переходят свинец-212 и висмут-212. Радионуклидное равновесие в ряду тория-232 в скелете и внутренних органах нарушено в значительной степени как при введении нерастворимого, так и растворимого соединений тория.

Проведены исследования скорости выведения тория-232 из легких крыс в том случае, если радионуклид поступает в составе руды или концентрата. Установлено, что в случае хронического ингаляционного поступления аэрозолей, содержащих труднорастворимые редкоземельные минералы типа цирконна и лопарита, эффективный период полувыведения  $\text{Th}^{232}$  из легких белых крыс в случае лопарита был равен  $440 \pm 50$  суткам, в случае цирконна  $470 \pm 140$  суткам. При экстраполяции этих данных на человека  $T_{\text{эфф.}}$  для тория, входящего в состав труднорастворимых природных соединений, составит 4,8-5,2 года.

Исследования, проведенные на крысах, которые подвергались хроническому ингаляционному воздействию пыли цирконна и лопарита, позволили установить, что минимально эффективная доза равна ~200 бэр. Для установления величины минимально-эффективной дозы были использованы такие критерии, как средняя продолжительность жизни, оценивавшаяся по  $ET_{50}$  - эффективному времени выживаемости 50% взятых в опыт крыс, по blastomagenному эффекту и по ряду биохимических, морфологических и других показателей, характеризующих не только состояние легочной ткани, но и организма в целом. Так, производилась оценка весовых показателей легких, содержания в них растворимых и нерастворимых белков, изучалась интенсивность обмена белков легочной ткани (с помощью аминокислот лизина и глицина, меченых по  $^{14}\text{C}$ ), степень склерозирования легочной ткани, а также состояние организма в целом по ряду гематологических, иммунологических и других физиологических показателей.

Итак, полученные в эксперименте данные позволили уточнить периоды полувыведения радионуклидов ряда тория ( $T_{\text{эфф.}}$ ), доли ( $f$  возд.), доли радионуклидов от количества во всем теле ( $f_2$ ), эффективные энергии радионуклидов ряда тория ( $E_{\text{эфф.}}$ ) и др., а также установить величину ПДМД при поступлении в организм природного тория в составе труднорастворимых руд и концентратов.

На основании этих данных проведен расчет гигиенических норм тория-232, тория естественного, а также природного тория, входящего в состав труднорастворимых руд и концентратов.

Расчет проводили для частиц пыли с массмедианным аэродинамическим диаметром (ММАД) равным 0,06 мкм, 1 мкм, 10 мкм. Использовали формулу, основанную на экспоненциальном законе выведения. В результате расчета установлены следующие значения:

СДН  $\text{Th}^{232}$  от  $0,37 \cdot 10^{-14}$  до  $2,6 \cdot 10^{-14}$  юри/л

СДН  $\text{Th}$ -вст. от  $0,8 \cdot 10^{-5}$  до  $6,2 \cdot 10^{-5}$  мг/л

СДН  $\text{Th}$  в составе  $2,5 \cdot 10^{-15}$  юри/л  
нерастворимых руд

Из расчетов следует, что гигиенические нормы варьируют в

зависимости от дисперсности пыли.

В НРБ<sup>4</sup> в настоящее время принятой величиной считается среднее значение, полученное для ММAD-1 мкм и равно:

СДН  $\text{Th}^{232}$  =  $1,16 \cdot 10^{-14}$  кюри/л

СДН  $\text{Th}^{\text{вст.}}$  =  $3,7 \cdot 10^{-5}$  мг/л.

#### Литература.

1. Ballou. The translocation of thorium decay products from thorium dioxide in the dog lung. Health phys. 1970, 19, 2, 344.
2. Л.Г.Макеева, Н.А.Павловская. Выведение тория-228 и продуктов его распада из легких в эксперименте "Гигиена и санитария", 1970, 11, 42-43.
3. Н.А.Павловская, Л.Г.Макеева. Поведение радионуклидов ториевого ряда ( $\text{Th}^{232}$ ,  $\text{Th}^{228}$ ,  $\text{Ra}^{224}$ ,  $\text{Rb}^{212}$ ,  $\text{Bi}^{212}$ ) в организме крыс при интратрахеальном поступлении двуокси тория. Гигиена и санитария", 1972, 2, 65-68.
4. Нормы радиационной безопасности НРБ-69. Москва, Атомиздат. 1972, стр. 65 и 80.



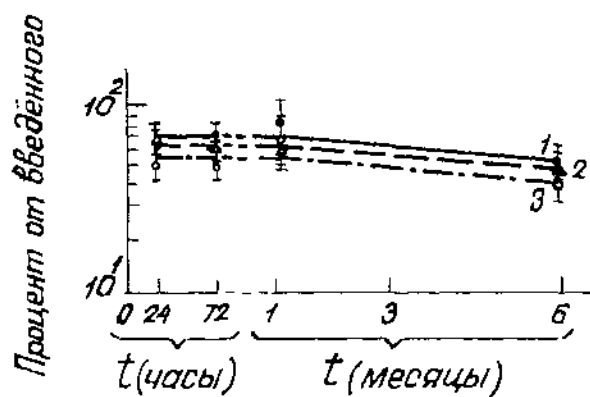


Рис. 1. Удержание тория-228(1), радия-224(2) и свинца-212(3) в лёгких крыс при интратрахеальном поступлении двуокиси тория.

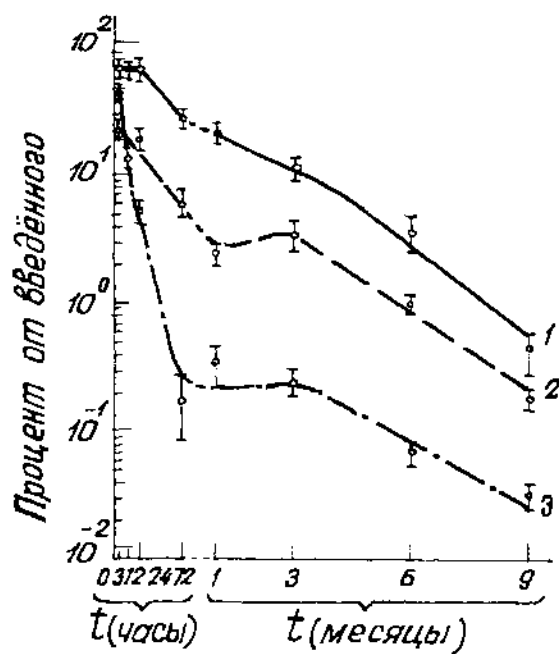


Рис. 2. Удержание тория-228(1), радия-224, свинца-212(3) в лёгких крыс при интратрахеальном поступлении хлорида радиотория.

## INHALATION STUDY OF A SUBMICRON SIZE LEAD-212 AEROSOL

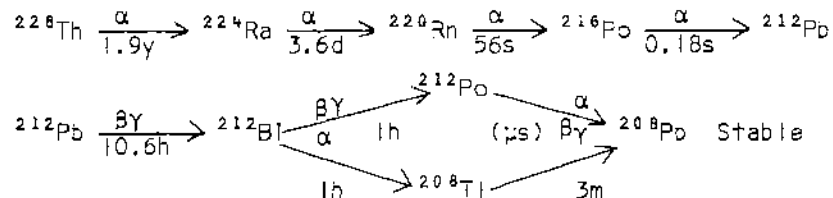
Andrea Bianco\*, F. R. Gibb and P. E. Morrow  
Department of Radiation Biology and Biophysics  
University of Rochester School of Medicine and Dentistry  
Rochester, New York 14642

### Abstract

Dogs (purebred female beagles ~ 10 kg) were briefly exposed to a submicron aerosol of  $^{212}\text{Pb}$  having an effective diffusion diameter between .05-120  $\text{\AA}$ , ( $\sigma_g$  1.2) produced in a chamber of new design. The resulting lead burden was followed for up to 72 hours postexposure by external counting of the 240 keV gamma ray. The concentration of  $^{212}\text{Pb}$  in the arterial and mixed venous blood were regularly measured and no significant A-V difference could be found. The blood concentration data were used to differentiate between the  $^{212}\text{Pb}$  level actually in the lungs and that within the thoracic blood volume. This approach was facilitated by the intravenous injection of  $^{212}\text{Pb}$  so that an increment in  $^{212}\text{Pb}$  concentration in the blood could be correlated with the increment in thoracic radioactivity. Seven canine studies indicated that the submicron aerosol of  $^{212}\text{Pb}$  was cleared from the lungs with a single exponential process having a biological half-time of ~ 12 hours. This mean value corresponds closely to the biological half-times in human lungs reported by other investigators for  $^{212}\text{Pb}$  and  $^{210}\text{Pb}$  labelled aerosols of diverse physical and chemical forms, viz. 8-12 hours, suggesting that the clearance value can be generalized for atmospheric lead. Evidence is presented that the blood is a principal clearance pathway and that external assessment of radioactive lead in the thorax must also consider the lead content of the thoracic wall structures.

### Introduction

An aerosol of  $^{212}\text{Pb}$  is formed by the decay of airborne  $^{220}\text{Rn}$ , an alpha-emitter having a half-time of 56 seconds. This aerosol has been the subject of several recent investigations because: (a) the aerosol produced is of submicron dimensions and provides an interesting physical system for investigating the production of aerosol by radioactive emissions<sup>1-5</sup>, and (b) of its short half-life, and physical and chemical nature, which make it of pertinence and value for biological studies including those in man<sup>6-9</sup>. The radioactive schema for the production of  $^{212}\text{Pb}$  and its decay are as follows:



\*on leave of absence from: Lab. Radiobiologia Animale, C.S.N.  
Casaccia, C.N.E.N. 00060 Roma, Italy

## Methods and Materials

The experimental animals used in the study were purebred, female, beagle dogs of approximately 10 kg body weight. Individual exposures to  $^{212}\text{Pb}$  aerosols were accomplished through an endotracheal tube connected to a special aerosol chamber (Figure 1) while the canine subject was anesthetized with pentobarbital sodium (ca 28 mg/kg i.v.).

### $^{212}\text{Pb}$ AEROSOL GENERATION SYSTEM

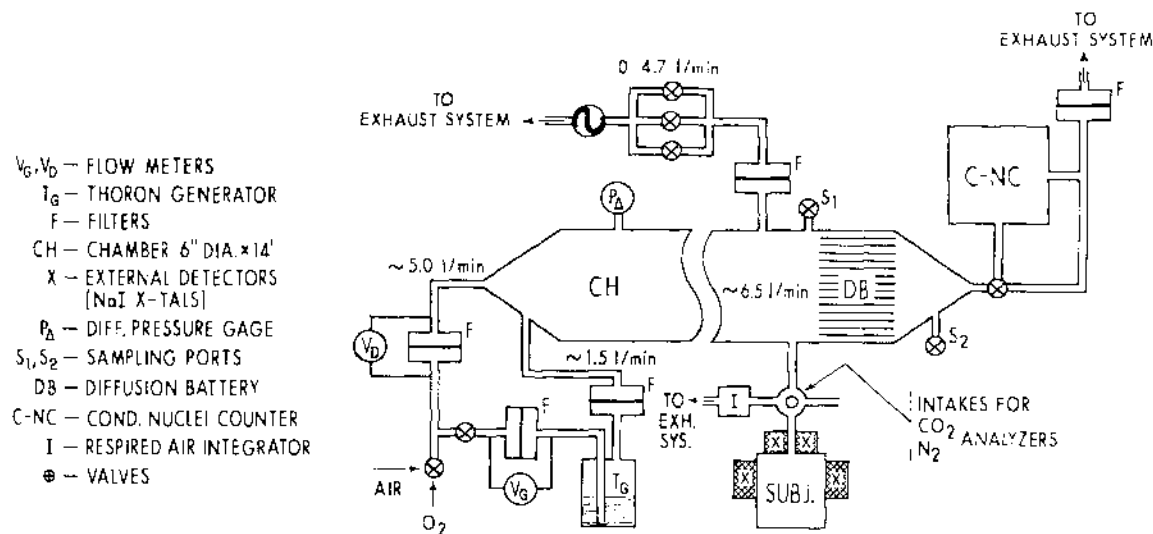


Figure 1. Schematic of the Aerosol Generation and Exposure System.

The linear chamber, CH, is complemented by four sub-units: a thoron source and aerosol generation unit (left); an aerosol analysis unit (right) consisting of sampling ports  $S_1$ ,  $S_2$ , the diffusion battery, DB, the condensation nuclei counter, C-NC, and a variable exhaust to control the flow through DB; the exposure section (rt. center) with its associated physiological monitors and the exhaust (right) for the disposal of unused aerosol.

The  $^{212}\text{Pb}$  aerosol was produced as the result of passing a clean air stream (1.5 l/min) through a thoriumnitrate solution enriched with  $^{228}\text{Th}$  and extracting the  $^{220}\text{Rn}$  gas. The  $^{220}\text{Rn}$ -air mixture was then passed through a coarse filter and a molecular filter before being diluted further by filtered clean air (5.0 l/min). The resulting radon mixture was conducted through a linear chamber 15 cm diameter by  $\sim 430$  cm long. At this point, connections were located which provided for (a) exposing the subject; (b) analyzing the aerosol; and (c) exhausting the unused aerosol. The volumetric flow in the chamber determined the extent to which the  $^{220}\text{Rn}$  decayed to  $^{212}\text{Pb}$  at the point of use, i.e. determined the transit time. At 6.5 l/min, the chamber transit time was 10 minutes which is equivalent to approximately 11 physical half-lives for the radon gas. This implies that at the point of use, the  $^{220}\text{Rn}$  had undergone 99.95% decay.

In the linear-chamber system, the aerosol can be produced continuously under highly controlled conditions so that the characteristics of the aerosol

are quite constant and highly reproducible from day to day. The dog exposure studies utilized a submicron aerosol with the following average characteristics: an effective diffusion diameter between 105-120 Å; geometric standard deviation  $\sim 1.2$ ; a mean  $^{212}\text{Pb}$  activity concentration of 0.7  $\mu\text{Ci } ^{212}\text{Pb}/\text{l}$  air corresponding to slightly less than 1 picogram of Pb/l. The exposures were generally of about 5 minutes duration and the initial respiratory burdens in the dog lungs averaged  $\sim 5 \mu\text{Ci}$  of  $^{212}\text{Pb}$ .

The analyses of the  $^{212}\text{Pb}$  aerosol depended upon the determination of the mass and numerical penetration values in a diffusion battery (Fig 1). Methods of estimating the distributional parameters of a diffusive, heterodisperse aerosol had been described by Sinclair<sup>10</sup>, Nurse and Mercer<sup>7</sup>, and by Fuchs<sup>11</sup>.

After the brief aerosol exposure, each dog was positioned in a body holder and placed within a shielded thoracic counting chamber<sup>12</sup>. Two 3.5" by 6.0" scintillation counters (NaI) were collimated so as to view the lateral aspects of the dog's chest between the first and fifth intercostal spaces thereby avoiding activity from subdiaphragmatic structures and the subglottic airways. The outputs of the two counters were added in the multichannel analyzer and obtained by high-speed digital print-out. Analyzer channels corresponding to the energy range 210-270 keV were used for the  $^{212}\text{Pb}$  measurements. For  $^{212}\text{Bi}$ , the 560-620 keV range was utilized. Dogs were serially measured by external counting for as long as was practicable, normally 2 days postexposure.

The contribution of the  $^{212}\text{Bi}$  daughter activity to the *in vivo* and *in vitro* measurements of  $^{212}\text{Pb}$  was determined by studying freshly-acquired filter samples of  $^{212}\text{Pb}$  either as a distributed source, e.g. in the lung fields of a beagle phantom, or in the same geometry as other samples were analyzed, i.e. blood. Serial blood samples were required in order to correct the thoracic counts for the contribution of  $^{212}\text{Pb}$  which was bloodborne. A sealed thorium standard (in equilibrium with its daughter products) was also utilized in the correction and calibration procedures. The  $^{212}\text{Pb}:^{212}\text{Bi}$  ratios were determined as a function of time and evaluated in terms of the excessive  $^{212}\text{Pb}$  counts measured. The appropriate relationships were then used to account for the daughter contribution to all experimentally determined  $^{212}\text{Pb}$  activities. The counting efficiency for measuring  $^{212}\text{Pb}$  within the canine thorax was found to be approximately 0.7% whereas for  $^{212}\text{Pb}$  in blood samples, the efficiency was  $\sim 10\%$  for the geometry used.

It was known from previous experiments<sup>6,7,9</sup> that external measurements of the chest soon after an exposure to a  $^{212}\text{Pb}$  aerosol depicted the combined processes of lead build-up in the blood and of lead removal from the lungs. Consequently, a knowledge of the blood build-up rate and a "blood correction factor" were required in order to reveal the actual time-course of  $^{212}\text{Pb}$  removal from the lungs. This "correction" procedure<sup>7</sup> in essence, assumes that at least during most of the lung clearance (0-48 hr), there is no other important extrapulmonary compartment within the thoracic field except the thoracic blood volume. This point will be dealt with more fully in the Discussion section of this paper.

### Results

In seven dog studies, measurements of  $^{212}\text{Pb}$  activity as directly obtained by external counting during the first 24-48 hr postexposure gave an apparent effective half-life for the thorax of 8.3 hr implying a simple exponential clearance for lead with a biological half-time of approximately 40 hours ( $\lambda = .0173$ ). However, when the raw activity data were corrected for the contribution of daughter activity, viz.  $^{212}\text{Bi}$ , then the average thoracic clearance half-time (effective) and the estimate for the biological

half-time decreased

(Figure 2).

When the daughter-corrected, thoracic activity of  $^{212}\text{Pb}$  was also adjusted for the contribution of the bloodborne  $^{212}\text{Pb}$ , the average, effective clearance half-time for the thorax further decreased to 5.6 hours and the corresponding biological clearance half-time decreased to 11.9 hours ( $\lambda = 0.054$ ). In Figure 3, therefore, the average curve can be depicted by the simple exponential equation:  $R = Ae^{-\lambda t}$  where R is the fractional activity retained, A the initial activity,  $\lambda$ , the decay coefficient, is equal to 0.054, and t is the time in hours.

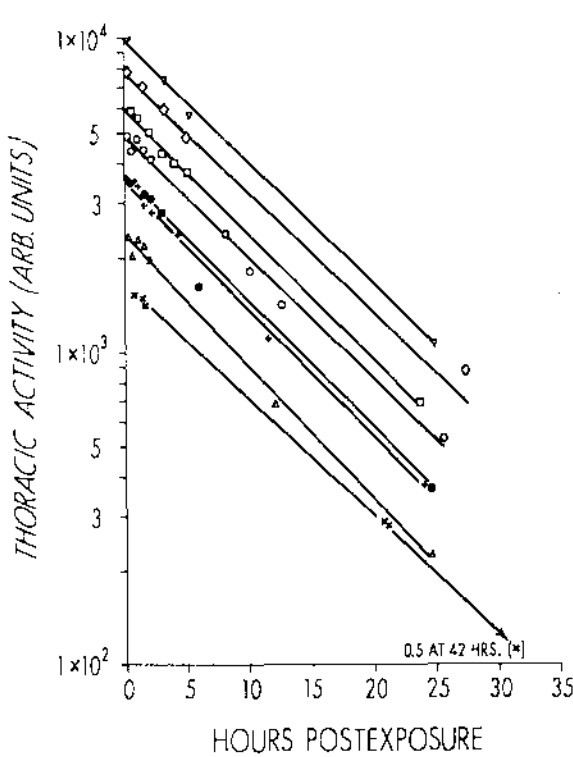


Figure 2. Graphic Summary of Thoracic Retention Data for  $^{212}\text{Pb}$ .

These data are corrected for the Compton scattering in the  $^{212}\text{Pb}$  energy spectrum due to  $^{212}\text{Bi}$  activity. The intercepts of the curves were adjusted so as to avoid overlap in their presentation.

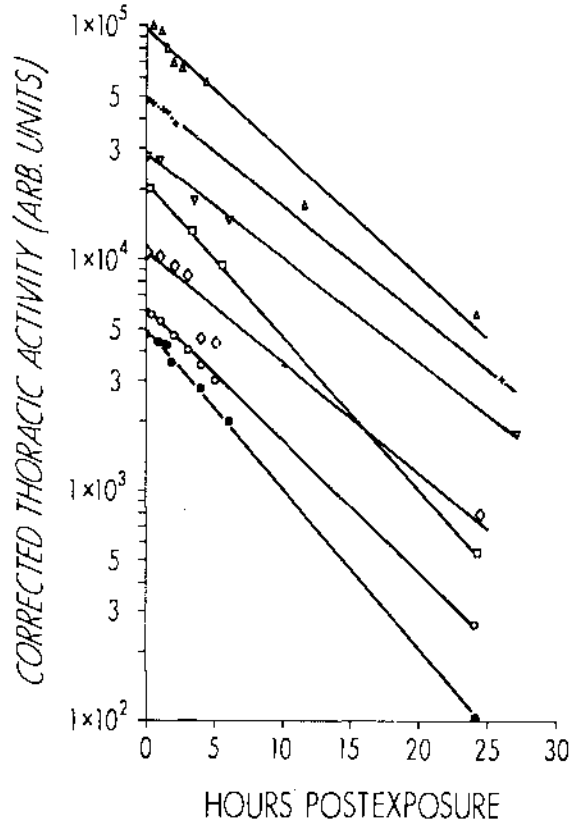


Figure 3. Corrected Thoracic Retention Data.

These curves have been corrected for bloodborne  $^{212}\text{Pb}$  by the procedure described in the text. For this graphic presentation, the data have been arbitrarily adjusted to reduce overlaps.

Whereas, all of the clearance data can be considered exponential, the range of half-time values in the daughter-corrected data was 7.0-7.8 hours; in the data adjusted for bloodborne radioactivity, the variation increased, i.e. 4.3-6.9 hours, indicating that the blood correction procedure was largely responsible for this.

Measurements of bloodborne activity in the mixed venous and arterial blood revealed no significant  $^{212}\text{Pb}$  concentration gradient during the first 24 hours. An A-V difference was suspected since there had been a report of an early high renal clearance of  $^{212}\text{Pb}$  after intravenous administration<sup>8</sup> which was soon

dissipated presumably due to the shift of  $^{212}\text{Pb}$  from the plasma to red blood cells.

The blood build-up curve for  $^{212}\text{Pb}$  peaked at approximately 10 hours post-exposure. In terms of stable or decay-corrected lead, there was no significant decrease in blood lead concentration below this peak concentration during the first 24 hours. The build-up phase in the blood appeared to be exponential and have a half-time of 4.7 hours. This is substantially faster than the rate of  $^{212}\text{Pb}$  disappearance from the lungs. This indicated some inconsistency in the data if it is assumed that all (>95%) of the lead clearance is directly into the blood as did Hursh and Mercer<sup>7</sup>. This point is under further study.

One dog was given a large initial  $^{212}\text{Pb}$  burden with the expectation that the thoracic clearance  $^{212}\text{Pb}$  could be followed after 24 hours more accurately. At 96 hours postexposure, the thoracic level was found to be 0.18% of the initial value and far in excess of that commensurate with 11.9 hours biological half-time. The activity in the blood measured at this time was insufficient to account for more than 30% of the thoracic activity, but since the blood activity was so low, we considered this value highly speculative. Consequently the animal was sacrificed and the lungs, pulmonary nodes and blood corresponding to the blood volume of the thoracic viscera, were removed. The chest cavity was filled with a mass of wet, absorbent paper corresponding to the weight of the tissues removed and then the thorax was recounted as it was in the intact animal. Ninety-four percent of the terminal thoracic activity was found associated with the eviscerated thorax. We conclude, therefore, that the retained  $^{212}\text{Pb}$  activity was based entirely upon the lead content of the blood and the thoracic wall structures and no lead retention could be assigned to the lungs and pulmonary lymphatics. This conclusion is entirely consistent with the biological half-time assigned to the lungs.

#### Discussion

Thoracic measurements in all dogs were subjected to a correction procedure to account for the bloodborne  $^{212}\text{Pb}$ . Hursh and Mercer<sup>7</sup> first reported this procedure, which entails the intravenous injection of a freshly-acquired  $^{212}\text{Pb}$  sample and to determine the increments in systemic blood and thoracic radioactivities post-injection. Our preparation was made by sonification of a  $^{212}\text{Pb}$  filter sample in isotonic saline followed by the injection of ~ 2 ml into a leg vein. The ratio of the increments in  $^{212}\text{Pb}$  activity constitutes a correction factor which is derived from both geometric and blood volume considerations.

In this study, the initial thoracic measurements (0-1 hr) and the subsequent blood-corrected measurements were taken as pertaining to the removal of  $^{212}\text{Pb}$  from the lung parenchyma. Certain corroborative evidence was cited. From other studies<sup>6,7,9</sup> there is evidence of the removal of lead from the blood by biliary and urinary excretion and by redistribution to other tissues, especially skeletal. These pharmacodynamic features of blood lead are not easily studied with the  $^{212}\text{Pb}$  isotope, but the available data indicate a very slow clearance rate for lead in the blood relative to that in the lungs.

The biological half-time of 11.9 hours for lead in the lungs is in close agreement to the 10.5-11.5 hr values reported by Hursh and Mercer<sup>7</sup> for human subjects following single exposures to 0.02-0.23  $\mu\text{m}$  AMAD aerosols. The clearance value is also similar to those reported by Booker, et al<sup>9</sup> for  $^{212}\text{Pb}$  labelled lead vapor in man, viz. ~ 10 hr biological half-time and by Jacob<sup>13</sup> for  $^{212}\text{Pb}$  attached to atmospheric dust particles giving a biological half-time of 8 hours. Dog studies by Gibb and Hursh (unpublished), utilizing  $^{210}\text{Pb}$  and involving ~ 0.1  $\mu\text{g}$  burdens of stable lead in the form of a basic carbonate (0.2-0.6  $\mu\text{m}$  AMAD), also revealed an initial lung clearance rate (after a blood-borne lead correction was applied) of 12.2 hours. The possibility that the

external counting data in these investigations were complicated by tracheo-bronchial clearance is discounted by the rate of  $^{210}\text{Pb}$  build-up in the blood which was nearly identical to that found in this study.

These several studies with highly different aerosols of lead, both physically and chemically, give remarkably similar results, so that it appears practical to generalize the clearance of lead from the lungs of man following exposure to various atmospheric forms. In possible disagreement to this viewpoint are the data of Morken<sup>14</sup> who reported a five minute biological half-time for  $^{214}\text{Pb}$  on atmospheric dust from mouse lungs and the data of Albert and Arnett, who studied  $^{212}\text{Pb}$  attached to kaolin particles<sup>15</sup> and reported a 60 hour biological clearance half-time. It should be noted, however, that Albert and Arnett failed to account for the bloodborne  $^{212}\text{Pb}$ .

#### References

1. Chamberlain, A.C., W.J. Megaw and R.D. Wiffen. Role of Condensation Nuclei as Carriers of Radioactive Particles. *Geofisica Pura e Applicata* **36**, 233-242, 1957.
2. Billard, F., J. Bricard, M. Cabane and G. Magelaine. Évolution des Noyaux de Condensation qui se forment par Radiclyse des Impuretés Gazeuses de l'Air. *C R Acad Sc Paris* **265**, 1376-1379 Series B, 1967.
3. McGreevy, G. Towards the Production of a Reproducible Aerosol by Radiolytic Methods. *Pure Appl Geophys* **71**, 190-197, 1968.
4. Huertas, M. L., A.M. Marty, J. Fontan and D. Blanc. Measurement of the Mobility and the Diffusion Coefficient of Ultrafine Radioactive Particles in the Air. *J Atmos Sci* **26**, 274-280, 1969.
5. Duggan, M.J. Some Aspects of the Hazard from Airborne Thoron and its Daughter Products. *Health Phys* **24**, 301-310, 1973.
6. Hursh, J.B., A. Schraub, E. L. Sattler and H.P. Hofmann. Fate of  $^{212}\text{Pb}$  Inhaled by Human Subjects. *Health Phys* **16**, 257-267, 1969.
7. Hursh, J.B. and T.T. Mercer. Measurement of  $^{212}\text{Pb}$  Loss Rate from Human Lungs. *J Appl Physiol* **28**, 268-274, 1970.
8. Stover, B.J.  $\text{Pb}^{212}$  (Th B) Tracer Studies in Adult Beagle Dogs. *Proc Soc Exp Biol Med* **100**, 269-272, 1959.
9. Bocker, D.V., A.C. Chamberlain, C. Newmon, and A.N.B. Stott. Uptake of Radioactive Lead Following Inhalation and Injection. *Br J Radiol* **42**, 457-466, 1969.
10. Sinclair, J. A Portable Diffusion Battery. *Am Ind Hyg Assoc J* **33**, 729-735, 1972.
11. Fuchs, N.A. Some New Methods and Devices for Aerosol Studies. Assessment of Airborne Particles: Fundamentals, Applications, and Implications to Inhalation Toxicity, T.T. Mercer, P.E. Morrow and W. Stöber, editors, Charles C. Thomas, Springfield, Illinois, 1972, pp. 200-210.
12. Morrow, P.E., F.R. Gibb and L.J. Leach. The Clearance of Uranium Dioxide Dust from the Lungs Following Single and Multiple Inhalation Exposures. *Health Phys* **12**, 1217-1223, 1966.
13. Jacobl, W., K. Aurand and A. Schraub. Advances in Radiobiology, Oliver and Boyd, Edinburgh, Scotland, p. 310, 1957.
14. Morken, D. A. and J.K. Scott. The Effects on Mice of Continual Exposure to Radon and its Decay Products on Dust. University of Rochester Atomic Energy Project Report #UR-659, 1966.
15. Albert, R.E. and L.C. Arnett. Clearance of Radioactive Dust from the Human Lung. *AMA Arch Ind Health* **12**, 99-106, 1955.

*This paper is based on work performed under contract with the U.S. Atomic Energy Commission at the University of Rochester Atomic Energy Project and has been assigned Report No. UR-3490-334.*

THE EFFECT OF BREATH-HOLDING ON THE  
DEPOSITION OF HALF-MICRON AEROSOL  
PARTICLES IN THE HUMAN LUNG

M.C. Subba Ramu  
Air Monitoring Section  
Bhabha Atomic Research Centre  
Bombay-400085, India

ABSTRACT

Measurements of deposition of particles in the human lung during breath-holding can be made use of to calculate the diameter of the tubular passageways of the respiratory tract in life. When the inspired volume is sufficiently great for most of the aerosol to penetrate beyond the anatomical dead space, the rate of deposition during the breath-holding can be regarded as an index of the average diameter of the alveolar airways. Studies of the effect of breath-holding on deposition and recoveries of  $0.5 \mu\text{m}$  particles of unit density in different lung volumes are described in this paper. Based on the results of steady-state breathing experiments, the average diameter of the airways in the alveolar region of the lung has been found to be 0.74 mm.

INTRODUCTION

Studies on the deposition of particles in human lungs show that it is possible pauses may occur in between inhalation and exhalation and also between successive breaths. The duration of these pauses should have certain effects on deposition of particles in the lung. According to Altshuler<sup>1</sup>, breath-holding experiments are useful in assessing an average dimension of the alveolar spatial units in life. When the inspired volume is sufficiently large for most of the aerosol to penetrate beyond the anatomical dead space, the rate of deposition during breath-holding can be regarded as an index of the average dimension of the alveolar space in life.

This paper gives an account of the studies made to find the effect of breath-holding on the deposition of  $0.5 \mu\text{m}$  spherical particles of unit density during the steady-state and single-breath experiments.

EXPERIMENTAL TECHNIQUES AND RESULTS

The apparatus used for the measurement of deposition is the one used by Davies, Heyder and Subba Ramu<sup>2</sup>. Three different techniques were employed for measuring the effect of breath-holding on aerosol deposition. In the first case, the subject breathed the aerosol till steady-state condition was reached, with a pause for a known period of time between successive



breaths. Table 1 gives the results of the measurements for these two cases. Average breath-holding times ( $t$ ) are given along with the tidal volumes (TV) expiratory reserve volumes (ERV), breathing frequencies ( $f$ ) and the fraction deposited ( $D$ ). It can be seen that the deposition increased by about 34% for an average breath-holding time of 1.07 secs in the first case whereas there was negligibly small difference in the second case.

Table-1

The effect of breath-holding on deposition during steady-state breathing

TV ( $\text{Cm}^3$ )	ERV ( $\text{Cm}^3$ )	$f$ (Breaths/min)	$t$ (secs)	(1-D) (Average)	Remarks
678	1260	16.8	0	0.906	Normal; No pause
678	1260	16.8	1.07	0.874	Pause between inhalation and exhalation
696	1200	17.8	0	0.905	Normal; No pause
696	1200	17.8	1.36	0.913	Pause between successive breaths

In the third case, the subject inhaled about  $600 \text{ cm}^3$  of aerosol-laden air, held the breath for a known period of time and exhaled more or less the same volume of air. This was followed by inhalation of clean air and maximal exhalation. The total recovery ( $R$ ), recovery in tidal volume ( $R_t$ ) and the recovery ( $R_r/1-R_t$ ) of aerosol particles are calculated separately as shown in table 2.  $R_r$  is the ratio of the number of particles recovered in the

Table-2

The effect of breath-holding on the recovery of  $0.5 \mu\text{m}$  particles in the exhaled air (single-breath experiments)

TV ( $\text{Cm}^3$ )	ERV ( $\text{Cm}^3$ )	$f$ (breaths/min)	$t$ (secs)	$R$	$R_t$	$R_r/1-R_t$
660	980	16.5	0.0	0.96	0.90	0.72
690	820	15.4	3.0	0.86	0.76	0.54
665	941	14.4	5.7	0.79	0.67	0.42
615	950	14.2	7.6	0.73	0.62	0.33
585	1010	17.0	15.3	0.57	0.46	0.18
635	1170	15.3	24.0	0.45	0.36	0.13

reserve air to that in the inhaled air. ( $R_r/1-R_t$ ) is the recovery of particles

in the reserve air expressed as a fraction of particles lost from the tidal air into the reserve air. Figure 1 gives the values of  $R$ ,  $R_t$  and  $(R_r/1-R_t)$  for different breath-holding times.

The build-up of aerosol particles in the lung before steady-state is reached is shown in figure 2 for normal breathing and breathing with pauses. Breath-holding seems to have no effect on the time needed for attaining the steady-state condition. The plot of  $R_n/R_{AV}$  against the number of breaths shows that, in all cases, steady-state is reached in four breaths.  $R_n$  is given by

$$R_n = E_n/I_n \quad \dots(1)$$

where  $I_n$  is the amount of aerosol inhaled in the  $n^{\text{th}}$  breath during build-up, and  $E_n$  the amount of aerosol exhaled in the same breath.  $R_{AV}$  is the average fraction recovered during steady-state breathing.

#### CALCULATION OF PASSAGEWAY DIAMETER

Landahl<sup>3</sup> gives the following equation for the fraction deposited ( $D$ ) in the human lung during breath-holding time 't':

$$-R_p \ln(1-D) = (1.8 \times 10^5 d^2 Ct) + (4 \times 10^{-6} \sqrt{Ct/d}) \quad \dots 2$$

where,

$d$  = diameter of the particle (cm)

$R_p$  = radius of the tubular passage (cm)

and

$C = 1+1.8 \times 10^{-5}/d$ , a size correction factor.

Table 3 gives the average diameters of the tidal volume ( $D_{TV}$ ) and the expiratory reserve volume ( $D_{ERV}$ ), calculated using equation (2) and table 2.  $D(TV+ERV)$  is obtained from the results of the total recoveries for different breath-holding periods. The diameter of the tidal volume varies from 0.33 to 0.38 mm and that of the expiratory reserve volume from 0.16 to 0.20 mm.

The diameter of the airways in the alveolated region has been calculated using the deposition measured during steady-state breathing. The deposition increased from 9.4%, when there was no pause, to 12.6% when the pause between inhalation and exhalation was, on an average, 1.07 secs (Table 1). Using equation (2), the diameter of the passageway was calculated to be 0.74 mm. The diameter of the passageway calculated in the case of steady-state breathing works out to be more than that calculated in the case of the single-breath experiments.

Table-3

Average diameters of the tidal( $D_{TV}$ ) and the expiratory reserve volumes ( $D_{ERV}$ ) in the lung, calculated using equation (2) and table (2)

t (secs)	TV (cm <sup>3</sup> )	$D_{TV}$ (mm)	ERV (cm <sup>3</sup> )	$D_{ERV}$ (mm)	(TV+ERV)	$D(TV+ERV)$ (mm)	$D_{TV}+D_{ERV}$
3.0	690	0.33	820	0.19	1510	0.51	0.52
5.7	665	0.34	941	0.19	1606	0.52	0.53
7.6	615	0.34	950	0.16	1565	0.50	0.50
15.3	585	0.35	1010	0.17	1595	0.46	0.52
24.0	635	0.38	1170	0.20	1805	0.48	0.58
Average	638	0.35	980	0.18	1618	0.50	0.53

## DISCUSSION

Experiments using 0.5  $\mu\text{m}$  particles, conducted by Palmes, Altshuler and Nelson<sup>4</sup>, showed that the passageway diameter varied from 0.3 to 0.4 mms. The results of similar experiments given in table 3 show that the diameter is about 0.5 mms. The diameter of the passageways calculated in the case of steady-state breathing works out to be 0.74 mms (Table 1). The difference in the diameters obtained for these two cases indicate that the most of the aerosol particles lost from the tidal air reach the walls of the lung after passing through the expiratory reserve volume and the residual volume.

Let us consider that the shape of the reserve volume is same as that of residual volume, then the radius (0.09 mms) and the corresponding expiratory reserve volume (980  $\text{cm}^3$ ) are related by

$$0.09 \propto \sqrt[3]{980} \quad \dots(3)$$

and if the radius of the residual volume is denoted by  $r_{RV}$  and the residual volume is 2040  $\text{cm}^3$  which is the value for the subject under consideration, then we have,

$$r_{RV} \propto \sqrt[3]{2040} \quad \dots(4)$$

Dividing (4) by (3), we get

$$r_{RV} = 0.09 \times \sqrt[3]{2040/980}$$

Thus,  $r_{RV} = 0.12$  mms

Therefore the diameter of the residual volume is 0.24 mms.

Now if we add the diameters of the tidal, reserve and residual volumes, we get 0.76 mms which is in close agreement with the diameter (0.74 mms) calculated for the steady-state breathing experiment.

The calculated diameter (0.74 mms) of the alveolar airways works out to be greater than the values given by weibel<sup>5</sup> in his regular dichotomy model of the human lung at 3/4 maximum inflation. If the diameters of the airways are large, it means it takes a longer time for the particles to travel from the main stream towards the walls and if, meanwhile, the subject exhales out, the fraction deposited would be the lowest and in some cases, all the 0.5  $\mu\text{m}$  particles inhaled would be exhaled out as in the case of single-breath experiments.<sup>6</sup>

The theoretical curves (figure 1) calculated by equation (2) do not vary much from the experimental ones, showing that the relationship given by Landahl<sup>7</sup> can be used for obtaining the diameter of the airways in the human lung using 0.5  $\mu\text{m}$  particles as tracers. Better agreement between the theory and experiments would perhaps be possible if a correction is made to account for the inertial movement caused during the breathing cycle in the region of functional residual volume. Another discrepancy pointed out by Palmes, Altshuler and Nelson<sup>4</sup> is the relatively small contribution of the Brownian motion term to the calculated deposition. As has been pointed out by Landahl<sup>7</sup> the difference becomes serious only for particles 0.1  $\mu\text{m}$  or less.

An important implication of these studies is that the dose delivered to the tissues by breathing radioactive particles of different sizes reduces considerably if the airway diameter, especially in the alveolated region of

the lung, is larger. This would result in appreciable changes in the m.p.l. values to the advantage of the progress of nuclear industry. The method of measuring the diameter of the airways by breath-holding technique is also useful for diagnostic purposes. Persons suffering from constrictive diseases will have a higher deposition of particles in the lung. Also the breath-holding technique would show the extent of constriction when compared with the airway diameter of a normal lung.

#### ACKNOWLEDGEMENTS

The author is grateful to Dr. C.N. Davies for his kind guidance in carrying out the studies in the London School of Hygiene and Tropical Medicine, supported by a fellowship from IAEA. The author is also thankful to Dr. K.G. Vohra for his keen interest and kind encouragements.

#### REFERENCES

1. Altshuler, B. 1969. "Behaviour of Airborne particles in the Respiratory Tract", In Circulatory and Respiratory Mass Transport (ed. by Wolstenholme, G.E.W. and Knight, J), p.215.
2. Davies, C.N., Heyder, J. and Subba Ramu, M.C. 1972. "Breathing of Half-micron Aeroecols I. Experimental", J. Appl. Physiol. 32, p. 591.
3. Landahl, H.D.1950. "On the Removal of Air-Borne Droplets from the Human Respiratory Tract: I. The Lung". Bull. Math. Biophys. 12. p. 43.
4. Palmes, E.D., Altshuler, B. and Nelson, N. 1967. "Deposition of Aerosols in the Human Respiratory Tract during Breath-Holding", In Inhaled particles and Vapours II (ed. by C.N. Davies), p. 339.
5. Weibel, E.R. 1963. "Morphometry of the Human Lung", Academic Press, New York.
6. Subba Ramu, M.C. 1972. "On the Physiological Implications of Aerosol Inhalation", Paper Presented in the National Symposium on Environmental Pollution, October 28-30, Bombay.
7. Landahl, H.D. 1963. "Note on the Removal of Air-Borne Particles by the Human Respiratory Tract with Particular Reference to the Role of Diffusion". Bull. Math. Biophys. 25, p.29.

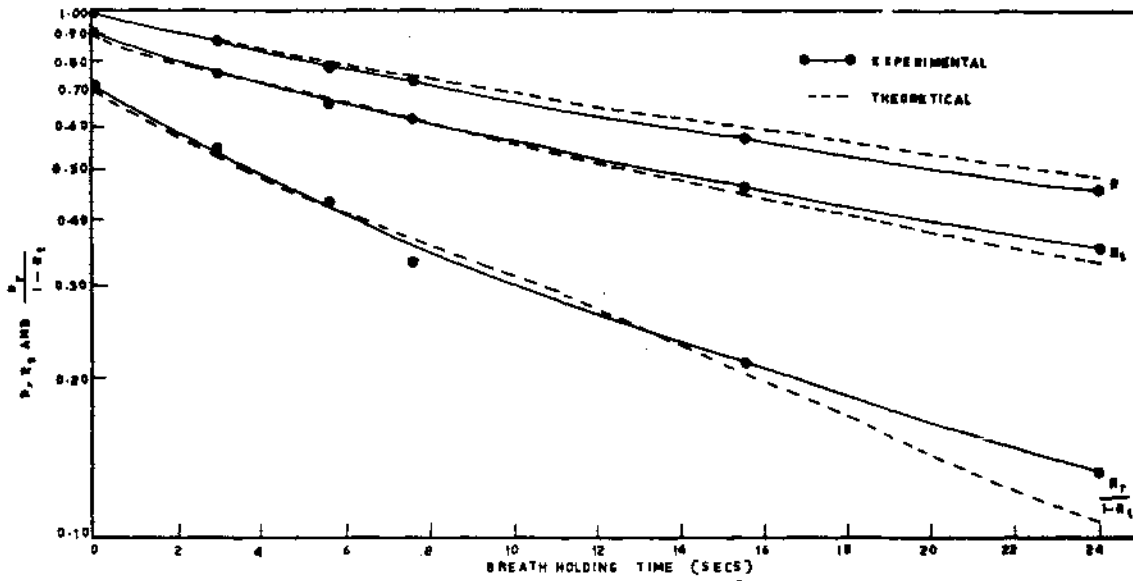


FIGURE:1. THE EFFECT OF BREATH HOLDING ON  $R$ ,  $R_t$  AND  $\frac{R_t}{1-R_t}$  (SINGLE BREATH EXPERIMENTS)

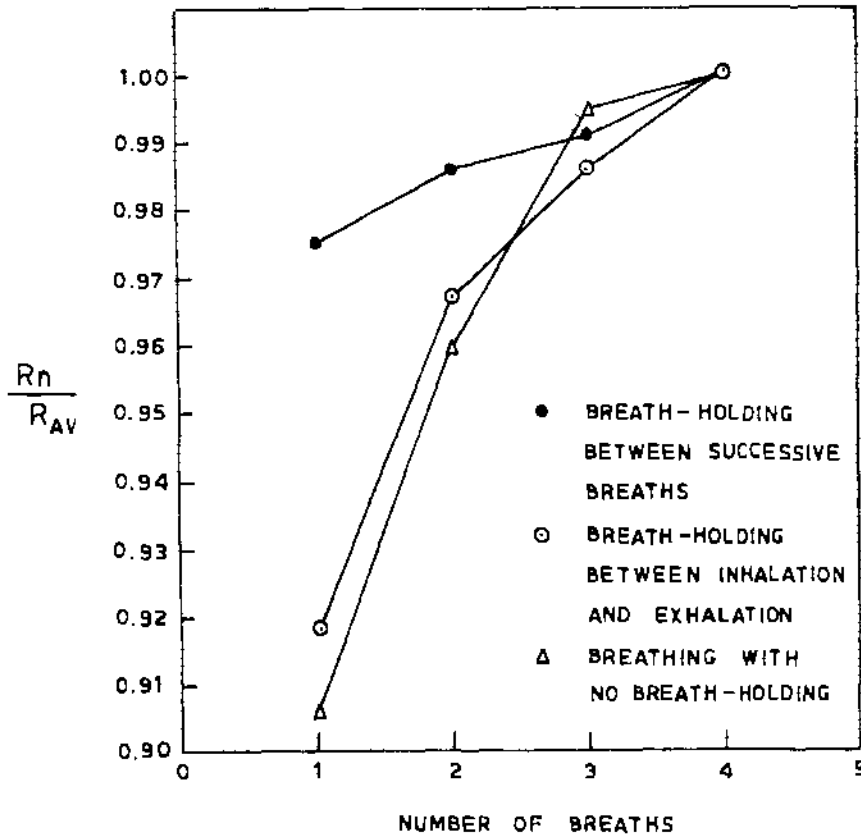


FIGURE:2. BUILD-UP OF AEROSOL PARTICLES IN THE LUNG DURING THE STEADY-STATE BREATHING.

HASL CYCLONE AS AN INSTRUMENT FOR MEASURING  
AEROSOL PARAMETERS FOR NEW LUNG MODEL

P.Kotrappa, S.K.Dua, D.P.Bhanti and P.P.Joshi  
Health Physics Division  
Bhabha Atomic Research Centre  
Bombay-400085, India

Abstract

Recently proposed ICRP Lung Model stipulates that a knowledge of an aerosol parameter, activity median aerodynamic diameter (AMAD) is essential to predict the fractional depositions in various parts of the respiratory tract. It is shown that this method of determining AMAD does not reflect significant error in the estimation of pulmonary deposition over a size range of 1.5 to 3.0  $\mu\text{m}$ . Results of a large number of measurements made in Trombay Fuel Reprocessing Plant are reported. Results compare well with similar measurements made by other techniques such as Centripeter and Autoradiography. Significance of the measured AMAD is discussed with respect to maximum permissible concentrations (MPC) in air.

Introduction

Currently used MPC<sup>1</sup> values in air for radioactive nuclides assume that 25% of whatever inhaled goes to pulmonary region of the lung and 12% undergoes long term retention for insoluble aerosols. This is assumed with no regard to the particle size distribution of the inhaled aerosol. In reality this assumption is not valid and hence ICRP constituted a Task Group<sup>2</sup> to evolve deposition and clearance models taking into account among other parameters, the particle size distribution of the inhaled aerosol. After considerable deliberation Task Group came to the conclusion that the deposition fractions in various parts of the respiratory tract can be predicted fairly accurately, if one knows a particle size parameter, activity median aerodynamic diameter (AMAD). Task Group further states that the spread in particle size distribution (the geometric standard deviation) does not have significant effect on the deposition fractions.

The popular instruments among others<sup>3</sup> available for the measurement of AMAD are cascade impactor<sup>4</sup> and cascade centripeter<sup>5</sup>, which have severe limitations with the loading and wall losses respectively. In the present work an approach of using the fraction penetrating the HASL cyclone<sup>6</sup> at a particular flow rate as a measure of AMAD is presented.

Penetration Characteristics of HASL Cyclones

Health and Safety Laboratory of USAEC developed miniature cyclones<sup>6</sup>. These have penetration characteristics matching the so called Los Alamos Respirable Curves, when operated at a particular flow rate. Half inch cyclone<sup>7</sup> is operated at 9 litres/min and one inch cyclones<sup>8</sup> at 56 litres/min. Therefore penetration characteristics of these cyclones can be taken identical to Los Alamos Respirable Curve. Lynch<sup>9</sup> fitted an analytical expression for

this penetration curve and worked out mass penetrations for different particle size distributions. He gives a table in which mass penetrations are listed for a given count median aerodynamic diameter and a given geometric standard deviation, assuming that the distribution is log-normal. We have taken this data and computed mass penetration fractions with respect to AMAD for various geometric standard deviations using Hatch and Choate equation<sup>10</sup>. The data thus calculated was fitted to a least squares line on a semilog graph paper. Fig.1 gives such lines for geometric standard deviations between 1.5 and 3.0. A master line given in the figure is a computer fitted least squares line taking into account the entire set of data points used for drawing various lines.

Moss and Ettinger<sup>11</sup> have plotted penetration curves on a linear graph sheet for slightly different penetration curves (ACGIH Criterion).

#### Mass Penetration Fractions and AMAD

If one measures mass penetration fractions through a cyclone at proper flow rates, one can find AMAD using the master line in Fig.1. As can be seen, the value of AMAD depends to some extent on the geometric standard deviation. Aerosol normally encountered in field conditions has a GSD between 1.5 and 3.0 and the situations giving rise to distributions outside these limits are rare (see Table-III). In the absence of the knowledge of GSD, lines corresponding to GSD of 1.5 and 3.0 enveloping the master line are taken to provide uncertainty in AMAD. For example, if penetration fraction is 0.70, AMAD is  $2.3 \mu\text{m} + 0.12 \mu\text{m}$   
 $- 0.30 \mu\text{m}$ .

#### Pulmonary Deposition Fractions and AMAD

The object of measurement of AMAD is to predict the pulmonary deposition fractions. We can now examine whether the possible errors in cyclone measurement of AMAD have significant influence on the estimated pulmonary deposition fractions. ICRP Task Group<sup>2</sup> gives curve between AMAD and the pulmonary deposition fractions. A part of it is reproduced on the left half of Fig.1 for the size range of  $1.5 \mu\text{m}$  and  $10.0 \mu\text{m}$ . Table-I gives for various AMADs, the errors involved in the estimation of pulmonary deposition fractions because GSD is not known. It is also indicated by error bands on the curve in Fig.1. It can be seen that the error is minimum in the size range of  $4 \mu\text{m}$  (AMAD) and is less than 10% for the size range of 1.5 to  $7.0 \mu\text{m}$ . It is therefore concluded that for the size range of interest, the error in estimating the pulmonary deposition fraction is not significant.

Table-I

Errors Associated with the Estimation of Pulmonary Deposition by Cyclone Method for Various Particle Size (AMAD)

Particle size (AMAD) $\mu\text{m}$	1.5	2.0	3.0	4.0	5.0	6.0	7.0	8.0
% Pulmonary Deposition	19.7	17.6	14.9	13.0	11.4	10.3	9.2	8.2
with errors	+ 1.5 - 1.2	+ 1.3 - 0.6	+ 0.1 - 0.1	+ 0.5 - 0.6	+ 1.1 - 0.9	+ 1.2 - 1.5	+ 1.7 - 1.9	+ 2.0 - 2.1

## Field Measurements of AMAD Using Cyclones

### Measurements

Fig.2 shows experimental arrangement for cyclone air sampling. Flow rates were set at 9 litres/min for a half inch cyclone using a wet-test meter and also a soap bubble flow meter. Whatever penetrates the cyclone gets collected on an air sampling filter paper (Whatman GF/A glass fibre paper). Other sampling head collects a gross sample. The ratio of the activity on the filter paper following the cyclone and the filter paper from gross sample, provides fractional penetrations. Samples are generally taken over a period of 6 to 8 hours at a location close to breathing zone of workers. Cyclone is thoroughly cleaned before using for a subsequent measurements. Such determinations are made in different operating areas of Trombay Fuel Reprocessing Plant.

### Discussion of Results

Number of measurements are made at each of the locations mentioned in Table-II. These measurements are grouped as shown in Table-II. Mean per cent penetrations and the corresponding AMAD are also listed in the table. It can be seen that AMAD does not stay constant from day to day or from one operation to the other. However a mean AMAD can be associated with each location. First three areas are the areas normally used for handling Plutonium and the other areas mostly fission products. It is of interest to compare our results with the AMAD reported in the literature. Table-III lists the range of values obtained by various investigators, using different techniques. Our values compare well with the values shown in Table-III.

Table-II

Results of Measurements of AMAD by Cyclone Method Measurements in Various Areas of Trombay Fuel Reprocessing Plant

Sampled Areas	Number of Measurements					Weighted mean per cent penetrations	AMAD ( $\mu\text{m}$ )
	10 to 20	21 to 30	31 to 40	41 to 60	61 to 80		
Crane Space	12	3	5	1	-	22.9	7.5
Pu-Lab	4	4	3	2	-	28.1	6.8
Pu-Lab Maintenance	1	4	1	6	-	37.5	5.2
Operating Gallery	-	1	5	6	7	52.1	3.6
Control Lab	3	3	2	10	2	41.5	4.7
Service Corridor	1	-	2	3	1	43.6	4.4

Note: First three areas give aerosol data for Pu and the subsequent four for fission products.

### Revision of MPC<sub>a</sub> Values

In light of the actual field measurements described above, we can examine the currently used MPC<sub>a</sub> values. Currently used MPC<sub>a</sub> values assume that 12½% of the inhaled undergoes long term retention in the pulmonary region for relatively insoluble isotopes. According to the proposed Lung Model, this percentage depends upon AMAD. Table-IV gives the per cent undergoing long term retention with respect to AMAD. It is seen that we are under-estimating the hazard if AMAD is greater than 1.5  $\mu\text{m}$ . Therefore



for Fuel Reprocessing Plant, we are over-estimating the hazard by a factor of 2.0 in using the current  $MPC_a$  values for insoluble aerosols.

Table-III  
AMAD Measurements Available in Literature

Author	AMAD ( $\mu m$ )	GSD	Comments
Andersen <sup>12</sup>	3.4 to 7.2	2.0 to 2.3	Autoradiographic method. Pu aerosols covering different operations.
Sundararajan <sup>13</sup>	2 to 6	1.7 to 2.3	-do-
Stevens <sup>14</sup>	3.5 to 6.0	2.0 to 3.4	Centripeter technique for Pu-aerosols.
	4.7 to 7.0	2.5 to 3.4	Centripeter technique for fission product aerosols.
Langnead <sup>15</sup>	4.3	1.9	UF <sub>4</sub> aerosol. Centripeter technique.
	5.33 $\pm$ 1.39	2.46 $\pm$ 0.62	Pu-aerosols. Centripeter technique.
	2.5 to 11.0	-	-do- (Windscale)
	5 to 6.0	-	-do- (Springfield)

Table-IV

Ratios of  $MPC_a$  (for Insoluble Aerosols) for Old and New Lung Models

Particle size, AMAD ( $\mu m$ )	1.0	1.5	2.0	3.0	4.0	5.0	6.0	7.0	8.0
% long term pulmonary retention	14.4	11.8	10.6	9.0	7.8	6.9	6.2	5.5	4.9
$\frac{(MPC)_a \text{ (Old model)}}{(MPC)_a \text{ (New model)}}$	1.15	0.94	0.85	0.72	0.62	0.55	0.50	0.41	0.39

#### Conclusion

Cyclone method of measuring AMAD provides an acceptable method as long as we intend to use the information for estimating inhalation hazards. The method does not provide geometric standard deviation. The basic assumption used in this method, viz. that the penetration characteristics are identical to the Los Alamos Respirable Curve, should be borne in mind. Proper flow rates must be employed. The method is simple and operational health physicists can adopt this method without needing additional skills. The measurement incidentally provides respirable fractions. Recent trend in defining TLV<sup>7</sup> (Threshold Limit values) for Silica and such other particulates, is to define TLV in terms of the respirable fractions. The method has a much wider applications.

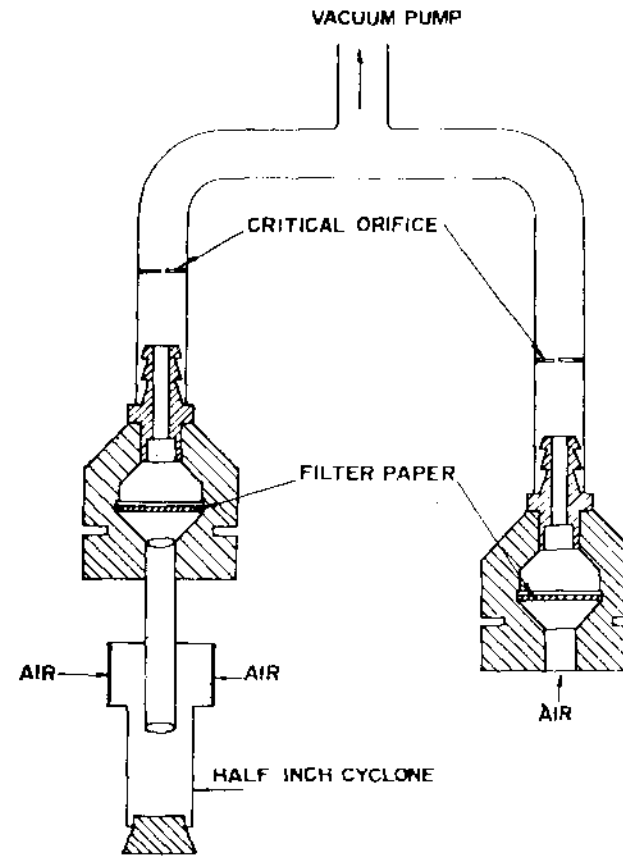
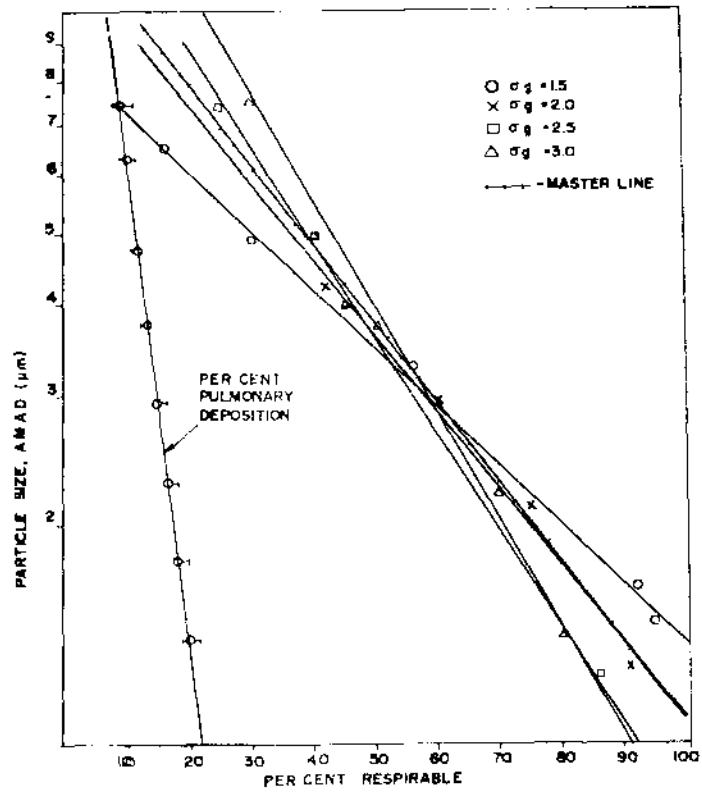
#### Acknowledgements

Our sincere thanks are due to Dr.A.K.Ganguly and Shri S.D.Soman for the helpful discussions and to Shri A.N.Prasad and Shri M.N.Nadkarni for providing facilities for the studies.

#### References

1. ICRP Committee: "Report of ICRP Committee II on Permissible Dose for Internal Radiation(1959)", Health Physics 3, 1 (1960).
2. Task Group on Lung Dynamics: "Deposition and Retention Models", Health Physics 12, 173 (1966).
3. T.T.Mercer: "Air Sampling Problems Associated with the Proposed Lung Model", CONF-661018, 87 (1966).
4. K.R.May: "The Cascade Impactor: An Instrument for Sampling Coarse Aerosols", J.Sci.Instr. 22, 187 (1945).
5. R.F.Hounam and R.J.Sherwood: "The Cascade Centripeter: A Device for Determining the Concentration and Size Distribution of Aerosols", Amer.Indust. Hyg.Assoc.J. 26, 122 (1965).
6. M.Lippmann: "Respirable Dust Sampling", Amer. Indust.Hyg.Assoc.J. 31, 138 (1970).
7. Aerosol Technology Committee: "Guide for Respirable Mass Sampling", Amer.Indust.Hyg.Assoc.J. 31, 133 (1970).
8. M.Lippmann - Personal Communication (1972).
9. J.R.Lynch:"Evaluation of Size Selective Presamplers", Amer.Indust.Hyg. Assoc.J. 31, 548 (1970).
10. O.G.Haabe: "Particle Size Analysis Utilisers Grouped Data and the Log-Normal Distribution", J.of Aerosol Science 2, 289 (1971).
11. O.R.Moss and H.J.Ettinger: "Respirable Dust Characteristics of Polydisperse Aerosols" Amer.Indust.Hyg.Assoc.J. 31, 546(1970).
12. B.V.Andersen et al: "Supplementary Data Sources for Evaluation of Insoluble Actinide Inhalation Exposures" BNWL-SA-1572 (1968).
13. A.R.Sundararajan et al.: "Particle Size Distribution of Pu Aerosols in Laboratory Air", II International Congress of IRPA (1970).
14. D.C.Stevens: "The Particle Size and Mean Concentration of Radioactive Aerosols Measured by Personal and Static Air Samples", Ann.Occup.Hyg. 12, 33(1969).
15. W.A.Langmead: "The objectives of Air Monitoring and the Interpretation of Air Sampling Results", I International Congress of IRPA (1967).

1231



CYCLONE SAMPLING ASSEMBLY

DOSE COMMITMENT TO THE LUNG FROM INHALATION  
OF RADIOCOBALT IN POWER REACTOR OPERATIONS

P.Kotrappa, S.D.Soman & B.Raghunath  
Health Physics Division  
Bhabha Atomic Research Centre  
Bombay-400085, India

Abstract

Personnel working in the operational and maintenance areas of Tarapur Atomic Power Plant (BWR Type) have shown detectable amounts of  $^{60}\text{Co}$  in their system, notably in the lung area. Large number of gross and respirable air samples taken in the working areas provided the air activity levels of  $^{60}\text{Co}$  and also the aerosol parameter, activity median aerodynamic diameter (AMAD). These data in conjunction with the long term clearance half-life available in literature are used to compute the quantity of  $^{60}\text{Co}$  in lungs at any time due to chronic inhalation. Parameters of the new ICRP lung model are used in the computation. Radiation dose commitment to the lung due to the retained  $^{60}\text{Co}$  is computed.

Introduction

Personnel working in Tarapur Atomic Power Plant, a 400 MWe BWR Type Power Reactor, have shown detectable amount of  $^{60}\text{Co}$  in their system, notably in chest area<sup>1,2</sup>. Air samples collected in the working environment also indicated that the major portion of the long lived component is due to  $^{60}\text{Co}$ . Chest clearance studies have shown that  $^{60}\text{Co}$  behaves like a class Y radionuclide as defined by the ICRP Task Group on Lung Dynamics<sup>3</sup>. Airborne concentration and the particle size parameter, activity median aerodynamic diameter (AMAD) provide the data to calculate both the material and dose commitments to the lung<sup>4</sup>.

Airborne Radiocobalt

The gamma spectra of air samples collected shows predominantly  $^{60}\text{Co}$ ,  $^{54}\text{Mn}$  and  $^{137}\text{Cs}$ . Nearly 80 to 90% of the entire airborne activity is found to be due to  $^{60}\text{Co}$  coming in air, probably as insoluble cobalt oxide aerosol. Origin of this activation product is traced to the material of the piston (stellite) used in the water circulating pumps. It has been reported<sup>1</sup> that during July 1970-July 1971, 182 cases were detected with chest burdens less than 10 nCi and 8 cases with chest burdens more than 10 nCi within a year of the operation of the TAPP.

Clearance of  $^{60}\text{Co}$  from lungs has been studied by several investigators. Most of these studies relate to accidental inhalation of  $^{60}\text{Co}$  oxide and a few relate to the occupational exposure in reactors and hot cell areas. Table-I gives the results of these studies. It is seen that effective clearance half-life varies considerably. 500 to 1000 days may be taken as long term effective clearance half-life. It is typically a class Y radionuclide as per the classification of the ICRP Task Group on Lung Dynamics<sup>3</sup>. Using this data in conjunction with the rate of inhalation and the particle size

parameter (AMAD), it is possible to calculate the lung burden and dose commitment to the lungs making use of the equations given by Kotrappa<sup>4</sup>.

Table-I

Long Term Clearance Half-Life for <sup>60</sup>Co from Chest Area

Reference	Effective Half-life (days)
Sill <sup>2</sup> et al, (1964)	172 to 700
Rundo <sup>6</sup> et al, (1966)	90*; 720 **
ICRP <sup>7</sup> -10 (1968)	115 - 400 40 -1000
Edvardsson <sup>8</sup> (1970)	180
Newton and Rundo <sup>9</sup> (1971)	935; 1460; 1210; 400; 750
Gupton and Brown <sup>10</sup> (1972)	225

\*Single exposure

\*\* Multiple exposure

Measurement of Airborne Concentration and AMAD

Technique of measuring the particle size parameter, AMAD, using a cyclone has been described in greater detail by Kotrappa<sup>5</sup> et al. In brief, it consists of measuring the respirable fraction of the activity which simply is the fraction of the activity penetrating the cyclone at a proper flow rate. Making use of a graph between the fractional penetration and AMAD, corresponding AMAD can be read out. Sampling arrangement consists of taking two simultaneous air samples in close proximity to each other. One sample is normal air sample through a filter paper and another is through a filter paper following the cyclone. Ratio of the activities of the latter to the former provides respirable fraction or the fraction penetrating the cyclone at a proper flow rate. Samples are counted in 512 channel gamma spectrometer, and the ratio of the peak counts are used for calculating the respirable fractions. Absolute activities of <sup>60</sup>Co on these papers are also determined by standard calibration techniques.

Results of Measurements

Table-II gives a summary of the measurements carried out in different areas of TAPP. Rad waste conveyor area and Rad waste air receiver area are normally restricted. These areas show high airborne concentration, at times more than the presently adopted maximum permissible concentration of 10000 pCi/m<sup>3</sup>. Areas at 103 and 125 feet are the operating areas from where the operators control the rod movements. 200 feet level area is generally an operating area. Fuel pond is located here and general maintenance is carried out in this area. High airborne activities are observed only during special maintenance works. AMAD of 4 μm can be taken to represent an average value for all the areas of TAPP.

Computation of Lung Commitments

Method of calculating the burden and dose to different respiratory compartments from continuous inhalation of a radioactive aerosol, making use of the proposed ICRP lung model has been described in detail elsewhere<sup>4</sup>. Fig.1 gives the computed burdens in pulmonary region and in lymph node

regions after chronic inhalation. These are general curves for the inhalation rate of 1 pCi/day, for an aerosol of 1  $\mu\text{m}$  (AMAD). To obtain results for  $^{60}\text{Co}$  for an aerosol of 4  $\mu\text{m}$  (AMAD), retention should be multiplied by 0.5. Fig. 2 gives the integrated dose commitments. Again these are general curves. To obtain results for  $^{60}\text{Co}$  one has to multiply the doses by 0.36 (0.5, correction for AMAD of 4  $\mu\text{m}$  and 0.72, correction for effective energy). A few examples of such computations for two different effective half-lives are given in Table-III.

Table-II  
Activity and particle size data in various areas of TAPP

Area	No. of samples	RANGE OF VALUES			Median AMAD ( $\mu\text{m}$ )	$\sigma_g$	
		$^{60}\text{Co}$ (pCi/m <sup>3</sup> )		Per cent respirable			
		Total	Respirable		AMAD ( $\mu\text{m}$ )		
Rad Waste Air receiver room	10	575-175463	245-84388	37-69	2.3-5.2	3.9	1.23
Rad Waste conveyor area	10	344-188826	132-59299	11-74	2.05-10.0	4.7	1.74
103 & 125 ft elevations including interior of Dry well	8	25-3062	19-908	30-76	2.0-6.2	3.8	1.42
200 ft. elevation	14	33-3921	23-1371	33-93	1.3-5.8	3.7	1.43

Table-III  
Cumulative Retention and Dose for Continuous Inhalation  
of 4  $\mu\text{m}$  (AMAD) Aerosols at the rate of 1 pCi/day.

Inhalation Time (days)	Retention in Pulmonary region (pCi)	Retention in Lymph region (pCi)	Dose to Pulmonary region (mrems)	Dose to Lymph Region (mrems)
<u>Effective Half-life = 500 days</u>				
100	7.1	0.13	0.03	0.01
500	27.1	2.57	0.55	1.10
1000	40.7	8.30	1.73	7.55
5000	54.0	73.5	17.0	400.0
10000	54.0	158.0	24.8	1820.0
<u>Effective Half-life = 1000 days</u>				
100	7.3	0.064	0.03	0.005
500	31.8	1.44	0.62	0.610
1000	54.0	5.2	2.24	4.45
5000	105.0	63.0	28.6	322.0
10000	108.0	148.0	66.0	1610.0

### Computation for Typical Inhalation Case (Pulmonary Region)

It is seen that the concentration of  $^{60}\text{Co}$  is of the order of 200  $\text{pCi}/\text{m}^3$  in continuously occupied operating areas such as 200 feet level. During a working day a standard man inhales  $10 \text{ m}^3$  and therefore daily intake is about 2000  $\text{pCi}/\text{day}$ . From Table-III, the retention after 500 days in pulmonary region will be  $(27.1 \text{ pCi} \times 2000)$  of the order of 54  $\text{nCi}$ . Saturation burden (after 5000 days) will be of the order of 108  $\text{nCi}$ . Therefore it is not surprising to find lung burdens of the order of 50  $\text{nCi}$ , if they work for 500 days in the atmosphere that has airborne concentration of  $200 \text{ pCi}/\text{m}^3$ . Corresponding dose to pulmonary region will be 1.10  $\text{rem}$  in 500 days. At saturation (that is after 5000 days), there will be a steady dose rate of  $\int (24.8-17.0) \times 2000/5000 \int 3.1 \text{ mrem}/\text{day}$ , which is about 6% of the daily allowed dose of 50  $\text{mrem}/\text{day}$  for lungs. Probably 10% of the allowed exposure to a person should be kept aside to take care of the internal exposure due to  $^{60}\text{Co}$  even at saturation.

### Computation for Lymph Node Region

Calculations can be made for lymph node regions on the same line as is done for pulmonary region. Currently used lung model<sup>3</sup> assumes that 70% of the material reaching the lymph node region is retained permanently and only 10% undergoes biological elimination. Recently Morgan<sup>11</sup> has stated that experimental data supports the view that 90% undergoes biological elimination with a 1000 days half-life and only 10% undergoes permanent retention. Further Morgan states that for calculating the doses to lymph nodes, controversy still exists whether one should use the mass of pulmonary lymph nodes or whether one should use the entire mass of circulating lymphocytes. He states again that lymph node tissue is known to be highly radiation resistant and the tissue is not likely to be a critical organ. Based on these facts, calculations made on the basis of the currently used lung model<sup>3</sup> may not be right. Pulmonary exposures would be controlling the hazard. Therefore we have not attempted to calculate the results for typical inhalation case and to draw any conclusion regarding the dose commitments.

### Conclusion

Detectable amount of  $^{60}\text{Co}$  is expected in the lungs of the workers in operational areas of TAPP. Probably 10% of the annual allowed dose should be left as the dose commitments for these workers due to inhalation of  $^{60}\text{Co}$  alone. Routine chest monitoring programme is essential. It is recommended that further investigation be done to see whether this problem can be eliminated once for all by stopping the source of cobalt.

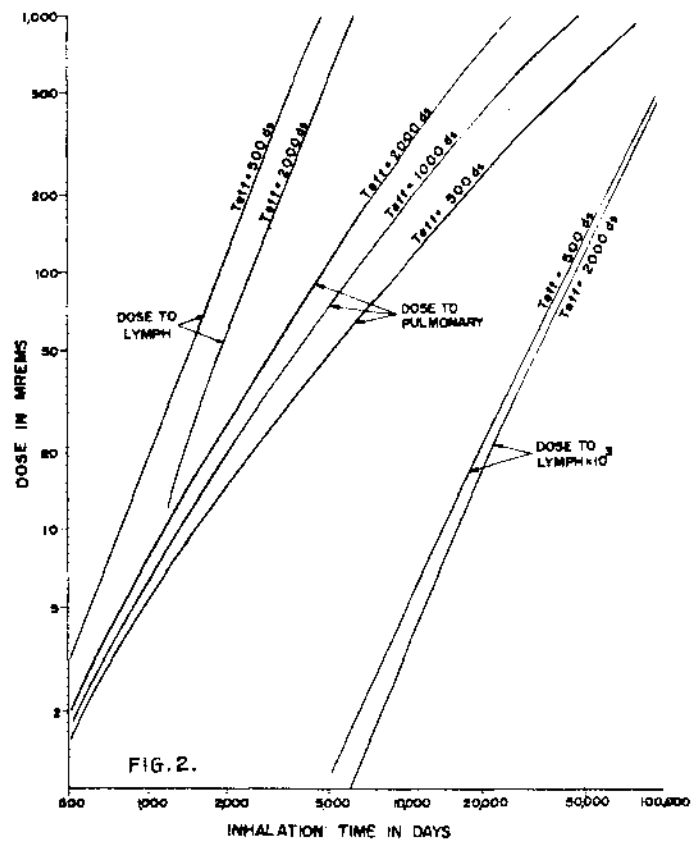
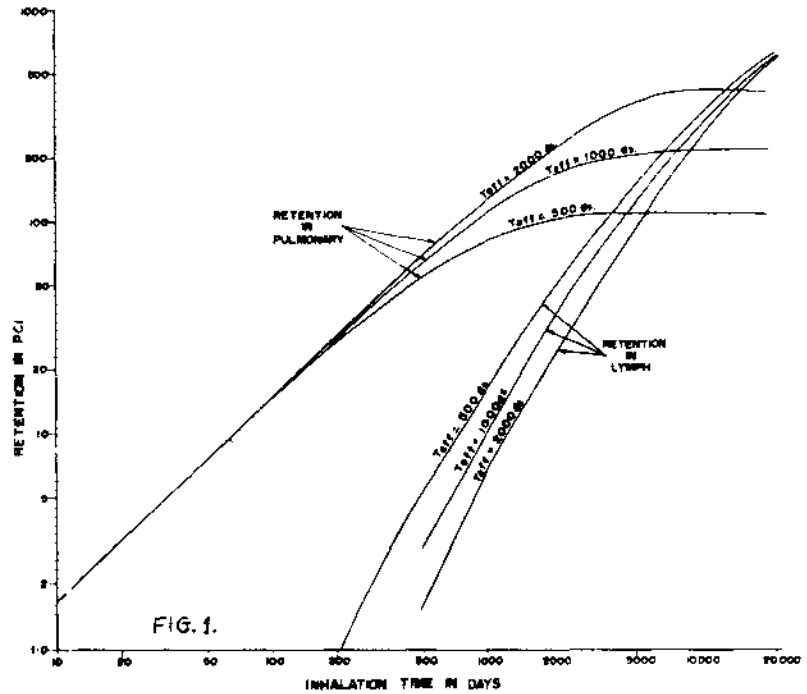
### Acknowledgements

Our sincere thanks are due to Dr. A.K. Ganguly for his interest in this work, Shri P. Abraham and Shri D. Pattnaik for their co-operation during our work at TAPP.

### References

1. I.S. Bhat et al. "Assessment of Radioactive Contamination in Man" IAEA-SM-150/54, STI/PUB/290 (1971).
2. C.W. Sill et al. "Assessment of Radioactive Contamination in Man" Vol. 1, STI/PUB/84, IAEA, 217 (1964).
3. ICRP Task Group on Lung Dynamics, Health Physics 12: 173 (1966).
4. P. Kotrappa. "Calculation of the Burden and Dose to the Respiratory Tract from Continuous Inhalation of a Radioactive Aerosol, Health Physics 17: 429 (1969).
5. P. Kotrappa et al, "HASL Cyclone as an Instrument for Measuring Aerosol Parameters for New Lung Model" Current IRFA Proceedings (1973).
6. J. Rundo, Health Physics and Medical Division Progress Report AERE-PR/HFM 5(4) (1964).
7. ICRP Publication 10, Report of Committee IV, (1968).

8. K.A.Edvardsson "Assessment of Radioactive Contamination in Man" IAEA-SM-150/53, STI/PUB/290 (1972).
9. D.Newton and J.Rundo, "The long-term retention of inhaled  $^{60}\text{Co}$ " Health Physics 21: 377 (1971).
10. E.D.Gupton and P.E.Brown, "Chest Clearance of  $^{60}\text{Co}$  Oxide", Health Physics 23: 767 (1972).
11. K.Z.Morgan, "Assessment of Radioactive Contamination in Man" IAEA-SM-150/50, STI/PUB/50 (1971).





# A METHOD FOR DETERMINING THE DISSOLUTION CHARACTERISTICS OF ACCIDENTALLY RELEASED RADIOACTIVE AEROSOLS\*

G. M. Kanapilly, O. G. Raabe and H. A. Boyd

Lovlace Foundation for Medical Education and Research  
Albuquerque, New Mexico 87108

## Abstract

Early assessment of the dissolution characteristics of radioactive aerosol particles involved in inhalation accidents is important for hazard evaluation and selection of proper therapeutic procedures. Since accidentally produced aerosols are usually of undetermined physicochemical properties, prediction of their dissolution characteristics is difficult. Therefore, a rapid experimental evaluation of their dissolution behavior is desirable. An *in vitro* method is described which provides both early and long-term measurements of dissolution behavior of aerosol samples in a parallel-flow system which simulates *in vivo* dissolution as in the lung after inhalation deposition. Health physics applications of dissolution data are discussed.

## Introduction

The biological hazards of an inhaled radioactive material depend upon the kinetics of deposition, retention, distribution and absorption of the inspired aerosol. The dissolution pattern of the aerosol particles is especially important in determining the retention and absorption from the site of deposition. Also, for estimations of lung burdens of radionuclides by urine analysis, *in vivo* dissolution rates of the lung-deposited aerosol particles are important.<sup>1,2</sup> Further, the effectiveness of chelation therapy for inhaled actinide and lanthanide nuclides will depend on the degree of solubility of the aerosol.<sup>3</sup>

The *in vivo* dissolution behavior of an accidentally released aerosol may not be readily predicted because its physicochemical nature may be unknown and because the *in vivo* dissolution is a highly complex process.<sup>4,5</sup> Due to particle size effect alone, rates of dissolution of inhaled radioactive aerosol particles may vary several orders of magnitude. Further, the chemical forms of the aerosol may greatly influence its rate of dissolution. Therefore, rapid experimental evaluation of the dissolution characteristics of radioactive aerosols involved in accidental inhalation is highly desirable. *In vitro* dissolution rates of aerosol particles comparable to *in vivo* rates of dissolution may be measured experimentally with flowing solvents.<sup>4</sup> In this report a parallel-flow system is described which provides both early and long-term measurement of *in vitro* rates of dissolution of aerosol samples. The results of *in vitro* dissolution experiments on a variety of test aerosols, and health physics application of *in vitro* rates of dissolution are discussed.

## Materials and Methods

After measurement of the radioactivity of samples of aerosol particles collected on filters, the filters were sandwiched between two 47 mm diameter 100 mpm pore size cellulose acetate membrane filters and held together

\* Research performed under AEC Contract AT(29-2)-1013.

tightly with plastic holders for elution with a flowing solvent, a simulant of biological fluid.<sup>4</sup> The experimental arrangement with a cross sectional view of the filter holder is shown in Fig. 1. Two cavities were formed when the filter was placed in this holder. The solvent passed into the lower cavity (0.6 ml) and eluted any dissolved material diffusing into this region from the filter. Tests with colored compounds showed that the upper static layer of "solution" (0.2 ml) could be eluted out within a few hours. Thus, in this system the dissolution and removal of dissolved material from the aerosol particles occurred by the indirect contact of a flowing solvent through filters. One can envision a general analogy between this dissolution system and the dissolution of particles and transport of the dissolved material from the pulmonary region to the blood capillaries.

The solvent from a large reservoir (20 l) was introduced into the secondary reservoir (200 ml) at 1 ml/min by gravity flow and controlled with a valve. The secondary reservoir, the filter holder and the associated tubing, all non-corrosive plastics, were immersed in water bath at 37°C. The secondary reservoir allowed temperature equilibration and served as a bubble trap. The filter holder was placed in the bath at an angle to prevent reduction in solvent flow rate by gas bubbles. Aliquots of eluates on planchets were counted with a low-background proportional counter for beta emitters and alpha emitters were counted with a liquid scintillation counter using Aquasol. Low levels of plutonium were analyzed by alpha spectroscopy. Fractional dissolutions were calculated for each period of observation as the fraction of activity remaining on the filter dissolved during that period. After each dissolution, the activity remaining on the sample was also determined.

The solvent was prepared with the composition similar to that of blood serum by dissolving analytical grade chemicals in distilled water (Table 1). The basis for the choice of this composition for the solvent was discussed earlier.<sup>4</sup> An antibacterial agent, 50 ppm of alkyl benzyl dimethyl ammonium chloride was also used in this solvent. In some studies a strong chelating agent, either ethylenediaminetetraacetic acid (EDTA) or diethylenetriamine-pentaacetic acid (DTPA), was also added to the solvents. The pH of the solvent was kept at 7.3 to 7.4 by equilibrating with 5% CO<sub>2</sub> in N<sub>2</sub> contained in a weather balloon (Fig. 1). In this study all experiments were done with a solvent flow rate of 1 ml/min and temperature of 37°C.

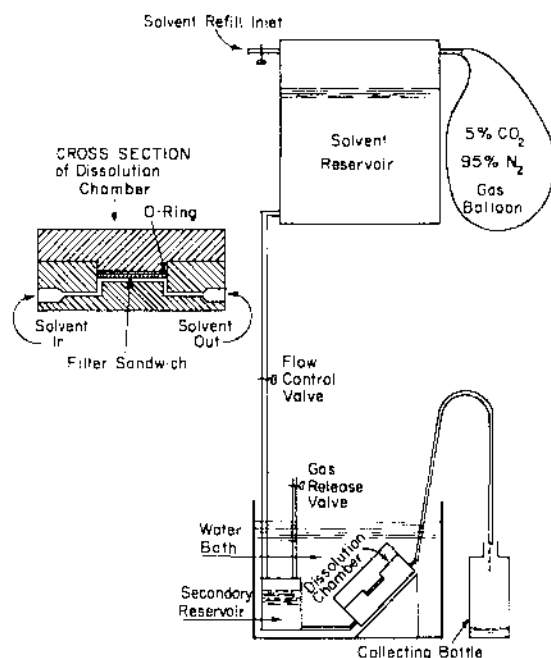


Figure 1. Schematic of parallel-flow dissolution system and filter holder assembly.

Table 1  
Preparation of Serum Simulant for *In Vitro* Elutions

Chemical Compound	m moles/liter
NaCl	116
NH <sub>4</sub> Cl	10
NaHCO <sub>3</sub>	27
NaH <sub>2</sub> PO <sub>4</sub>	1.2
Na <sub>3</sub> Citrate	0.2
Glycine	5.0
L-Cysteine	1.0
H <sub>2</sub> SO <sub>4</sub>	0.5
CaCl <sub>2</sub>	0.2

The methods of production and characterization of polydisperse aerosols of  $^{95}\text{Zr}$ - $^{95}\text{Nb}$  labeled zirconium oxalate, carbonate and dioxide and  $^{90}\text{Sr}$  in fused montmorillonite clay have been described earlier.<sup>6</sup> Samples of monodisperse particles of  $^{238}\text{PuO}_2$  and  $^{239}\text{PuO}_2$  were prepared under identical conditions with  $1100^\circ\text{C}$  heat treatment by the method described by Kotrappa et al.<sup>7</sup> To demonstrate the usefulness of *in vitro* dissolution studies, a filter sample containing a  $^{239}\text{Pu}$  aerosol accidentally released at another laboratory and involved in a human exposure was also used in this study.

### Results and Discussion

*In vitro* rates of dissolution of  $^{90}\text{Sr}$  fused clay and  $^{95}\text{Zr}$ - $^{95}\text{Nb}$  labeled aerosol particles (zirconium oxalate, zirconium carbonate and zirconium dioxide) were found comparable to the corresponding dissolution in the lungs of animals.<sup>4</sup> The dissolutions in the fractions collected during the first two minutes were 85%, 0.2% and 0.1%, respectively, for the oxalate, carbonate and dioxide aerosols. Both the dioxide and carbonate particles were comparatively insoluble; only 0.6% and 2%, respectively, dissolved during 150 minutes. The oxalate particles dissolved 93% during 100 minutes. The fractional dissolution of  $^{90}\text{Sr}$  in fused clay particles during the 5th to 11th day of continuous elution was a relatively constant value of  $(1.3 \pm 0.1 \text{ S. D.}) \times 10^{-3}$  per day.<sup>5</sup> The *in vitro* rate constant of dissolution of  $^{90}\text{Sr}$ -fused clay particles was about  $3.3 \times 10^{-8} \text{ g. cm}^{-2} \cdot \text{day}^{-1}$ .<sup>4</sup> Good agreement between the *in vitro* rates of dissolution of monodisperse  $^{137}\text{Cs}$ -fused clay particles in Beagle dog lungs, after inhalation deposition, has also been observed.<sup>8</sup>

The dissolution of monodisperse ( $\sigma_g < 1.2$ )  $\text{PuO}_2$  aerosols of both  $^{239}\text{PuO}_2$  and  $^{238}\text{PuO}_2$  (80%  $^{238}\text{Pu}$  + 20%  $^{239}\text{Pu}$ ) was studied for 3 to 4 weeks (Fig. 2). Serum simulant containing  $2 \times 10^{-4}$  MEDTA was used as the solvent for the study with  $0.36 \mu\text{m}$   $^{238}\text{PuO}_2$  and during the later parts of the  $^{239}\text{PuO}_2$  study. The  $\text{PuO}_2$  aerosol particles (density,  $\rho \approx 7 \text{ g. cm}^{-3}$ ) were relatively insoluble. The average dissolution rates of  $\text{PuO}_2$  and other pertinent data are summarized in Table 2. If the rate of dissolution of a particle is proportional to its surface area, for spherical particles the rate constant  $k = \rho Df/6$  when  $D$  is the diameter of the particle and  $f$  is the fractional dissolution per unit time.<sup>4</sup> The dimensions of  $k$  are  $\text{g. cm}^{-2} \cdot \text{day}^{-1}$  when the units of  $\rho$ ,  $D$  and  $f$  are  $\text{g. cm}^{-3}$ ,  $\text{cm}$  and fraction per day, respectively.

During the first day, about 3% of  $^{238}\text{PuO}_2$  ( $D = 0.36 \mu\text{m}$ ) was dissolved in serum simulant containing EDTA and higher dissolution of this sample,

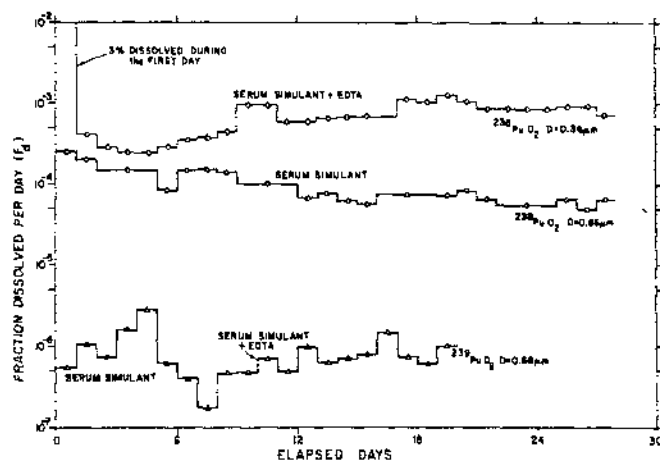


Figure 2. Dissolution data histogram for monodisperse particles of  $^{238}\text{PuO}_2$  and  $^{239}\text{PuO}_2$  as determined in a parallel-flow dissolution system.

Table 2  
Summary of Dissolution of PuO<sub>2</sub> at 37°C

Sample Form	Original sample activity $\mu$ Ci	Physical diameter, D $\mu$ m	Solvent	Span of study Days	Average fraction dissolved Per Day	Dissolution rate constant, k $g \cdot cm^{-2} \cdot day^{-1}$
<sup>239</sup> PuO <sub>2</sub>	6.8	0.66	Serum Simulant and Serum Simulant + EDTA	0 to 20	$(8.5 \pm 3.4) \times 10^{-7}$	$(6.5 \pm 2.6) \times 10^{-11}$
<sup>238</sup> PuO <sub>2</sub>	4.6	0.66	Serum Simulant	0 to 28	$(9.7 \pm 5.2) \times 10^{-5}$	$(7.5 \pm 4.0) \times 10^{-9}$
<sup>238</sup> PuO <sub>2</sub>	4.9	0.36	Serum Simulant + EDTA	1 to 28	$(7.1 \pm 3.0) \times 10^{-4}$	$(4.1 \pm 1.7) \times 10^{-8}$

compared to that of 0.66  $\mu$ m <sup>238</sup>PuO<sub>2</sub> particles in serum simulant, persisted. Increased dissolution of <sup>238</sup>PuO<sub>2</sub> due to the presence of EDTA is indicated by the higher rate constant (Table 2). The presence of EDTA in the solvent did not increase the rate of dissolution of <sup>239</sup>PuO<sub>2</sub> particles. It is clearly shown that the <sup>238</sup>PuO<sub>2</sub> particles are much more soluble than <sup>239</sup>PuO<sub>2</sub> particles.

Generally the *in vivo* dissolution of PuO<sub>2</sub> is considered to be extremely small.<sup>9-11</sup> Higher absorption of <sup>238</sup>Pu compared to <sup>239</sup>Pu from the lungs of Beagle dogs to the systemic burden, after inhalation deposition of <sup>238</sup>PuO<sub>2</sub> and <sup>239</sup>PuO<sub>2</sub>, has also been reported.<sup>12</sup> Ineffectiveness of chelates to enhance <sup>239</sup>Pu removal from <sup>239</sup>PuO<sub>2</sub> particles has also been observed in DTPA chelation therapy treatments.<sup>3, 11</sup> These *in vivo* observations are in qualitative agreement with the *in vitro* studies. Increased *in vitro* dissolution of <sup>238</sup>PuO<sub>2</sub> in the presence of EDTA suggests the usefulness of chelation therapy in <sup>238</sup>PuO<sub>2</sub> inhalation exposure.

Two dissolution experiments with an accidentally released <sup>239</sup>Pu aerosol sample (97.5% <sup>239</sup>Pu and 2.5% <sup>241</sup>Am), one with serum simulant (1.7  $\mu$ Ci) and another (1.2  $\mu$ Ci) with serum simulant containing  $1 \times 10^{-3}$  M DTPA were conducted. The dissolution experiment with serum simulant as the solvent consisted of 8 days of continuous elution and then elution with serum simulant containing  $1 \times 10^{-3}$  M DTPA for an additional four days. The early dissolution data obtained from these studies, during the first five hours, are shown in Fig. 3. The data obtained during the entire dissolution experiment are summarized in Fig. 4. Higher rates of dissolution in serum simulant containing DTPA, compared to that in serum simulant alone, were seen between 2 hours and up to about 5 days of elapsed time. When the sample eluted with serum simulant was subsequently eluted with serum simulant containing DTPA, increased rates of dissolution were observed. The total dissolutions were 29.0% in serum simulant containing DTPA during 7 days and 31.5% from the second sample during 12 days. The dissolution in serum simulant during the first 8 days was 17.9% and in serum simulant containing DTPA, during the next 4 days, 13.6%. About 30% of the sample was in a relatively soluble form and exhibited higher dissolution in the presence of DTPA.

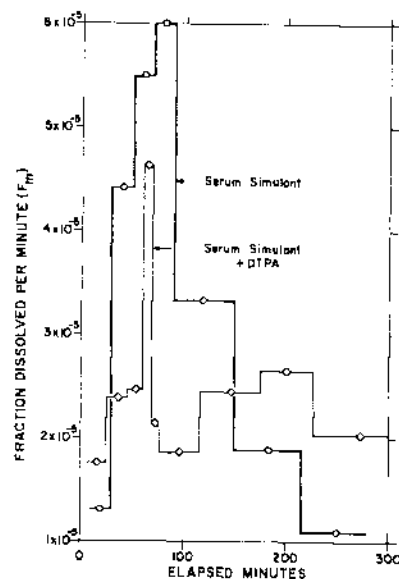


Figure 3. Early dissolution data histogram of an accidental <sup>239</sup>Pu aerosol sample as determined in a parallel-flow dissolution system with serum simulant and serum simulant containing DTPA.

The usefulness of *in vitro* dissolution studies on accidentally released aerosols may be illustrated with the data presented. The aerosol was originally considered to be of insoluble  $^{239}\text{PuO}_2$ . An individual who inspired this aerosol had an estimated lung burden of about 450 nCi of  $^{239}\text{Pu}$  and underwent treatments consisting of bronchopulmonary lavage and DTPA therapy beginning on the eighth day after exposure.<sup>13, 14</sup> The urinary excretion on the seventh day was measured as 0.112 nCi. The *in vitro* dissolution data for the aerosol sample may be used to estimate the initial lung burden (after the first phase clearance),  $Q_0$ , by Healy's method.<sup>1</sup> Urinary excretion,  $E_u$ , at the  $R$ th day after inhalation deposition is given as:<sup>1</sup>

$$E_u = 0.002\lambda_s Q_0 \int_0^R e^{-\lambda t} (R-t)^{-0.74} dt$$

with  $t$  the time in days after inhalation deposition,  $\lambda_s$  the daily fractional rate of dissolution and transfer to the blood (assumed constant by Healy), and  $\lambda$ , the total daily fractional removal rate from lower respiratory tract due to both dissolution,  $\lambda_s$ , and ciliary clearance,  $\lambda_c$ . The *in vitro* fractional dissolution of the aerosol sample in serum simulant varied from  $5.2 \times 10^{-2}$  to  $4.5 \times 10^{-3}$  per day (Fig. 4). However, the total dissolution during the first 7 days was 17.1% with an average fractional dissolution of  $2.44 \times 10^{-2}$  per day. This value may be used as  $\lambda_s$  in the Healy equation. The numerical values for the integral  $\int_0^R e^{-\lambda t} (R-t)^{-0.74} dt$  when  $R = 7$ , are relatively insensitive to  $\lambda$  values; these values are 5.58 and 4.86 for  $\lambda$  of  $2.44 \times 10^{-2}/\text{day}$  ( $\lambda = \lambda_s$ ) and  $5.0 \times 10^{-2}/\text{day}$  ( $\lambda \approx 2\lambda_s$ ), respectively.<sup>2</sup> Also, after the rapid clearance phase, the ciliary clearance rate from the pulmonary region, is relatively slow,  $< 8 \times 10^{-3}$  per day.<sup>15</sup> If  $\lambda = \lambda_s = 2.44 \times 10^{-2}/\text{day}$  then  $Q_0 = 0.112 / 0.002 \times 2.44 \times 10^{-2} \times 5.58 = 411$  nCi. Similarly if  $\lambda = 5.0 \times 10^{-2}$  then  $Q_0 = 472$  nCi. These values are in general agreement with the estimate of  $Q_0$ , 450 nCi obtained by whole body counting.<sup>13</sup>

#### Conclusions and Recommendations

When human exposure to an accidental aerosol is involved, an *in vitro* dissolution study should be done on a representative sample collected on a filter as soon as practicable. If size selective sampling is available only the respirable size particles should be used in the *in vitro* dissolution. Glass fiber filters for the collection of the aerosol sample should be avoided, because the elution of polyvalent nuclides from this type of filter is less than quantitative. For rapid solubility range finding, however, any filter sample containing a known activity of the accidental aerosol may be used. The method of choice for the determination of *in vitro* dissolution is continuous elution with the parallel flow system (Fig. 1) with serum simulant, 37°C, at a flow rate of 1 ml/min. Direct assaying of eluted radioactivity during the first hour of elution should indicate the range of solubility of the aerosol. If significant fractional dissolution is shown, DTPA treatment of exposed individuals may be useful. *In vitro* dissolution studies of the aerosol sample with DTPA added to the serum simulant may provide useful additional information on the efficacy of DTPA therapy. The rates of dissolution of a relatively insoluble aerosol

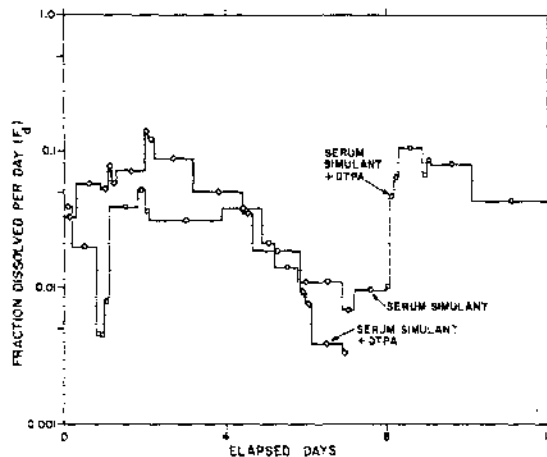


Figure 4. Dissolution data histogram of an accidental  $^{239}\text{Pu}$  aerosol sample during several days as determined in a parallel-flow dissolution system. The solvents used are indicated in the figure.

should be determined for longer periods, several weeks, so that relatively constant fractional dissolution may be obtained. Such data then may be used for the evaluation of biological disposition of the inhaled material and for the estimation of lung burden from urine and blood radioactivity levels.<sup>1</sup>

Acknowledgements - The authors are indebted to Drs. R. O. McClellan, R. G. Thomas, B. B. Boecker, R. G. Cuddihy and J. A. Mewhinney for their suggestions; to Mrs. P. K. Peterson, Mr. R. K. Latven and Mr. J. J. Miglio for technical assistance; to Mr. F. C. Rupprecht for editorial assistance and to Mr. E. E. Goff for preparation of the illustrations.

#### References

1. Healy, J. W., Amer. Ind. Hyg. Qtly. 18, 261 (1957).
2. Nelson, I. C., Health Phys. 22, 191 (1972).
3. Lafuma, Jr., Accidental Irradiation at Place of Work, Proceedings of the International Symposium on Accidental Irradiation at Place of Work, pp. 551-559, EURATOM, Nice, France, 26-29 April, 1966.
4. Kanapilly, G. M., Raabe, O. G., Goh, C. H. T. and Chimenti, R. A., Health Phys. 24, 497 (1973).
5. Kanapilly, G. M. and Goh, C. H. T., Health Phys. (in press).
6. Raabe, O. G., Kanapilly, G. M. and Newton, G. J., in Inhaled Particles III, Vol. I, pp. 3-17, (Edited by W. H. Walton) Unwin Brothers, Ltd. Surrey, England, 1971.
7. Kotrappa, P., Boyd, H. A. and Wilkinson, C. J., Health Phys. 22, 837 (1972).
8. Thomas, R. G., Thomas, R. L., Boecker, B. B., Kanapilly, G. M. and McClellan, R. O., Lovelace Foundation Fission Product Inhalation Program, Annual Report, 1971-1972, LF-45, pp. 37-49.
9. Morrow, P. E., Gibb, F. R., Davies, H., Mitola, J., Wood, D., Wraight, N. and Campbell, H. W., Health Phys. 13, 113 (1967).
10. Bair, W. J. and Park, J., in Radiation Protection (Edited by W. S. Snyder), pp. 181-197 (1968).
11. Schofield, G. F. and Lynn, J. C., Health Phys. 24, 317 (1973).
12. Park, J. F., Howard, E. B. and Bair, W. J., Report No. AFWL-TR-69-75 (1969).
13. Mann, J. R., Health Phys. 23, 426 (1972).
14. McClellan, R. O., Boyd, H. A., Benjamin, S. A., Cuddihy, R. G., Hahn, F. F., Jones, R. K., Mauderly, J. L., Mewhinney, J. A., Muggenburg, B. A. and Pfleger, R. C., Health Phys. 23, 426 (1972).
15. P. E. Morrow (Editor), Health Physics 12, 173 (1966).

## BIOLOGICAL MODELING FOR PREDICTING RETENTION PATTERNS OF INHALED CONTAMINANTS\*

R. G. Cuddihy, W. C. Griffith and B. B. Boecker

Lovelace Foundation for Medical Education and Research  
Albuquerque, New Mexico 87108

### Abstract

Biomathematical models were developed for simulating the tissue distribution and clearance of inhaled Ba and Ce aerosols in Beagle dogs. Incorporating physical size characteristics of particles for predicting deposition and chemical solubility for estimating systemic absorption, application of these models was studied over a broad range of aerosol forms. The results illustrate the importance of determining these aerosol characteristics to aid in developing well founded predictions for its biological behavior. Procedures developed to date were more reliable in describing Ba metabolism and illustrate the more complex biochemical nature of Ce retention.

### Introduction

Retention of inhaled particulate material after deposition in the respiratory tract is of vital interest to those concerned with radiation protection in nuclear industries and those concerned with other areas of industrial and environmental health. Inhaled particles are retained for minutes to hundreds of days depending upon physical size, chemical composition, solubility and respiratory patterns during inhalation. Inhalation exposures of humans to toxic materials may result from single acute, intermittent or chronic environmental contamination at relatively constant or highly variable air concentrations. All of these factors profoundly influence the doses received by the respiratory tract and other organs. Thus, some rapid accounting system which uses physical, chemical and biological factors is necessary for managing inhalation exposure data to provide time integrated tissue dosimetry information.

Dosimetry problems are simplified when whole body or tissue accumulations of the toxic materials can be measured with confidence. Whole-body counting and isotope distribution scanning after inhalations of radioactive materials with relatively high energy gamma or beta emissions are routine, however, difficulties arise when nonradioactive or very weak beta-gamma or alpha emitting isotopes are inhaled. Then, the only certain information which can be easily obtained relates to the air concentration, general chemical form of the aerosol and material recovered in excreta. All data can add to the evaluation of possible consequences of an exposure if a versatile framework is developed for relating information. One method is the use of mathematical models of the biological behavior of inhaled materials and their solution with readily available techniques. These models can be developed from experimental animal studies; however, since aerosols inhaled in accidents are likely to differ chemically and physically, simple adjustments must be provided.

Models of radionuclide metabolism have been studied in Beagle dogs to simulate inhaled radioactivity cleared from the respiratory tract to other organs as first order compartmental exchanges. The initial conditions include

\* Research performed under U. S. Atomic Energy Commission Contract No. AT(29-2)-1013 and conducted in facilities fully accredited by the American Association for Accreditation of Laboratory Animal Care.

aerosol particle size, air concentration, respiratory rates and an estimation of the solubility of the aerosol. Applying the models to human exposures requires adjusting transfer rates for species differences, variations in chemical form and deposition pattern of the particular exposure. Adjustments can be made during early stages of an evaluation and refined as data become available. In this way, a unified picture of an exposure can be developed and long-term evaluation facilitated especially with regard to prospective therapy.

### Biomathematical Model

A biomathematical model used in analyses of inhalation studies of  $^{140}\text{Ba}$ ,  $^{140}\text{La}$  and  $^{144}\text{Ce}$  in Beagle dogs is in Fig. 1. The respiratory tract is divided into the nasopharynx, tracheobronchial tree and pulmonary regions as defined by the Task Group on Lung Dynamics.<sup>1</sup> Aerosol deposition in these compartments depends on its physical characteristics. These regions as well as the stomach, small intestine and tracheobronchial lymph nodes are divided into two or more parallel compartments leading into a compartment in series. The parallel compartments represent inhaled material in a particular state which may be converted by dissolution or other change in chemical form and pass into a transformed state, represented by the series compartment, from which absorption into the circulation may occur. The number of parallel compartments in these organs corresponds to the number of first-order exponential functions required to represent the dissolution mathematically. The rate constants of the exponential functions were used for the corresponding compartmental transfer rates, and coefficients of the exponential functions were used to apportion the material deposited in each region among the parallel compartments. Mechanical clearance of deposited material is shown by solid arrows in Fig. 1 to avoid confusion and represents transfers between corresponding subunits of each organ configuration. All internal organ exchanges of ionic or molecular forms with blood and excretory pathways, shown as individual arrows were used with a constant fraction of the compartmental content transferring per unit time interval. Transfer of material from blood to small intestine indicates a possible association with liver and biliary secretion, however, sufficient information on this is not available.<sup>3</sup> Analog and digital computer programs were developed for simulation of the model.

### Retention of Inhaled Barium Aerosols

In previously reported experiments, Beagle dogs inhaled differing chemical forms of barium:  $^{133}\text{BaCl}_2$ ,  $^{133}\text{BaSO}_4$ , heat treated  $^{133}\text{BaSO}_4$  and  $^{133}\text{Ba}$  in fused clay.<sup>3</sup> Respiratory tract retention of  $^{133}\text{Ba}$  was measured for 16 days and then the dogs were sacrificed for tissue analyses. An additional group of dogs inhaled aerosols of  $^{133}\text{Ba}$  in fused clay and was studied for 512 days. Samples of the aerosols from the exposures were enclosed between 25  $\mu\text{m}$  cellulose acetate membrane filters and dissolved in a simulated serum solvent.<sup>4</sup> The aerosol samples were placed in 100 ml of solvent which was refreshed periodically to measure filter retention of  $^{133}\text{Ba}$  as

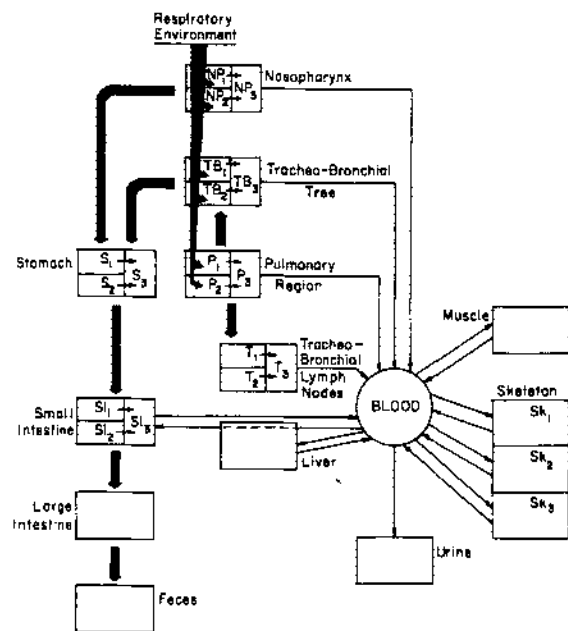


Figure 1. Biomathematical model for describing the retention of inhaled aerosols. Intercompartmental transfers of material as indicated by arrows were represented by first order rate constants.



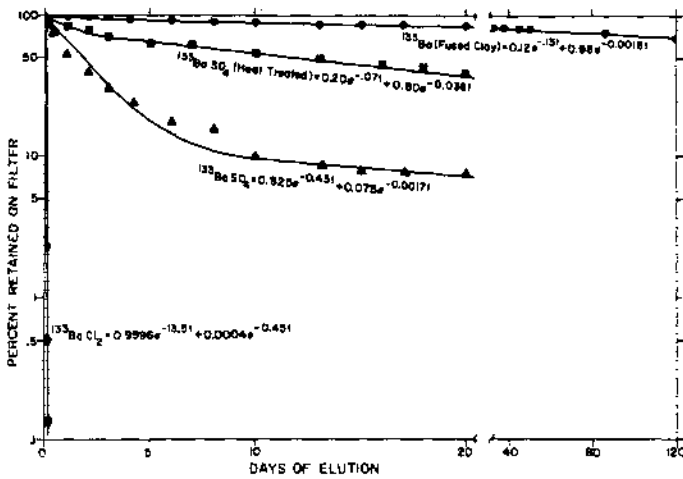


Figure 2. Retention of  $^{133}\text{Ba}$  on aerosol filter samples subjected to dissolution in static contact with serum simulant.

shown in Fig. 2. Retention half-lives ranged from 1.2 hr for  $^{133}\text{BaCl}_2$  to more than 430 days for  $^{133}\text{Ba}$  in fused clay. Respiratory tract clearance and skeletal accumulation of  $^{133}\text{Ba}$  following inhalation of  $^{133}\text{BaSO}_4$  is shown in Fig. 3. Solid lines are the predictions of the model using transfer constants in Table 1 and initial deposition fractions in Table 2. Comparisons of the observed and predicted total body, lung, skeletal and tracheobronchial lymph node activities at sacrifice are also in Table 2 for all aerosols of  $^{133}\text{Ba}$ . In general, the predicted tissue activities at sacrifice were sufficiently similar to observed activities to be useful in dosimetric projections for barium aerosols.

#### Retention of Inhaled Cerium Aerosols

The relationship between *in vivo* and *in vitro* solubility was also studied with aerosols of cerium:  $^{144}\text{CeCl}_3$ ,  $^{144}\text{CeCl}_3$  in  $\text{CsCl}$  ( $\text{Ce}:\text{Cs} \approx 1:10$  by mass) and  $^{144}\text{Ce}$  in fused montmorillonite clay. Particle sizes were about  $2 \mu\text{m}$  AMAD for all aerosols. Dissolution was studied in different solvents including 0.001 M citric acid (pH 3.3), 0.001 M acetic acid (pH 3.9), saline (pH 6.9) and serum simulant (pH 7.3 to 7.9) under static solvent conditions as described previously. A parallel flowing solvent system<sup>4</sup> was also used with serum simulant at pH 7.3 stabilized in an atmosphere of 5%  $\text{CO}_2$  and 95%  $\text{N}_2$ . All dissolution studies were conducted at  $37^\circ\text{C}$ . Typical retention patterns of  $^{144}\text{Ce}$  on the filter samples are in Fig. 4 along with the respiratory tract retention of  $^{144}\text{Ce}$  in Beagle dogs after inhalation of  $^{144}\text{CeCl}_3$  in  $\text{CsCl}$  with  $\text{Ce}:\text{Cs} \approx 1:10$  by mass.<sup>5</sup> Dissolution of Ce aerosols was greatly influenced by solvent composition. Filter retention after 4 days ranged between 0.5% in citric acid and 80 to 99% in serum simulant. The higher pH and presence of phosphate, sulfate and carbonate species in serum simulant were likely responsible for slower aerosol dissolution. Clearance of  $^{144}\text{CeCl}_3$  from the respiratory tracts of Beagle dogs had an intermediate

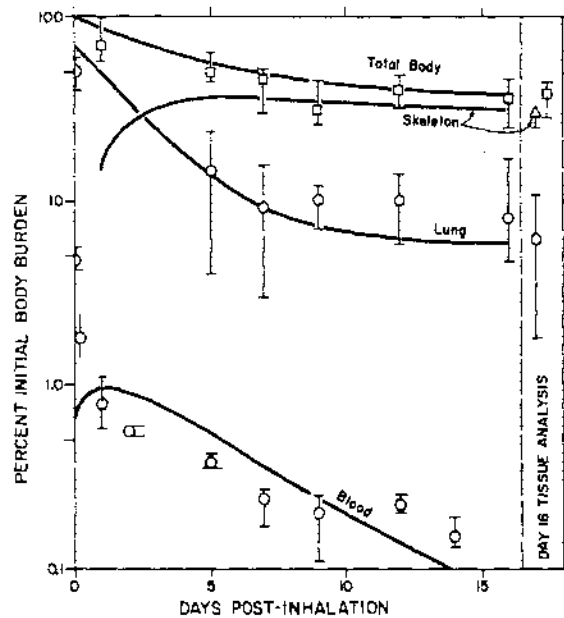


Figure 3. Retention and tissue distribution of  $^{133}\text{Ba}$  following inhalation of  $^{133}\text{BaSO}_4$  by Beagle dogs.

Table 1  
 First Order Kinetic Constants Applied to Model Shown in Fig. 1  
 for the Description of Ba and Ce Retention Following Inhalation

Origin	Compartment Of Destination	Transfer Rates (fraction/day)	
		Ba	Ce
Nasopharynx	Stomach	25	25
Nasopharynx*	Blood	100	100
Trachea and Bronch.	Stomach	0.9	0.9
Trachea and Bronch.**	Blood	100	100
Pulmonary	Stomach	≈ 0	.0009
Pulmonary*	Blood	100	100
Stomach	Sm. Intestine	15	15
Sm. Intestine	Lg. Intestine	5	5
Lg. Intestine	Feces	0.7	0.7
Pulmonary	TB Lymph N.	0.0001	0.0001
TB Lymph N.*	Blood	100	100
Sm. Intestine	Blood	0.35	≈ 0
Blood	Sm. Intestine	4	8.5
Blood	Urine	2.5	7
Blood	Liver	1.3	11.5
Liver	Blood	3.2	≈ 0
Blood	Muscle	40	≈ 0
Muscle	Blood	10	≈ 0
Blood	Skeleton (Sk <sub>1</sub> )	16.5	9
	(Sk <sub>2</sub> )	2.4	---
	(Sk <sub>3</sub> )	2.4	---
Skeleton (Sk <sub>1</sub> )	Blood	0.65	≈ 0
(Sk <sub>2</sub> )		0.014	---
(Sk <sub>3</sub> )		0.0007	---

Origin	Destination	Aerosol	Transfer Rates
NP <sub>1</sub>	NP <sub>3</sub>	BaCl <sub>2</sub>	13.5
TB <sub>1</sub>	TB <sub>3</sub>	BaSO <sub>4</sub>	0.43
P <sub>1</sub>	P <sub>3</sub>	BaSO <sub>4</sub> (heat treated)	0.70
S <sub>1</sub>	S <sub>3</sub>	Ba-fused clay	0.13
SI <sub>1</sub>	SI <sub>3</sub>	Ce-fused clay	0.34
T <sub>1</sub>	T <sub>3</sub>		
NP <sub>2</sub>	NP <sub>3</sub>	BaCl <sub>2</sub>	0.45
TB <sub>2</sub>	TB <sub>3</sub>	BaSO <sub>4</sub>	0.0017
P <sub>2</sub>	P <sub>3</sub>	BaSO <sub>4</sub> (heat treated)	0.0038
S <sub>2</sub>	S <sub>3</sub>	Ba-fused clay	0.0016
SI <sub>2</sub>	SI <sub>3</sub>	Ce-fused clay	0.0005
T <sub>2</sub>	T <sub>3</sub>		
P <sub>1</sub>	P <sub>4</sub> **	CeCl <sub>3</sub> in CsCl	0.65
P <sub>2</sub>	P <sub>4</sub>		0.018
P <sub>3</sub>	P <sub>4</sub>		0.001

\* Represents transfer from dissolved or transformed state to blood only.

\*\* Three parallel compartments were used for CeCl<sub>3</sub> in CsCl and the same rate constants were used for NP, TB, S, SI and T compartment configurations.

Table 2

Physical Characteristics and Deposition Fractions of Aerosols Used in Inhalation Studies with Beagle Dog Tissues Activities at Time of Sacrifice

Aerosol	BaCl <sub>2</sub>	BaSO <sub>4</sub>	Heated BaSO <sub>4</sub>	Ba in Fused Clay	CeCl <sub>3</sub> in CsCl	Ce in Fused Clay
AMAD*	2.3	1.0	0.9	2.2	2.2	1.4 - 2.7
<u>Regional Deposition as Fractions of Total Body Deposition</u>						
NP <sub>1</sub>	.45	.14	.03	.05	.05	44
NP <sub>2</sub>	0	.01	.12	.40	.40	4
NP <sub>3</sub>	---	---	---	---	---	2
TB <sub>1</sub>	.10	.09	.02	.01	.01	8.6
TB <sub>2</sub>	0	.01	.08	.09	.09	.8
TB <sub>3</sub>	---	---	---	---	---	.4
P <sub>1</sub>	.45	.69	.15	.05	.05	15
P <sub>2</sub>	0	.06	.60	.40	.40	3
P <sub>3</sub>	---	---	---	---	---	2

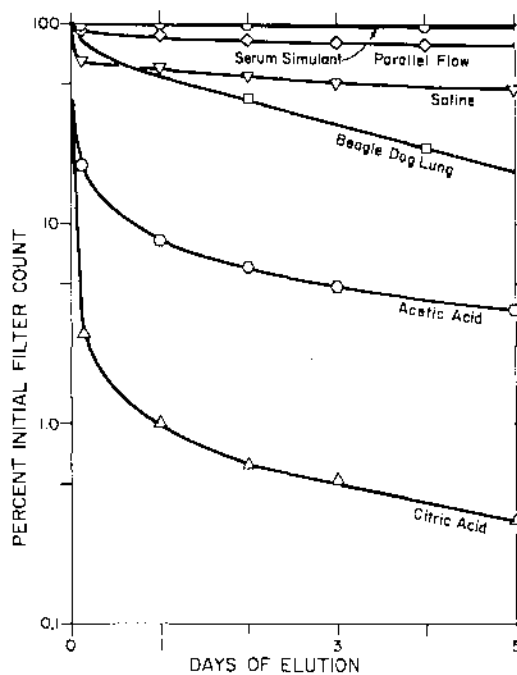
Tissue Activities and Fractions of Initial Body Burdens

Sacrifice Time (days)	16	16	16	16	512
<u>Observed</u>					
Lungs	---	.06	.20	.37	.19
Skeleton	.45	.30	.30	.008	.036
TBLN	---	---	---	---	.31
Total Body	.45	.36	.50	.38	.24
<u>Predicted</u>					
Lungs	---	.06	.32	.40	.16
Skeleton	.30	.31	.20	.025	.06
TBLN	---	---	---	---	.009
Total Body	.30	.37	.52	.43	.23

See Fig. 5 for sacrifice times and tissue activities

\* Activity Median Aerodynamic Diameter

Figure 4. <sup>144</sup>Ce retention in <sup>144</sup>CeCl<sub>3</sub> aerosol samples in static contact with various solvents. Data for filter retention of <sup>144</sup>Ce in a system with parallel solvent flow and respiratory tract retention of inhaled <sup>144</sup>Ce by Beagle dogs were studied with aerosols of <sup>144</sup>CeCl<sub>3</sub> in CsCl.



clearance rate, however, the exact relationship of the aerosols used for the *in vitro* and *in vivo* studies is not known.

The biomathematical model was used to simulate data from the studies of the retention of inhaled  $^{144}\text{CeCl}_3$  and  $^{144}\text{Ce}$  in fused clay in Beagle dogs using transfer rates in Table 1. A comparison is in Fig. 5. A reasonable fit was obtained, however, dissolution of chloride aerosols in the respiratory tract as used in these simulations was considerably more than observed *in vitro* except with dilute solutions of citric or acetic acid.

Dissolution of  $^{144}\text{Ce}$  in fused clay aerosols had a half-life of 450 days in saline during the first 25 days and a half-life of 200 days in serum simulant. Both of these dissolution rates include more rapidly soluble fractions of the aerosol samples normally observed at early times. Dissolution rates would probably have declined after longer times, however, the half-lives for clearance from the respiratory tracts of dogs were 1440 days for 90% and 2 days for 10% of the deposited aerosol.

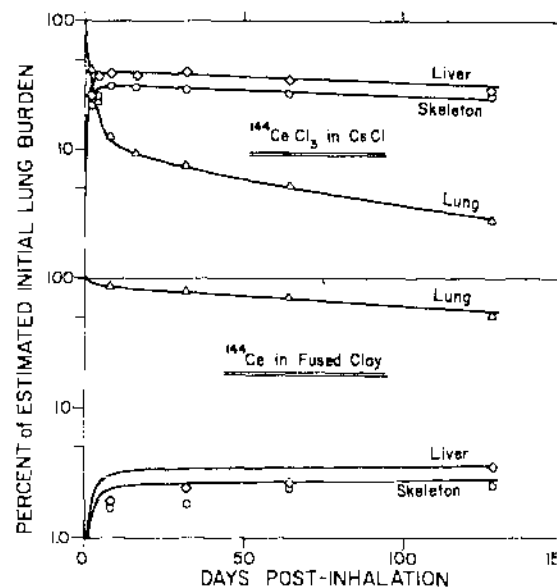


Figure 5. Tissue retention of  $^{144}\text{Ce}$  in Beagle dogs after inhalation of aerosols of  $^{144}\text{CeCl}_3$  and  $^{144}\text{Ce}$  in fused clay.

#### Summary

In general, mathematical models incorporating *in vitro* measurements of aerosol solubility facilitate complex analyses of inhalation exposures, however, exact correspondence between dissolution rates of specific aerosols *in vivo* and *in vitro* should not be accepted without other supporting evidence. Such methods are likely to aid in distinguishing very soluble from moderate and insoluble aerosols with further study in the selection of solvents with aerosols for trivalent and tetravalent elements. Studies with serum simulant solvent predicted very long respiratory tract retention of  $^{144}\text{CeCl}_3$  whereas elution of  $^{144}\text{Ce}$  in dilute solutions of acetic and citric acid or saline more closely resembled the clearance of  $^{144}\text{Ce}$  in Beagle dogs. This discrepancy was not observed with inhaled aerosols of barium, perhaps due to the more soluble nature of barium in most aqueous solutions.

**Acknowledgements** - The authors are grateful to members of the Department of Radiobiology and Veterinary Medicine of the Lovelace Foundation, Dr. Patricia Durbin of the Lawrence Berkley Laboratory and Mr. Fred Ruppert for assistance in developing this paper.

#### References

1. International Commission on Radiological Protection, Task Group on Lung Dynamics, 1966, (Oxford: Pergamon Press) and *Health Phys.*, 12, 173.
2. Ballou, J. E. and Hess, J. O., 1972, *Health Phys.*, 22, 369.
3. Cuddihy, R. G., Hall, R. P. and Griffith, W. C., Inhalation Exposures Barium Aerosols: Physical, Chemical and Mathematical Analysis, submitted for publication to *Health Physics*.
4. Kanapilly, G. M., Raabe, O. G., Goh, C.H.T. and Chimenti, R. A., 1973, *Health Phys.*, 24, 497.
5. Boecker, B. B. and Cuddihy, R. G., Toxicity of  $^{144}\text{Ce}$  After Inhalation  $^{144}\text{CeCl}_3$  by Beagle Dogs: Metabolism and Dosimetry, to be submitted for publication.

DEPOSITION AND ELIMINATION OF IRON OXIDE  
AEROSOL FROM THE LUNG OF RATS:  
COMPARISON WITH ICRP PREDICTIONS FOR MAN

P. J. Hewitt  
Radiation Protection Service  
University of Bradford England

Abstract

Male albino rats have been exposed to iron oxide aerosol (MAD 0.3  $\mu\text{m}$ ) at iron concentrations up to 700  $\text{mg}/\text{m}^3$  for continuous periods up to 235 min, and for intermediate periods of 30 min over a ten day period. From 0.3-3.0 mg of deposited iron were found in the rat lungs removed one day post exposure which corresponds to initial pulmonary fractional depositions of between 0.3 and 0.7 - which is comparable to that proposed for man by the ICRP Lung Dynamics Task Group (0.35). The elimination of deposited iron from the rat lung can be described by a two component exponential expression with parameters similar to those of ICRP for man except that the phase two clearance in rats was found to be shorter ( $T_{1/2} = 33\text{d}$ ).

Introduction

The ICRP Lung Dynamics Task Group derived curves for man relating the aerodynamic diameter of particulates to the fractional deposition factor for the nasal, tracheo bronchial and pulmonary regions<sup>1</sup>. Opportunities for testing these curves with human subjects are restricted due to the practical difficulties involved in long term exposures to high concentrations of aerosol, and the various analytical limitations. Nevertheless some useful data has been gained following relatively short term exposures using radio-tracer purposefully administered or resulting from unplanned exposure<sup>2</sup>. Rats have been used in this present study, which could be exposed for up to five hours in a controlled fashion to relatively high concentrations of a series of metal oxides of interest.

The research programme is directed to the assessment of the hazard to health of industrial workers exposed to metallic fumes having a median aerodynamic diameter (MAD) below 1  $\mu\text{m}$ . Metallic particulates of this size range are generated in a number of common industrial processes, such as welding and metal grinding, and are believed to be toxicologically significant<sup>3</sup>. The ICRP deposition curve for man indicates that about a quarter of airborne particulates of 1  $\mu\text{m}$  MAD will be initially deposited in the pulmonary region (fractional deposition  $\sim 0.25$ ) decreasing to a fractional deposition of  $\sim 0.1$  at 5  $\mu\text{m}$  MAD, as the larger particulates are deposited in the nasal and tracheo bronchial region. Below 1  $\mu\text{m}$  diameter the pulmonary curve shows a steep rise so that at 0.3  $\mu\text{m}$  MAD for example fractional deposition of 0.35 are predicted. Experimental verification of this portion of the curve is clearly of considerable interest in the assessment of the health hazard of metallic fumes. Of equal importance are investigations into the clearance kinetics of particulates initially deposited in the pulmonary region.

## Exposure to Iron Oxide Particulate

Male albino rats were exposed to iron (MAD 0.3  $\mu\text{m}$  geometric standard deviation 1.8) in a specially constructed chamber using experimental techniques described elsewhere<sup>4</sup>. The concentration of fume throughout each exposure period was determined by collection of the total particulate flowing through the chamber and measuring the air volume throughput. Electron microscopy was used for the particle size measurement.

Both continuous and intermittent exposures were used (Table 1) Rats were killed one day post each exposure and also at several predetermined periods up to 100 d post exposure in order to follow the elimination of deposited iron from the lung. Control rats were housed alongside and then killed at the same time as those exposed; the total lung being then removed and iron was determined by instrumental (Ge/Li) neutron activation analysis. A number of blood and body tissue samples were also analysed.

Table 1. Exposure Conditions and Deposition Factors for Rats exposed to Iron Oxide Aerosol

Exp	Duration (min)	Fe conc of inhaled air ( $\text{mg}/\text{m}^3$ )	Fe deposited in lung 1d post exp (mg)	Fractional deposition at t=0 (D)
1	16	392	0.35	0.50
2	30	250	0.60	0.72
3	Intermittent (Table 3)		3.21 3.12	0.35
4	235	143	1.45 1.21	0.46 0.38

## Results and Discussion

The lung burden was calculated using the total lung weight and the iron concentrations of the exposed rat's lung corrected for iron in control rat lung. The shape of the pulmonary elimination curve can be expressed by an exponential expression of the form

$$R = A_s e^{-\frac{0.693t}{T_{s\frac{1}{2}}}} + A_1 e^{-\frac{0.693t}{T_{l\frac{1}{2}}}}$$

where R is the % of the initial lung burden retained at time t,  $T_{s\frac{1}{2}}$  and  $T_{l\frac{1}{2}}$  are short term and longer term elimination half times,  $A_s, A_1$  are constants.

Substituting values for  $A_s, A_1$  and the corresponding elimination half times  $T_{s\frac{1}{2}}$  and  $T_{l\frac{1}{2}}$  taken from ICRP data<sup>1</sup> the elimination curve for man<sup>2</sup> can be compared with that determined for the rat (Fig 1).

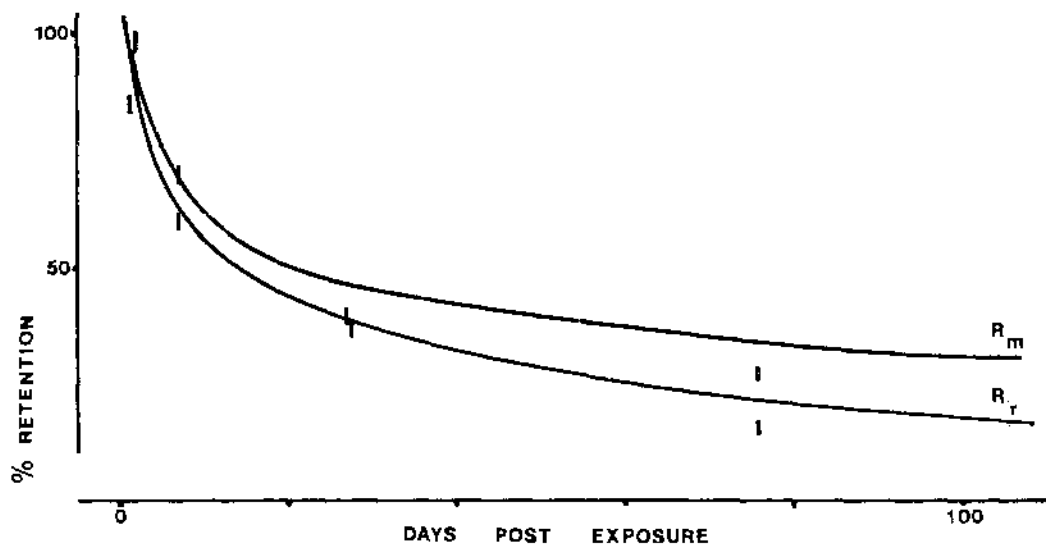


Fig 1 RETENTION CURVES FOR MAN ( $R_m$ ) AND RAT ( $R_r$ )

The experimental data obtained in this present study are insufficient to be definitive, but are compatible with a similar short term ( $T_{1/2}=1d$ ) elimination in both man and the rat, although the longer term elimination  $T_{1/2}$  for the rat is shorter ( $\sim 33d$ ) than that proposed by ICRP for man ( $T_{1/2}=70d$ ). The ICRP curve is based upon a two phase exponential clearance model, comprising a short phase I ( $T_{1/2}=24h$ ) and a phase II clearance of  $T_{1/2}=70d$ ). The slower clearance represents that fraction ( $\sim 40\%$ ) of the initial pulmonary deposited iron oxide translocated to the gastro-intestinal tract, through a process which depends upon endocytosis and ciliary mucus transport, and in addition the fraction that is translocated to the blood - according to the ICRP model some 15% of the initial pulmonary iron oxide deposit would be taken into the systemic blood directly whilst a fraction ( $\sim 5\%$ ) would follow a lymphatic drainage route.

A contribution to the blood may also be postulated following gastro-intestinal tract absorption, but in the case of iron oxide this will be negligible. In order to test this, blood, liver and kidney samples were removed from pairs of rats one day post exposure in both experiments 3 and 4, and also from an equivalent number of control rats, and all samples were analysed for iron. The contribution of inhaled iron oxide to the blood and to organs other than lung tested proved to be insignificant. This confirms the observations of Albert and Arnett<sup>5</sup> on man and also Morrow et al<sup>6</sup> on dogs, who found no detectable radioactivity in blood following inhalation studies with  $^{59}Fe$  labelled iron oxide, Morrow concluding that iron oxide behaves as an 'insoluble' material being efficiently eliminated from the gastro-intestinal tract in the faeces.

The very long term fate of pulmonary deposited iron oxide is probably inadequately described by a two component exponential expression: the rats were studied in this work for an insufficiently long period post exposure to define such components, and other

workers<sup>5,6</sup> terminated their measurements 70d post exposure. However the retention of iron and other elements occupationally ingested many years previously is clearly demonstrated in the results of human lung analysis (Table 2). The 'hard metal' workers were engaged in a process involving the risk of inhalation of cobalt, tantalum and tungsten, but clearly the iron concentration in the boiler maker's lung is of particular interest, being clearly indicative of long term retention since the sample was taken five years post exposure.

Table 2. Analysis Results on Lung of Controls and Industrially Exposed Metalworkers\*

	Hard Metal Workers				Boiler/ Welder	Controls	
	1	2	3	4		1	2
Co	1.0	2.6	2.0	19.2	3.9	1.0	0.9
Cr	1.0	1.7	1.7	10.6	1.6	0.4	0.3
Fe	1.0	1.1	0.7	1.7	16.6	0.7	0.8
Ta	1.0	-	0.5	1.8	-	-	-
W	1.0	26.0	48.0	130.0	-	-	-

\* All results are compared to HM 1, taking the concentration of each element as unity in that sample.

Fractional deposition factors can be calculated from the expression

$$D = \frac{\text{wgt of deposited particulate in lung at } t = 0}{(\text{air conc})(\text{duration})(\text{respiration rate})(\text{tidal vol})}$$

Casarett<sup>7</sup> reports deposition factors of 0.49-0.67 (mean 0.60) for iron oxide particulates (0.068 CMD  $\sigma_g$  1.62), these calculations being based on minute volumes of 75cm<sup>3</sup>. In general the rats used in this present study had minute volumes of 120cm<sup>3</sup> and the calculated deposition factors for the 0.3  $\mu$ m iron oxide were lower (Table 1), being very similar to the 0.35 predicted from ICRP for man. Because of the dimensional differences it is not unexpected that for relatively large diameter particulates (>5 $\mu$ m) there should be greater deposition in the nasal and upper respiratory tract of the rat than in man, but the effect of anatomical difference on <1 $\mu$ m particulates is less clear, and in this study the deposition in rats could not be differentiated from man. Thomas<sup>8</sup> found that for <sup>137</sup>Cs in rats the total body deposition plotted against particle size followed a similar trend to that of the ICRP curve for man but at about half the deposition level - the deposition factors at the lowest particle size studied (~0.3 $\mu$ m MAD) being in all cases lower than that found for iron oxide in this work.

Thomas<sup>9</sup> identified in rats an intermediate clearance phase due to the anatomical structure of the upper respiratory tract



which because of its downward slope towards the mouth could be expected to enhance the clearance rate compared to man. This could partly account for the phase II elimination half time observed for rats in this study ( $T_{1/2}$  33d) compared to that for the dog ( $T_{1/2}$  62d)<sup>6</sup> and man ( $T_{1/2}$  70d)<sup>1</sup>.

Table 3. Daily Contribution to Lung Burden during Intermittent Exposure (Experiment 3)

Day	Duration (min)	Fe conc of inhaled air ( $\text{mg}/\text{m}^3$ )	Initial pulmonary* deposition (mg)	Remaining at <sup>e</sup> T = 11 (mg)
0	33	378	0.52	0.25
1	31	450	0.59	0.28
2	30	402	0.51	0.26
3	30	367	0.46	0.24
4	32	524	0.70	0.37
5				
6				
7	30	583	0.73	0.42
8	29	664	0.81	0.50
9	26	599	0.65	0.44
10	32	622	0.84	0.66

Total 3.42

cf Table 1(3.21, 3.12)

\* Calculated from (air conc)(duration)(min vol)(0.35)

<sup>e</sup> Calculated using elimination equation

Using the two component elimination equation and the deposition factor which for the purpose of illustration is taken as that of the ICRP (0.35), the daily contributions of iron to the rat's lungs are easily calculated, and the total can then be compared with the actual iron content of the lungs found at autopsy (Table 3).

### Conclusion

This preliminary study of the iron oxide inhalation and elimination has indicated the value of the ICRP Task Group approach. For the particle size examined, the deposition factors found for the rat were similar to those proposed by ICRP for man. Pulmonary clearance could be reasonably described in terms of a two component exponential but the phase II component was of shorter half time than that in man.

### Acknowledgements

The assistance of Dr. R. Hicks, School of Pharmacology, with the rat exposures is gratefully acknowledged.

### References

1. ICRP Task Group on Lung Dynamics (1966) Health Physics 12, p173.
2. Rundo, J. (1964) Health Physics and Medical Division Progress Report, AERE-PR/HPM 5 (U).
3. NASA CR-72492. (1966) "Survey and analysis of health hazards resulting from ultrafine metal and metal oxide powders at the NASA Lewis Research Center, Cleveland, Ohio".
4. Hewitt, P.J. and Hicks, R. (1972) Nuclear Activation Techniques in the Life Sciences. IAEA, Vienna. p219-232.
5. Albert, R.E. and Arnett, L.C. (1955) Arch. Ind. Health, 12, p99-106.
6. Morrow, P.E., Gibb, R.F. and Johnson, L. (1964) Health Physics, 10, p543-555.
7. Casarett, L.J. and Epstein, B. (1966) Amer. Ind. Hyg. Assoc. J., 27, p533-538.
8. Thomas, R.L. (1969) Health Physics, 16, p417-428.
9. Thomas, R.G. (1972) Assessment of Airborne Particles. (Editors, Mercer, T.T. et al) C. C. Thomas, Springfield, Illinois. p405-420.

## INHALATION HAZARDS: THEY COULD BE WORSE\*

R. G. Thomas  
Lovelace Foundation for Medical Education and Research  
5200 Gibson Blvd., SE  
Albuquerque, New Mexico 87108

### Abstract

The inhalation route of entrance of relatively insoluble radioactive toxic particles to the body may afford several mechanisms that act to reduce the potential radiation hazard, as compared to entrance by other possible routes. A relatively minor fraction of what is inhaled actually reaches the pulmonary spaces of the lung. In addition, those particles that do reach this region of the respiratory tract have the opportunity of being cleared by at least three methods; muco-ciliary movement, dissolution and subsequent removal by the blood, and translocation to tracheobronchial lymph nodes. With relatively insoluble particles all of these factors appear to work toward helping the individual to reduce the radiation insult. The methods proposed to bring about this reduction through these various factors are discussed in general terms.

### Introduction

The inhalation route of entry of radionuclides to the body generally represents the most prevalent hazard in incidents involving accidental releases that may occur in the nuclear industry. Radioactive materials that become airborne are likely to do so in a variety of physico-chemical states, depending upon the industrial operation. The temperature of release, for example, may play an important role in the subsequent behavior of the nuclide-containing substance once it is deposited in the respiratory tract. The chemical or physical form of the vehicle or matrix containing the radionuclide may play a similarly important role in the subsequent metabolism of the inhaled material. Thus, the practical aspects of concern in the evaluation of potential inhalation hazards involve an explicit knowledge of the conditions under which releases may occur. This point has been particularly emphasized recently by a number of authors investigating the factors involved in the increased incidence of lung cancer in miners and workers in other dusty trades.<sup>1</sup>

This paper explains the factors involved in assessing the relative potential hazards following human inhalation exposure and discusses many areas in which the body appears to aid in reducing the radiation risk to the body tissues.

---

\* Research performed under U. S. A. E. C. Contract AT(29-2)-1013 and conducted in facilities accredited by the American Association for Accreditation of Laboratory Animal Care.

## Lung Deposition

The Task Group on Lung Dynamics (TGLD) of Committee 2, the International Commission on Radiological Protection (ICRP) has separated the human respiratory tract into three major regions.<sup>2</sup> These are the nasopharyngeal (NP) region, beginning with the anterior nares and descending to the level of the larynx; the tracheobronchial (TB) region, continuing through the trachea and bronchial tree and including the terminal bronchioles; and the pulmonary (P) region consisting of the remainder of the respiratory tract, beginning with the respiratory bronchioles and including the alveoli. In general, larger particles deposit in the NP and TB regions and, if relatively insoluble in body fluids, soon (within 48 hours) find their way to the gastrointestinal tract to be eliminated by fecal excretion. According to the TGLD, the quantity deposited in these two areas combined might vary from a few percent up to essentially 100% of inhaled particulate material. Deposition in the TB region rarely exceeds 10% of the total inhaled aerosol. The NP region shows the greatest variation in deposition as a function of particle size, with deposition greater at the larger particle sizes and increasingly reduced as the aerodynamic particle diameters become smaller than 1  $\mu\text{m}$ . Any fraction of an inhaled aerosol of an insoluble nature that deposits in these two regions is acted upon in favor of defense against the toxicant, thus protecting the individual from depositing large quantities of inhaled particles in the pulmonary spaces.

This same argument may be used in discussing the pulmonary (P) region with regard to deposition of inhaled materials. Because of the large fractions inhaled that are deposited in the upper respiratory tract, plus the sizeable fraction that is exhaled and not deposited in any part of the tract, deposition in the P region is comparatively low. Only when the particle size is very small (less than 0.5  $\mu\text{m}$  aerodynamic diameter) does deposition in this region surpass 25% of the amount inhaled, as estimated by the TGLD.<sup>2</sup> Inasmuch as most practical aerosols are of a size greater than this, the body is afforded considerable protection from an inhaled, relatively insoluble substance.

The exception to these protective factors arises when inhaled particles are of a soluble nature. As an example, a recent study has shown that soluble substances deposited on the mucosa of the NP region of the Syrian hamster entered the blood at a rather rapid rate.<sup>3</sup> These studies indicated a nasal absorption of >50% for the chlorides of strontium, barium and cesium but less than 4% for the trivalent cerium. As is often the case, trivalent cations tend to form rather insoluble ligands with proteins and other biological molecules. The report indicates that absorption from the NP region can be at least as great as from gastrointestinal absorption for a given substance. Thus, the radiobiological effects from a very soluble particulate material following inhalation can be afforded little alleviation by the body forces, in that the radionuclide may enter the circulation very rapidly. Those substances that fall between the arbitrary soluble and insoluble categories present a complicated picture that will be discussed later.

## Pulmonary Retention

The major factor in considering inhalation hazards appears to be related much more to retention characteristics than to initial deposition. What is the likely fate of the inhaled fraction that is deposited in the pulmonary spaces? What lines of body defense favor the residence of a body burden of a given radionuclide to be in lung, as opposed to being initially deposited elsewhere as a consequence of a non-inhalation type of entry to the body? In one case the particle may be extremely soluble in body fluids and would act the same with

regard to localization sites and retention characteristics (metabolism) regardless of route of entry. In this instance the mode of assimilation is probably not very important. In another case, that of a relatively insoluble particle however, the particle may remain in the pulmonary spaces (alveolar region) and irradiate the surrounding tissue for a period of time dependent upon its effective half-life in that area. It may gradually dissolve and the radionuclide cation may enter the blood and either be excreted or translocated to the organ(s) most compatible with its chemical properties. The insoluble particle may also clear through the lymphatics to the regional (tracheobronchial) lymph nodes. It may also have the fortune of eventually being swept up the ciliated escalator, swallowed, and excreted in the feces. These are some of the factors that will be considered in terms of alterations, particularly reductions, in the potential radiation hazard after inhalation. The following remarks will be restricted to relatively insoluble particles deposited in the deep lung, unless otherwise noted.

#### Ciliary Removal:

It was mentioned that early clearance from the tracheobronchial region takes place through ciliary activity, in a matter of hours or days. This means of clearance is an important body mechanism for removing toxicant particles after accidental inhalation exposure. What of this method of clearance after the initial, rather large phase, has subsided? It is common knowledge that particles can be readily engulfed by macrophages following deposition in the alveolar region. It is also feasible and accepted that a small particle may reside on or within the surfactant lining of the lung, even perhaps prior to or following an engulfment by a phagocytic cell. What now is the fate of these particles? It makes sense that the mechanical movement of the lung alone creates some probability that the particle associated with surfactant, whether or not engulfed, will be swept upward via the muco-ciliary escalator and swallowed. In other words, it would essentially ride "piggy-back" on the normal processes of lung clearance. Such a probability may well be dependent upon the numbers of particles present that are available to be treated in this fashion, thus leading to an exponential (first order) loss with regard to the decrease in lung content with time.

This process of ciliary clearance from the pulmonary spaces is very important in clearing the lung of toxicants, including particulate material containing radionuclides. Without this process, regardless of the detailed mechanisms involved, the relative potential radiation hazard to the lung after inhalation would be considerably greater.

#### Dissolution of Particles:

All materials appear to be somewhat soluble in body fluids, and the fluids of the lung are no exception in the process. Mercer has emphasized the importance of solubilization of particles in removing materials (e.g., radionuclides contained in particles) from the deep lung.<sup>4</sup> The rate of dissolution of a particle in an erosive active medium is a function of the total amount of available surface area on that particle. Thus, the rate of removal of particulate material from the lung by this process is a function of the total surface area available to the fluids in the lung. In most cases this surface area is made up from millions of particles of all sizes, generally accepted as being log-normally distributed according to the number occurring at a given size. There are a few particles that are very large and which carry a great deal of radioactivity, compared to a large number of small particles, each containing relatively little radioactivity. Consider the dissolution of this size distribution, assuming all chemical characteristics of the particles to be the same.

The smaller particles may, in total, represent a considerable radiation source to the lung. These will dissolve much faster however, due to the much greater surface-to-mass ratios (surface area ÷ volume x density). The radionuclide cations released subsequently follow one of several pathways; (1) enter the blood and be excreted or localized in some other organ, (2) become associated into a chemical complex in the lung, (3) somehow find their way to the lymphatics or (4) somehow find their way up the muco-ciliary escalator. Of these possibilities, case 1 is the most likely route for the lone cation. In this case, the situation results in distributing the radioactivity to other organs in the body (often referred to hereafter as "internal organs"). Thus, as the dissolution process continues, more radionuclide accumulates internally or is excreted, primarily in the urine, and the initially higher dose rate to the lung is gradually reduced. The body, by this method, once again rises to the occasion by splitting the offensive lines and utilizing a means to dilute the total potential radiation dose to the body. With some other routes of entrance to the body, such as intravenous, the insult would be inflicted to two or three internal organs beginning almost immediately with no "reservoir" organ such as lung to dilute the attack. This would create a very high dose rate initially, a factor that may be very radiobiologically important. The slow migration from lung to the internal organs leads to a gradual build-up and simultaneous continuous loss from the tissues of localization, thus in most cases, never subjecting the internal organs to the larger dose rates. Such interaction between lung and radionuclides translocated to other organs has yet to be demonstrated experimentally to be less hazardous, and is no doubt dependent upon the chemical properties and physical half-lives of the materials involved. It would appear, however, that the inhalation route in this respect is somewhat favored in regard to being less hazardous for a given amount of radionuclide-containing material entering the pulmonary spaces as compared to some other route.

#### Translocation to Lymph Nodes:

Data are available from many sources that indicate a gradual concentration of a potentially toxic particulate substance in tracheobronchial lymph nodes following its entrance to the lung.<sup>5</sup> In most instances, the concentration (quantity/gram tissue) in these regional nodes surpasses that in lung at about 100 days post-commencement of exposure, regardless of whether this be single (acute) or chronic (repeated) inhalation exposure (Fig. 1). The material migrated to the nodes appears to be in particulate form, at least as can be discerned histologically and autoradiographically, depending upon the radionuclide and quantities involved.<sup>6</sup> Large accumulations of the particulate material may occur in the medullary areas, with little or no accumulation near the more peripheral germinal centers of the cortex. With alpha particle emitters, the particle appears to be localized such that the ionizing track length will not permit a release of large amounts (if any) of radiation energy to the germinal sites where lymphocyte production is manifested. Thus, large accumulations of alpha-emitting radionuclides in these nodes tend to irradiate the nutrient supply to the node (shut off the circulation), making it devoid of function after an extended period of time, but appear to act only to a minor degree directly on the cortical tissue, per se. The loss of nodal material under these conditions does not appear to be of grave consequence to the body in cases that have been experimentally observed.<sup>6</sup> In a long-term study involving inhalation by Beagle dogs of the alpha-emitter <sup>239</sup>Pu, large accumulations of the nuclide were found in the pulmonary lymph nodes.<sup>7</sup> In no case was there found a primary tumor in the lymphatic tissue of the nodes, but three cases of primary lesions of endothelial origin were reported. With beta particle emitters the length of the ionizing path is longer, and the extent (range) of damage is therefore greater. No primary tumors in the pulmonary

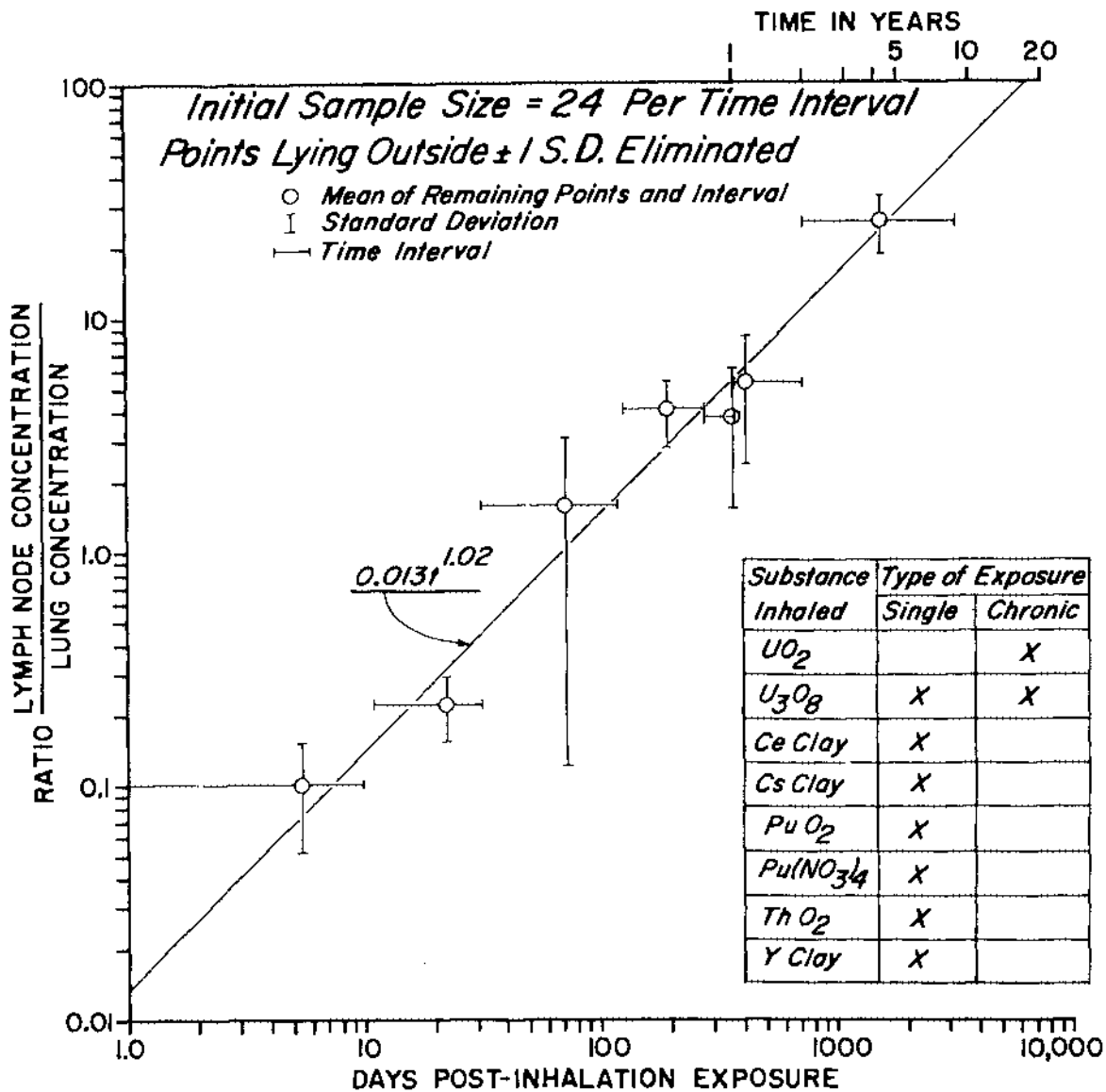


Fig. 1. Lymph node to lung concentration ratios following inhalation of various radionuclides by Beagle dogs. The total number of points (168) were segregated into groups of 24 and standard deviations (S.D.) calculated. All points outside  $\pm 1$  S.D. were eliminated from each group, a new mean and S.D. were calculated, and plotted, as shown. The time intervals spanning collection of each group of 24 points are shown, with the mean being plotted at the average time for the interval. All details concerning the individual experiments are reported elsewhere.<sup>5</sup>

lymph nodes have been reported, however, following inhalation of these type of emitters. It is as though these nodes were anatomically placed and physiologically devised to "clean-up" the more sensitive lung tissue and to, once again and emphatically, act as a means of effectively reducing the radiation hazard following inhalation.

## Therapy

The use of chelating (and other) agents to relieve the body of deposited radionuclides has been in practice for years; an excellent review has recently been published.<sup>8</sup> These chemicals have been used effectively to remove cations from bone, in particular the bone surfaces, and they act quite well for materials such as plutonium if administered soon after the nuclide reaches the bloodstream. The gross effect is one of increasing by quite sizeable amounts the quantity of radionuclide excreted in the urine; the basic fallacy is that the total reduction in internally deposited radionuclide is quite negligible. Following the inhalation route, however, there has been considerable recent evidence of significant reduction in lung burdens of radionuclide-containing particles by pulmonary lavage.<sup>9</sup> Through the process of flushing out the deep lung, alternating sides at intervals of a few days, as much as 50% of an initial lung burden may be removed. This is an order of magnitude better than the use of chemical agents such as chelators, in removing materials that may have been deposited internally by another route. In addition, for that material that leaves the pulmonary spaces for deposit in internal organs, as described earlier, one can also use the chelating agents quite effectively. The combined effect of lung washing (lavage) and DTPA (administered in the lavage fluid) has been recently described following the inhalation of relatively soluble  $^{144}\text{CeCl}_3$ , and the combination produced a "one-two" punch for removal of the inhaled cation.<sup>10</sup> When this combined treatment was used for an insoluble form of the same cation in fused clay particles, however, the DTPA appeared to be of little assistance in reducing the lung burden.<sup>11</sup> The lung washing technique may enhance entrance to the blood as well as performing its actual physical removal. The intravenous chelator then enhances excretion by sequestering the radionuclide as it enters the circulation.

The gross appearance from the inhalation route of entry is one of encouragement with regard to the ability to remove substantial quantities of deposited particulates. The fact that therapeutic removal of radioactive particles from the lung following inhalation can be accomplished to a degree, is an important factor in assessing relative radiological hazards as a function of route of entry to the body.

## References

1. Proceedings of the Conference on Morphology of Experimental Respiratory Carcinogenesis, P. Nettesheim, M. G. Hanna, Jr., and J. W. Deatherage, Jr., Editors, CONF-700501, December, 1970.
2. Task Group on Lung Dynamics, Committee 2, ICRP, P. E. Morrow, Chairman, Health Phys., 12, 173 (1966).
3. Cuddihy, R. G. and Jennie A. Ozog, Health Phys., in Press.
4. Mercer, T. T., Health Phys., 13, 1211 (1967).
5. Thomas, R. G., in Assessment of Airborne Particles, T. T. Mercer, P. E. Morrow and W. Stöber, Editors, Charles C. Thomas, Publisher, Springfield, Illinois, 1972, page 405.
6. Thomas, R. G., in Radiobiology of Plutonium, B. J. Stover and W. S. S. Jee, Editors, The J. W. Press, Salt Lake City, 1972, page 231.
7. Park, J. R., W. J. Bair and R. H. Busch, Health Phys., 22, 803 (1972).
8. Smith, V. R., Health Phys., 22, 765 (1972).
9. Pflieger, R. C., B. A. Muggenburg, D. H. Sesline, J. W. Harvey, R. G. Cuddihy and R. O. McClellan, Health Phys., 23, 595 (1972).
10. Muggenburg, B. A., R. C. Pflieger, R. G. Cuddihy and R. O. McClellan, Health Phys., 23, 611 (1972).
11. Boecker, B. B., B. A. Muggenburg and R. O. McClellan, Submitted for publication in Health Phys.



# LES PROBLEMES DE PROTECTION SOULEVES PAR LES RAYONNEMENTS NON IONISANTS

H. JAMMET

Commissariat à l'Energie Atomique  
Département de Protection  
Fontenay-aux-Roses (France)

## Abstract

Considering the growing utilization of non ionizing radiation and the spreading danger from workers to the general population, the nature, sources and conditions of exposure to non-ionizing radiations, their interaction with matter, their biological and pathological effects are reviewed and the present situation of protection as to regulations, operational principles, and monitoring is discussed at the international and national levels.

## Introduction

Le Comité Scientifique du congrès m'a fait l'honneur de me demander de vous exposer les problèmes de protection posés par les rayonnements non-ionisants. C'est la première fois, je crois, qu'une Organisation Internationale de Protection Radiologique inscrit ce sujet dans ses préoccupations.

Jusqu'à présent les rayonnements ionisants avaient accaparé leur attention toute entière. Cependant les rayonnements non-ionisants méritent qu'on examine attentivement les problèmes de protection qu'ils posent étant donné la puissance de certains générateurs, le développement de leur utilisation et l'extension des dangers du domaine professionnel au domaine public. Certains pays, tels les USA et l'URSS, ont notablement avancé dans ce domaine, la Communauté Economique Européenne s'en préoccupe et l'Organisation Mondiale de la Santé a créé des groupes de travail pour étudier ces problèmes. Je vais tenter de faire le point sur la situation actuelle, en m'efforçant de garder le juste milieu devant un auditoire hétérogène comprenant d'éminents spécialistes de la question, mais aussi beaucoup de personnes peu familiarisées. Je m'excuse donc auprès de certains des rappels généraux que mon exposé comportera obligatoirement. Nous examinerons successivement la nature, les sources et les modalités d'irradiation des rayonnements non ionisants, leur interaction avec la matière, leurs effets biologiques et pathologiques, les mesures de protection concernant la réglementation, la prévention et la surveillance.

## Nature physique

Les rayonnements non ionisants se caractérisent par le fait que leur interaction avec la matière n'engendre pas d'ionisation.

Nous n'étudierons dans le présent rapport que les rayonnements électromagnétiques non ionisants avec parfois leur comparaison avec les rayonnements électromagnétiques ionisants.

Le spectre des ondes électromagnétiques est illustré par la figure I. Il va des rayons gamma et des rayons X ultra-durs aux ondes radio de grande longueur d'onde. Compte-tenu de l'énergie des photons et du mode d'interaction avec la matière, il est possible de distinguer plusieurs zones spectrales dont les frontières ne sont pas délimitées avec netteté.

Lasers. En outre, une autre catégorie de rayonnements électromagnétiques mérite une mention particulière : les lasers et les masers. Il s'agit de rayonnements électromagnétiques cohérents dans le domaine des longueurs d'onde de l'ultra-violet à l'infra-rouge pour les lasers, des micro-ondes pour les masers.

Les lasers sont des émetteurs de faisceaux de lumière fortement collimatés dont la densité d'énergie et la densité de puissance peuvent être considérables. Ces faisceaux lumineux sont cohérents à la fois dans l'espace et dans le temps. Ils sont monochromatiques, les gammes d'émission allant de l'ultra-violet à l'infra-rouge en passant par le domaine du visible.

### Unités

Les unités utilisées en protection contre les rayonnements non ionisants sont de nature très différente de celles utilisées pour les rayonnements ionisants. Pour ces derniers on emploie des unités d'exposition (Roentgen) et des unités de dose absorbée dans la matière (rad) ; le rad est exprimé en Joule par gramme de matière. Pour les rayonnements non ionisants on emploie des densités d'énergie en  $J/cm^2$  ou des densités de puissance en  $W/cm^2$  ; il s'agit donc d'énergie passant au travers d'une surface et non dissipée dans un volume ou une masse. Il en résulte que les relations dose-effet, d'une part, et les normes réglementaires d'autre part, sont difficilement comparables entre rayonnements non ionisants et rayonnements ionisants.

### Les sources de rayonnements non ionisants

Les sources de rayonnements non ionisants sont très diverses et doivent être étudiées séparément pour chacun d'eux.

Lumière visible - Ultra-violet - Infra-rouge. La source principale de lumière visible, d'ultra-violet et d'infra-rouge est le rayonnement solaire. Les générateurs artificiels de rayonnement ultra-violet sont essentiellement constitués par des tubes au sein desquels on produit des décharges électriques dans un gaz. Les sources artificielles de rayonnement infra-rouge sont constituées par des matériaux ayant atteint une température suffisante.

Micro-ondes. Un générateur de micro-ondes est constitué par :

- le générateur proprement dit qui crée un rayonnement électromagnétique par déplacement d'électrons dans des champs électriques et magnétiques combinés. Suivant la fréquence et la puissance désirée on utilise des tubes électroniques, des magnétrons ou des klistrons.
- un organe de transmission constitué par une ligne bifilaire, un câble coaxial ou un guide d'onde.
- une antenne d'émission (radar) ou une cavité (four).

Lasers. L'effet laser est obtenu en stimulant le retour à l'état normal d'un certain nombre d'atomes qui ont une population d'électrons préalablement inversée de leur niveau atomique. Le laser amplifie et coordonne ce retour. Pour mettre en oeuvre l'effet laser, il faut disposer :

- d'un matériel atomique dont la population électronique peut être inversée,
- d'un moyen d'inverser cette population,
- d'un moyen de stimuler le retour à l'état normal à l'aide d'un résonateur optique.

La durée de l'impulsion laser dépend essentiellement du mode de fonctionnement :

- lasers à fonctionnement continu,
- lasers à impulsion normale (durée d'impulsion d'environ 1 milliseconde),
- lasers déclenchés ou à impulsion géante (durée d'impulsion d'environ 1 nanoseconde).

Les lasers à impulsion atteignent de grandes puissances dans des temps nécessairement très courts, surtout lorsqu'ils fonctionnent " en déclenché " (exemple : le gigawatt en moins de 30 nanosecondes). Les lasers à fonctionnement continu émettent des rayonnements allant de quelques milliwatts à 500 watts.

Du point de vue technologique on distingue : les lasers à cristal, les lasers à gaz, les lasers à semi-conducteurs, les lasers à liquide.

### Modalités d'irradiation

Les modalités d'irradiation par des rayonnements non ionisants sont extrêmement diverses tant par leur nature que par leur importance relative.

Elles tiennent tout d'abord à la production de ces rayonnements soit dans le milieu naturel, soit dans les activités humaines. Les applications des rayonnements non ionisants sont en effet scientifiques, industrielles, militaires, médicales, domestiques. Les personnes exposées peuvent être classées en trois catégories principales :

- les travailleurs,
- le public,
- les patients (irradiation médicale).

Lumière visible. Les modalités d'irradiation par la lumière visible sont trop communes pour qu'il soit nécessaire de les rappeler.

Ultra-violet. En milieu professionnel, une exposition peut survenir dans les cas suivants :

1. Rayonnements ultra-violetes solaires : travaux de maçonnerie, construction des routes, travaux agricoles, travaux en mer (pêche), travaux en haute montagne.

2. Rayonnements ultra-violetes artificiels : fabrication du verre, sidérurgie et coulée, métallurgie (laminage), contrôle des moules métalliques, travaux de soudage et de découpage à l'arc, fabrication de manchons à incandescence, fabrication et contrôle de lampes à ultra-violetes et de tubes à décharges, lithographie, irradiation des denrées alimentaires, des médicaments et du tabac.

Dans le domaine public, l'irradiation solaire est essentiellement liée aux loisirs, l'irradiation artificielle au bronzage esthétique.

Dans le domaine médical, l'irradiation ultra-violette est due à un certain nombre de traitements photothérapeutiques.

Infra-rouge. La surexposition professionnelle aux rayonnements infra-rouges menace les personnes qui travaillent dans le voisinage des fours ou sur des corps ou objets portés à haute température.

L'irradiation du public est essentiellement liée à l'utilisation des appareils de chauffage, l'irradiation médicale à certaines méthodes de thermo-thérapie.

Micro-ondes. Un risque d'exposition à des doses considérées comme supérieures pour l'absorption d'une certaine dose d'énergie aux niveaux tolérables pour l'organisme humain ou partielle est à signaler dans les professions suivantes : personnel volant ou personnel au sol des aérodromes; personnel maritime; mécanicien radar et personnel d'exploitation des stations radar; personnel travaillant dans les stations aérospatiales; personnel exposé autour de fours à haute fréquence; personnel travaillant avec des masers; personnes utilisant des appareils médicaux à micro-ondes; personnes affectées au contrôle des champs de micro-ondes; personnes utilisant des fours à micro-ondes (cuisines); personnes préposées à la stérilisation des denrées alimentaires et des produits pharmaceutiques; travailleurs du bois contre-plaqué.

L'irradiation du public revêt deux modalités essentielles :  
- un certain nombre de personnes peuvent être exposées soit du fait de leur logement, soit du fait de leur passage aux rayonnements émis par des sources puissantes (radar);  
- d'autre part, un nombre de plus en plus grand de foyers domestiques sont équipés avec des fours culinaires à micro-ondes dont les risques d'irradiation en fonctionnement normal ou anormal ne sont pas négligeables.

L'irradiation médicale est essentiellement due aux traitements diathermiques.

Ondes radio. Dans le domaine professionnel, les personnes exposées appartiennent aux techniciens de la radio, de la télévision et de certaines applications industrielles.

Le public d'une façon générale est soumis au champ d'ondes radio à des niveaux faibles sauf au voisinage des émetteurs puissants.

Dans le domaine médical, l'irradiation par ondes courtes est une modalité de la diathermie.

Lasers. Le laser permet de concentrer dans un très petit volume une puissance électromagnétique considérable et d'obtenir des températures extrêmement élevées capables de faire fondre tous les matériaux même les plus réfractaires ou de détruire les tissus. C'est pourquoi différents types de rayonnement laser trouvent d'ores et déjà de nombreuses utilisations :

- industrielle et technique : micro-soudage, micro-usinage des métaux, forage et découpage, détermination de niveaux à l'aide de rayons conducteurs dans le bâtiment, photochimie, optique appliquée (holographie), métrologie (précision extraordinaire des techniques de mesure), télémétrie (lidar : appareil fonctionnant sur le même principe que le radar et qui permet des mesures extrêmement précises), télécommunications.
- recherche scientifique. Le laser, en effet, continuera de plus en plus à faire l'objet de nombreuses recherches dans les universités et les laboratoires.

Dans le domaine public on commence à trouver des gadgets utilisant des rayonnements lasers et même des jouets.

Dans le domaine médical existent des applications médico-chirurgicales : interventions sur la rétine, destruction de tumeurs, etc ... Le rayonnement laser lorsqu'il est utilisé, assure une cautérisation instantanée et réalise une excellente hémostase.

#### Interaction des rayonnements électromagnétiques avec la matière

La compréhension des mécanismes d'action biologique des rayonnements non ionisants, et d'une façon générale des rayonnements électromagnétiques, est directement liée à la pénétration de ces rayonnements dans la matière vivante et à leur interaction avec elle.

La pénétration des rayonnements électromagnétiques dans la matière vivante est fonction de leur énergie et passe par un minimum pour les rayonnements visibles et voisins ainsi que le montre la figure 2. Certes, des différences existent selon la nature des tissus et les interfaces des organes. De même, des phénomènes de diffusion ou de résonance peuvent interférer avec les simples lois d'absorption. Mais d'une façon générale des interactions auront toujours lieu dans la peau, alors que les organes profonds du corps ne seront intéressés que par les rayonnements de grande énergie (ionisants) ou de grande longueur d'onde (micro-ondes et ondes radio). L'oeil constitue un cas particulier intéressant et la figure 3 montre qu'il est entièrement traversé par les rayonnements ionisants et les micro-ondes qu'il est opaque aux ultraviolets courts et aux infrarouges longs et qu'il concentre sur la rétine la lumière visible, les UV et IR voisins de celle-ci.

Les interactions des rayonnements électromagnétiques avec la matière peuvent intervenir à différents niveaux des édifices atomiques et moléculaires. De façon schématique et caricaturale, on peut admettre en première

approximation que les rayonnements ionisants perturbent les édifices atomiques et les rayonnements non ionisants les édifices moléculaires. Les rayonnements ionisants sont seuls capables, par absorption dans la matière, de former des électrons secondaires hautement énergétiques dont l'énergie cinétique s'épuise par ionisation et excitation. Les rayonnements non ionisants peuvent agir à trois niveaux différents : au niveau des électrons orbitaux externes atomiques et moléculaires avec corrélativement des réactions photochimiques; au niveau des atomes constitutifs des molécules considérées comme un tout avec des phénomènes de rotation. Les phénomènes de vibration, de rotation et de diverses déformations aboutissent à l'agitation thermique dans la matière.

On peut ajouter aux généralités précédentes les particularités suivantes concernant les effets biologiques des rayonnements électromagnétiques.

#### Effets biologiques

1. Rayons X et rayons gamma (rayonnements ionisants). L'une des caractéristiques des rayonnements ionisants est la distribution stochastique des atomes et molécules excités et ionisés dans la matière irradiée. Les excitations électroniques ainsi que les ionisations conduisent à des réactions radiochimiques non spécifiques. Ces réactions non spécifiques sont à l'origine de réactions biologiques secondaires de caractères généralement lésionnel. La chaleur dégagée par l'absorption des rayonnements, ainsi que les épiphénomènes thermiques des réactions radiochimiques sont sans importance sur le plan biologique.

2. Rayons ultra-violets. Les effets photobiologiques des ultra-violets sont une conséquence des réactions photochimiques. La chaleur produite par l'absorption des rayons ultra-violets, ainsi que les effets thermiques des réactions photochimiques sont d'une importance très secondaire sur le plan biologique.

3. Lumière visible. Les réactions photochimiques sont de faible importance et de caractère très spécifique (vision, photosynthèse). Chez l'homme, une grande partie de l'énergie lumineuse absorbée par la peau se transforme en chaleur. C'est la raison pour laquelle, dans le spectre de la lumière visible, il faut compter aussi bien avec des effets biologiques de caractère spécifique (tels que l'éblouissement et la photosensibilisation) qu'avec des effets thermiques de type pur (brûlures de la peau ou de la rétine).

4. Rayonnement infra-rouge. L'absorption du rayonnement infra-rouge par un tissu biologique se traduit non par une excitation électronique mais simplement par une excitation des niveaux vibrationnels et rotationnels. Au cours de ce processus d'absorption, seule l'énergie thermique de la substance irradiée augmente. Il s'ensuit que la production de chaleur est le seul effet biologique qui procède de cette absorption.

5. Micro-ondes. L'absorption des micro-ondes peut se traduire par l'excitation de quelques niveaux vibrationnels, mais ce sont essentiellement les niveaux rotationnels qui se trouvent excités. De plus, ces champs de haute fréquence engendrent des courants électriques. L'un et l'autre de ces effets se traduisent par une production de chaleur. L'effet biologique essentiel de

l'absorption des micro-ondes est une production de chaleur. Les effets non thermiques ne semblent jouer qu'un rôle accessoire.

6. Ondes radio. L'absorption a pour effet de créer dans les cellules et tissus des courants électriques qui se traduisent par de la chaleur. A des fréquences inférieures à environ 300 kHz, et pour certaines puissances de champ électrique, les cellules et les tissus peuvent subir une certaine excitation électrique, sans que cette excitation s'accompagne d'autres effets.

7. Lasers. Le rayonnement laser a une triple action biologique :

- effet thermique. L'effet thermique dépend du type et de la puissance du laser et de la structure de l'échantillon biologique exposé. En particulier, l'hétérogénéité des milieux biologiques peut entraîner une distribution thermique irrégulière et certaines différences d'action selon la nature du tissu.
- effet électrique. Le faisceau laser perturbe le cortège électronique des atomes de la matière irradiée. Il s'ensuit des perturbations qui peuvent être à l'origine de divers effets chimiques qui, à leur tour, pourront avoir des conséquences biologiques.
- effet mécanique. Le faisceau laser induit dans le milieu qu'il traverse des phonons (le phonon est la quantité élémentaire d'énergie élastique) stimulés transportant une énergie très grande qu'on appelle hypersons. Ces hypersons seraient à l'origine des effets lésionnels observés non pas au point d'impact du rayonnement laser sur le tissu mais à son point d'émergence.

#### Les dommages radiopathologiques

Après avoir étudié les mécanismes d'action biologique des rayonnements non ionisants, il convient d'envisager les dommages qu'ils peuvent engendrer.

1. Ultra-violet. Une surexposition aux rayons ultra-violet se traduit par des lésions à caractère aigu ou chronique. Parmi les lésions aiguës, citons les coups de soleil ainsi que l'érythème actinique. La surexposition aux rayons ultra-violet est susceptible de provoquer une kérato-conjonctivite douloureuse dont la guérison est cependant rapide et sans séquelles. Après une exposition prolongée ou répétée, des modifications cutanées irréversibles sont susceptibles de se produire. La peau devient brune et sèche, elle se ride et perd de son élasticité. En soi, il ne s'agit que d'affection bénigne. Cependant, sur cette dermatose peuvent se greffer des épithéliomas et des kératoses séniles. Il est connu que les rayonnements ultra-violet peuvent engendrer, à long terme, des cancers cutanés. Mais les relations "dose-effet" sont mal établies en cette matière.

2. Lumière visible. Les effets de la lumière visible sur l'organisme sont analogues à ceux des rayonnements ultra-violet. En outre, une exposition prolongée équivaut à une exposition à la chaleur. Enfin, les effets cataractogènes d'une telle exposition sont loin d'être négligeables.

3. Infra-rouge. Etant donné la faible pénétration, les seuls organes à pouvoir être atteints chez l'homme sont la peau et les yeux. Selon la densité du flux énergétique du rayonnement infra-rouge absorbé par la peau, on constate soit une légère hyperthermie locale, soit une hyperthermie généralisée plus ou moins grave, soit des brûlures. La lésion classique de l'oeil, en dehors des

brûlures de caractère aigu, est la cataracte.

4. Micro-ondes. Les micro-ondes sont absorbées par l'organisme. L'énergie absorbée est entièrement convertie en chaleur. L'augmentation de la température en un point donné de l'organisme humain dépend :

- de la quantité d'énergie des micro-ondes absorbée,
- de la conductibilité thermique du tissu ainsi que du transport thermique par l'intermédiaire du courant sanguin.

Les effets physiologiques et pathologiques sont une conséquence de l'augmentation locale de la température et de la charge thermique de l'ensemble de l'organisme (effet thermique). C'est la raison pour laquelle, pour de fortes densités de flux, il peut se produire localement des échauffements ou des brûlures, par exemple au niveau de la peau ou des tissus sous-jacents. Etant donné l'absence de vascularisation du cristallin, l'augmentation de température dans le cristallin peut devenir suffisamment grande pour provoquer le plus souvent et après un certain délai, l'opacification du cristallin. Une augmentation de la température dans de grandes régions de l'organisme humain conduit à une hyperthermie qui peut être létale.

En dehors des effets thermiques qui entraînent des lésions anatomiques, certains (école russe en particulier) envisagent des effets non thermiques. L'interprétation pathogénique est délicate et complexe; elle met en cause l'action directe des champs électromagnétiques et en particulier, des champs magnétiques variables sur la matière vivante. Des sujets exposés à de faibles intensités, ont présenté des troubles de type fonctionnel :

- troubles neuro-végétatifs : fatigabilité; hypersudation; somnolence (ou insomnie); céphalée; troubles sensoriels (visuels ou auditifs); instabilité émotionnelle; hyperexcitabilité neuromusculaire; perte d'appétit, nausées; troubles du rythme cardiaque, crises vagotoniques; état lipothymique, instabilité tensionnelle.
- troubles endocriniens : déficit surrénalien; hyperfonctionnement thyroïdien.
- modifications sanguines : ioniques; électrophorétiques; cytologiques - anémie, lymphopénie, polynucléose - coagulabilité sanguine et résistance globulaire, rarement touchées.

5. Ondes radio. En général, dans les utilisations professionnelles, les ondes radio induisent dans l'organisme des intensités tellement faibles qu'il n'en résulte pas d'effets biologiques. Cependant on aurait observé, pour certaines utilisations professionnelles, des manifestations neuro-végétatives, de la fatigue, de l'asthénie. Ces symptômes disparaissent en même temps que l'exposition. On n'a pas observé de séquelles.

6. Lasers. Les dommages corporels susceptibles d'être provoqués par le rayonnement laser peuvent résulter du rayonnement direct aussi bien que des rayonnements réfléchis diffusés. Les organes critiques sont l'oeil et la peau.

- Effets oculaires. La totalité du globe oculaire peut être atteinte, les dommages pouvant aller de la lésion minimale jusqu'à la déchirure de type explosif. Alors que les petites lésions de la cornée évoluent en général vers la guérison, les lésions de la rétine sont irréversibles. Les atteintes rétiniennes varient dans leur gravité en fonction de la topographie des lésions. Quelle que soit la topographie des lésions rétiniennes, il y a lieu de souligner le



caractère fréquemment indolore de celles-ci. Les facteurs aggravants des lésions sont les suivants :

Focalisation sur la rétine. Les rayons lasers qui traversent l'oeil se trouvent focalisés sur la rétine par le cristallin, ce qui se traduit par une forte augmentation de la densité énergétique. Pour l'établissement des consignes de sécurité, il faut faire entrer en ligne de compte le cas le plus défavorable où un faisceau de rayons parallèles est focalisé au niveau de la rétine sur une surface ayant le diamètre d'un disque de diffraction.

Transmission à travers l'oeil. La région de l'oeil située en avant de la rétine est transparente à la gamme des longueurs d'ondes, allant de 0,4 à 1,4  $\mu\text{m}$  à quoi viennent s'ajouter deux autres gammes dans l'infrarouge. Si le rayonnement est absorbé dans la partie de l'oeil située en avant de la rétine, la densité énergétique sur la rétine peut se trouver diminuée. Mais alors, les tissus antérieurs sont exposés à un danger plus grand. Dans ce cas cependant vu l'absence de focalisation, le seuil critique des lésions est nettement plus élevé.

Durée d'irradiation. Dans le cas des lasers pulsés ou de lasers à impulsions géantes, on a constaté sur le plan expérimental que la densité énergétique admissible diminuait en même temps que la durée des impulsions. Cela se comprend aisément si l'on considère qu'avec un apport énergétique lent, l'énergie calorifique produite peut partiellement se dissiper, relevant d'autant le seuil lésionnel critique. Par ailleurs, un certain rôle est joué à cet égard par la fréquence de la répétition des impulsions.

Grosseur de l'image. Les travaux expérimentaux montrent que la densité d'énergie nécessaire à l'apparition d'une lésion augmente lorsque la surface de la région irradiée devient plus petite.

Il demeure, en matière d'effets oculaires, un certain nombre d'inconnues relatives :

- à l'effet de sommation des faisceaux de basse énergie;
- à l'effet de sommation des effets punctiformes périphériques;
- aux effets à long terme du rayonnement laser après cessation de l'exposition au risque;
- à l'effet en profondeur (effet Brillouin).

- Effets cutanés. L'incidence sur la peau d'un rayonnement laser peut provoquer immédiatement une brûlure avec coagulation locale, ce qui permet une cicatrisation rapide. Néanmoins la réaction cutanée dépend de plusieurs facteurs qui sont :

- la longueur d'onde du faisceau,
- la durée d'exposition,
- les "qualités optiques" de la peau en matière d'absorption, de transmission et de propriétés réfléchissantes. C'est ainsi que l'absorption de l'énergie laser se trouve facilitée par une densité locale plus importante de la pigmentation.

On ne sait encore que peu de choses en ce qui concerne l'exposition cutanée chronique au rayonnement laser.

## Réglementation

En matière de protection, la réglementation constitue l'une des pièces maîtresses. En ce qui concerne les rayonnements électromagnétiques, un contraste saisissant existe entre l'état de la réglementation pour les rayonnements ionisants et pour les rayonnements non ionisants. Pour les rayonnements ionisants, un système cohérent et universellement adopté a été mis au point par une Commission Internationale compétente. Pour les rayonnements non ionisants, l'inventaire des réglementations fait apparaître une hétérogénéité, des lacunes, des contradictions.

### Rayonnements ionisants

La protection contre les rayonnements ionisants a été codifiée par la Commission Internationale de Protection Radiologique. L'I.C.R.P. a établi un corps de doctrine qui est représenté par des recommandations générales et des recommandations particulières aux différentes modalités d'irradiation.

De fait, les recommandations ont été transcrites de façon appropriée dans les réglementations nationales de la quasi totalité des pays. Elles sont par ailleurs reconnues de façon officielle par les grandes organisations internationales (O.M.S., B.I.T., F.A.O., A.I.E.A., Comité Scientifique des Radiations des Nations Unies) ou régionales (Euratom, O.C.D.E.). Il résulte de cet état de fait une homogénéité et une cohérence exemplaires en matière de protection contre les rayonnements ionisants.

### Rayonnements non ionisants

Dans le domaine des rayonnements non ionisants, il n'existe malheureusement pas d'organisme international comparable à la Commission Internationale de Protection Radiologique.

Il en résulte qu'aucune réglementation universellement acceptée n'existe, ni pour l'ensemble des rayonnements non ionisants, ni même pour certains d'entre eux. En outre, les textes réglementaires nationaux ou particuliers sont disparates avec de nombreuses lacunes. Enfin, il existe pour certains rayonnements tels que les micro-ondes des différences très sensibles entre les limites d'irradiation pouvant atteindre un facteur 1000.

## Généralités

Normes fondamentales. Quelques recherches ont été faites pour tenter de déterminer une limite fondée sur un seuil de lésion pour quelques types de rayonnements avec des longueurs d'ondes déterminées et un effet biologique donné, par exemple : érythème et kérate-conjonctivite pour UV de différentes longueurs d'onde ; lésion de la rétine et de la peau pour les lasers. Mais les conditions expérimentales et les résultats disparates rendent la comparaison difficile.

L'exposition limite de l'organe est généralement expérimentée en  $J \cdot cm^{-2}$ . Lorsqu'il s'agit de tissus internes cette norme est quelquefois déduite de

l'exposition des tissus externes par application d'un facteur de transmission ou de concentration.

Limites dérivées. Les recommandations, réglementations ou codes de pratique que l'on trouve pour certains types de rayonnements (UV, micro-ondes, lasers) concernent des limites dérivées. Ces dernières sont exprimées soit en densité d'énergie pour une impulsion de durée déterminée (surtout lorsqu'il s'agit d'impulsions relativement courtes) soit en densité de puissance  $W \cdot cm^{-2}$ .

Les valeurs recommandées sont très variables non seulement selon les pays, mais souvent dans un même pays selon l'organisme qui les propose. L'absence de méthodes de détermination, de conditions expérimentales et de moyens de mesure uniformes rend souvent leur comparaison difficile.

#### Etat actuel de la réglementation

UV. Des recherches existent pour déterminer un seuil de lésion au niveau de la peau ou de l'oeil en fonction de la longueur d'onde pour servir de base à une norme fondamentale. Mais, selon l'O.M.S., il est urgent de déterminer la relation dose-effet pour cancer de la peau. La seule recommandation officielle que l'on trouve est celle mentionnée par l'O.M.S., qui semble provenir d'une recommandation déjà assez ancienne de l'American Medical Association (1948): l'exposition ne doit pas dépasser :

0,5  $\mu W$  par  $cm^2$  pour une exposition  $\leq 7$  heures  
et 0,1  $\mu W$  par  $cm^2$  pour une exposition continue de 24 h par jour.

Infra-rouges. Les connaissances sur les effets possibles à long terme sont insuffisantes, et en particulier celles sur la relation dose-effet. Les seules données sur lesquelles on pourrait actuellement fonder des normes sont les seuils de lésion connus pour la cornée (O.M.S.) :

7,6  $J/cm^2$  aux longueurs d'onde 0,88 - 1,1  $\mu m$   
2,8  $J/cm^2$  aux longueurs d'onde 1,2 - 1,7  $\mu m$

#### Microondes

Normes fondamentales. Les bases sur lesquelles elles devraient être fondées, c'est-à-dire les effets biologiques, sont encore mal connues. Les données quantitative, lorsqu'il y en a, sont difficiles à interpréter. La question de l'existence d'un seuil n'est pas résolue et on ne peut exclure complètement la possibilité d'effets cumulatifs. D'autre part, une telle norme devrait être fondée sur l'énergie absorbée dans les tissus. Or, celle-ci ne peut être déterminée à l'heure actuelle avec le minimum de précision qui serait nécessaire pour en évaluer les risques. C'est pourquoi, il paraît difficile à l'heure actuelle de déterminer des normes fondamentales. Dunster cependant, propose comme norme fondamentale, une densité de puissance de  $10 mW/cm^2$ .

Limites dérivées. Les normes dérivées pour le domaine des microondes sont généralement exprimées en densité de puissance ( $mW/cm^2$ ) pour une durée d'exposition déterminée. Un grand nombre de pays et d'organismes ont promulgué ou recommandé des limites d'exposition (tableau 1). On constate des variations d'un organisme à l'autre, mais il se dégage essentiellement deux grands courants :

- Aux Etats-Unis tout d'abord, dans certains pays européens ensuite, ces limites sont fondées sur l'aptitude de l'organisme à supporter une charge thermique :

- 10 mW/cm<sup>2</sup> supportable pendant un temps relativement long (ex. journée de travail),
- 1 mW/cm<sup>2</sup> recommandé par certains organismes lorsqu'il s'agit d'une exposition permanente (ex. marins dormant à proximité d'une antenne sur un bateau) et pour la population (limite de fuite à 5 cm proposée pour les nouveaux fours à microondes).

Pour des expositions intermittentes très courtes inférieures à l'heure, deux formules :

1 - Committee of the American Standards Association : 1 mW.h/cm<sup>2</sup> par fraction de 0,1 heure, ce qui donne 10 mW/cm<sup>2</sup> pour 0,1 heure.

2 - La plupart des organismes militaires aux Etats-Unis comme en Europe : la durée d'exposition en minute :

$$t = 60 \times \left(\frac{10}{W}\right)^2$$

W devant rester inférieur à 100 ou 55 mW/cm<sup>2</sup> selon des pays.  
W étant la densité de puissance moyenne sur le temps t.

- La C.E.E. a créé un groupe de travail dont le rapporteur J. Dunster propose les limites simplifiées suivantes :

- exposition continue, densité de puissance dans le champ : 10 mW/cm<sup>2</sup>

- exposition intermittente, densité de puissance :  $D \leq 100$  mW/cm<sup>2</sup>

durée d'exposition :  $t = 60 \times \left(\frac{D}{100}\right)^2$

- En U.R.S.S. et certains pays d'Europe Centrale, les limites sont fondées sur des troubles fonctionnels liés à un déséquilibre neuro-végétatif plus ou moins accentué. Aussi, sont-elles beaucoup plus sévères :

- 1 mW/cm<sup>2</sup> pour moins de 20 minutes de séjour et port de lunettes de protection obligatoire,
- 0,1 mW/cm<sup>2</sup> pour une exposition de deux heures par jour
- 0,01 mW/cm<sup>2</sup> pour plus de deux heures par jour.

### Lasers

Normes fondamentales. Organes sensibles : oeil et peau. L'effet sur l'oeil dépend de la longueur d'onde, ainsi que des conditions d'éclairage extérieur qui déterminent le diamètre de la pupille. On a cherché à établir des seuils de lésion pour la rétine.

Ici encore pas de moyens de mesure de l'énergie absorbée. Aussi toutes les réglementations portent-elles sur des limites dérivées.

Limites dérivées. Elles s'expriment différemment selon le mode de fonctionnement du laser :

- en densité d'énergie par unité de surface ( $J/cm^2$ ) par impulsion pour les impulsions très courtes et les trains d'impulsions très courtes;
- en densité de puissance par unité de surface pour les rayonnements continus ( $W/cm^2$ ).

Le code de pratique Britanique indique les limites suivantes pour l'exposition de la cornée (tableau II). J. Dunster propose pour la C. E. E. les limites suivantes pour l'oeil et la peau pour les lasers émettant dans l'ultra-violet, le visible et l'infra-rouge (tableau III, IV, V).

### Prévention

La protection contre les rayonnements non ionisants doit comprendre la réglementation que nous venons de voir, la prévention et la surveillance. La prévention constitue le 2ème volet du triptyque de la protection radiologique. Dans le domaine des rayonnements ionisants, cette prévention a fait l'objet d'études très poussées qui ont contribué à faire de l'énergie atomique l'une des activités humaines où les risques sont les plus faibles. Elle est essentiellement basée sur la sécurité des installations, les protections liées aux appareils, les protections liées aux travailleurs, les consignes d'exploitation et les habitudes de travail.

Pour les rayonnements non ionisants de nombreuses méthodes de prévention ont été mises en oeuvre, essentiellement par les équipes scientifiques, militaires ou industrielles utilisant des sources relativement importantes. Les constructeurs également ont prévu des dispositifs de protection qu'il conviendrait cependant, dans un certain nombre de cas, d'améliorer de façon sensible.

D'une façon générale, on peut envisager des mesures de prévention au stade de la construction, au stade de l'utilisation sur un plan collectif ou sur un plan individuel.

Au stade de la construction, dans toute la mesure du possible, les sources de rayonnements non ionisants devraient posséder des écrans pour éviter tous les rayonnements parasites autres que le faisceau utile. Les appareils générateurs devraient faire l'objet d'une homologation officielle, en particulier pour ceux qui sont utilisés par le public. Il devrait être interdit d'apporter des modifications quelconques au système de protection. Peut-être conviendrait-il d'envisager un contrôle périodique des appareillages susceptibles de montrer des défauts au cours du temps.

Dans l'utilisation des sources de rayonnements non ionisants les mesures collectives devraient comprendre notamment les suivantes :

- les installations devraient être conçues de façon telle que dans tous les cas où cela est possible, elles constituent un système fermé sans rayonnement de fuite;
- pour le cas où les installations seraient en système ouvert, des zones interdites à séjour réglementé ou contrôlé devraient être établies;
- le nombre de personnes utilisant les installations devrait être réduit au minimum;

- des mesures du rayonnement ambiant devraient être effectuées afin de s'assurer que les limites d'irradiation sont bien respectées;
- des consignes strictes devraient être édictées pour le personnel.

A ces mesures collectives, des mesures individuelles peuvent s'ajouter lorsqu'il est impossible d'abaisser les niveaux d'irradiation au-dessous des limites réglementaires. Ce n'est que dans ce cas, qui devrait rester exceptionnel, que l'on peut envisager des moyens de protection individuelle portés par les travailleurs. Les organes les plus sensibles étant d'une façon générale les yeux, la peau et le corps entier selon le rayonnement, les moyens de protection consisteront soit en lunette, soit en vêtement protecteur.

### Surveillance

La surveillance constitue le 3ème volet de la protection radiologique. On distingue une surveillance physique et une surveillance médicale.

#### Surveillance physique.

1. La surveillance physique des rayonnements ionisants a fait l'objet au cours des dernières décades d'un nombre considérable de travaux et de réalisations. On est actuellement capable de déceler nettement au-dessous des limites d'irradiation les doses reçues par les différentes catégories de personnes exposées. Les méthodes utilisées peuvent être directes ou indirectes :

- Les méthodes directes portent sur la mesure de l'irradiation au niveau des personnes elles-mêmes au moyen de détecteurs individuels (films dosimètres, stylos électromètres etc...).
- Les méthodes indirectes consistent à évaluer l'irradiation à partir de mesures portant sur l'environnement professionnel ou publique (champ d'irradiation ou de contamination).

La dosimétrie des rayonnements ionisants est devenue une discipline en soi. Elle fait appel à des méthodes de mesures extrêmement diverses. Elle a à sa disposition une grande variété de dosimètres permettant des mesures instantanées ou cumulées avec des gammes d'utilisation extrêmement larges, des sensibilités très poussées et des précisions largement suffisantes.

2. En ce qui concerne les rayonnements non ionisants, la situation est toute différente. La dosimétrie de ces rayonnements est difficile et elle n'a pas fait l'objet d'études aussi étendues et poussées qu'en ce qui concerne les rayonnements ionisants. Les méthodes utilisées ont un caractère spécifique qui limite leurs possibilités pratiques. Les difficultés sont déjà grandes pour obtenir des informations précises sur les champs de rayonnements non ionisants. Il n'existe pratiquement pas de détecteurs individuels. Il en résulte que la surveillance pour les rayonnements non ionisants ne peut être actuellement que collective.

Certaines estimations sont faites à partir de mesures portant sur les faisceaux au voisinage immédiat des sources, d'autres estimations sont faites à distance, c'est à dire au voisinage des personnes exposées dans les champs de rayonnements non ionisants.

A titre d'exemple, nous allons passer en revue quelques méthodes dosimétriques relatives aux deux catégories importantes de rayonnements non ionisants : les micro-ondes et les lasers.

### Micro-ondes

Principe de mesure. Les micro-ondes sont constituées par un champ électrique et un champ magnétique ayant des composantes perpendiculaires et égales. Il est possible de mesurer l'une ou l'autre de ces composantes (le champ électrique est plus facile à mesurer) ou bien la densité totale de puissance.

Loin de l'émetteur, le faisceau est bien constitué. Dans la zone proche les deux composantes ne sont ni égales ni perpendiculaires et peuvent varier très rapidement d'un point à un autre. Il faudrait alors mesurer les deux composantes simultanément. Ces mesures sont particulièrement difficiles lorsqu'il y a plusieurs rayonnements de fréquence et de polarisation différentes et que viennent s'ajouter des phénomènes de réflexion (cas des navires de guerre et des bases aériennes).

En réalité, pratiquement tous les systèmes mesurent la densité de puissance par l'échauffement d'un capteur sensible.

#### Appareils de mesure :

- Pour les densités de puissance supérieures à  $1 \text{ watt/cm}^2$  : calorimètre.
- Pour les valeurs inférieures à  $1 \text{ watt/cm}^2$  : bolomètre.

### Lasers

Domaine de mesure. On peut distinguer 3 types de lasers :

- émission continue,
- laser relaxé,
- laser déclenché.

La difficulté des mesures dépend des facteurs suivants :

- diversité des longueurs d'onde
- diversité des durées d'impulsions
- origine de l'irradiation qui peut être due au rayonnement direct, réfléchi ou diffusé (importance de la diffusion atmosphérique)
- dimension des faisceaux - les faisceaux focalisés ont des diamètres qui peuvent être inférieurs à  $50 \mu\text{m}$ .
- densité d'énergie ou de puissance très élevées dans les faisceaux focalisés (pouvant atteindre  $100 \text{ MW/cm}^2$ ).

Unités de mesure. C'est le joule ou le joule/ $\text{cm}^2$  pour les lasers pulsés, le watt ou le watt/ $\text{cm}^2$  pour les lasers continus.

Principe des mesures. On peut distinguer trois types de dosimétrie :

- la mesure de la puissance du faisceau ou de l'énergie émise par pulse, dont la valeur est utile en cas d'irradiation par le faisceau direct,

- la mesure de l'intensité instantanée en un point, utile pour les cas d'irradiation par des faisceaux réfléchis,
- la mesure de l'exposition cumulée en divers points des laboratoires pour connaître les effets à long terme des rayonnements diffusés.

A ces mesures doit s'ajouter dans certains cas la détermination de la dimension du faisceau.

#### Appareils de mesure de la densité d'énergie ou de puissance :

- détecteurs photoémisifs
- détecteurs photoconducteurs
- détecteurs thermiques

#### Surveillance médicale.

Les pratiques de surveillance médicale sont fort différentes d'un pays à l'autre et ceci est vrai non seulement pour les rayonnements non ionisants mais également pour les rayonnements ionisants.

En fait, il est bon que les travailleurs exposés aux rayonnements soient soumis à une surveillance médicale analogue à celle des autres travailleurs lorsque les risques d'irradiation sont suffisamment faibles. Par contre, lorsque les risques d'irradiation sont élevés, il convient de mettre en oeuvre une surveillance médicale spécifique portant sur les tissus et organes critiques pour le rayonnement considéré.

C'est ainsi que pour les rayonnements ionisants la surveillance doit porter sur la peau, l'oeil, les tissus sanguiformateurs, les gonades etc...

En ce qui concerne les rayonnements non ionisants, et compte tenu des dommages radiopathologiques éventuels, la surveillance médicale doit porter essentiellement sur l'oeil et la peau.

Cette surveillance médicale devrait comprendre des examens à l'embauche pour juger de l'aptitude du travailleur, des examens périodiques en cas de risques suffisants, des examens après emploi quand des séquelles à long terme sont possibles.

A titre d'exemple, nous présentons la surveillance médicale envisagée pour le personnel exposé aux rayonnements laser :

1. Un examen médical d'embauche et des examens médicaux périodiques au cours desquels un ophtalmologiste compétent et entraîné fera un examen oculaire complet comportant :
  - un examen de l'acuité visuelle
  - un examen des annexes extérieures de l'oeil (paupières, conjonctives, etc..)
  - un examen des différents milieux réfringents
  - un examen du fond d'oeil après dilatation de la pupille
  - un examen de la vision binoculaire
  - un examen de la vision des couleurs
  - un examen du champ visuel.



Un examen identique devra être effectué lorsque le travailleur cessera d'être exposé au rayonnement laser.

Il est souhaitable, en raison des manipulations dangereuses que doit effectuer le personnel exposé, que celui-ci ait un équilibre psychocarcactériel satisfaisant et qu'il soit informé de l'existence et de la nature des risques. De même, il y aura lieu de procéder à un examen des téguments.

## 2. L'établissement d'une fiche de dommages.

Le rythme et la périodicité des examens systématiques doit être fonction de l'importance du risque et de la nature du travail effectué.

Les accidents cutanés se manifestent rapidement et peuvent aisément être rapportés à leur cause.

Les accidents oculaires ne sont quelquefois rapportés à leur cause que tardivement. Les lésions périphériques de la rétine peuvent n'être remarquées qu'à l'occasion d'un examen systématique ou par la victime après un laps de temps plus ou moins long, l'accident causal étant passé inaperçu et la lésion initiale étant généralement indolore.

### Conclusion

Au terme de cet exposé nous sommes arrivés à conclure que les problèmes posés par la protection contre les rayonnements non ionisants sont nombreux, importants et difficiles. La situation actuelle apparaît entièrement différente de celle existante pour les rayonnements ionisants. En matière de rayonnements non ionisants des recherches importantes doivent être poursuivies dans le domaine de leurs interactions avec la matière, de leurs effets biologiques, des dommages radiopathologiques. Un effort international d'harmonisation doit être entrepris pour les systèmes d'unités, les normes réglementaires, l'homologation des appareils. Des études technologiques doivent être développées sur la sûreté des installations, les équipements de protection et les méthodes de dosimétrie pratique. Ceci rejoint les vœux émis par le groupe de travail de l'O.M.S. en 1971 que j'ai résumés dans le tableau VI. La Commission International de Protection Radiologique en 1971 a reconnu que des contrôles adéquats devraient être établis sur les sources de rayonnements non ionisants et qu'il était à présent nécessaire d'avoir des discussions internationales sur les critères biologiques servant de base aux normes. Cependant, l'I.C.R.P. a considéré que ce sujet était en dehors de ses préoccupations courantes. L'I.C.R.P. espérait que sa déclaration faciliterait une action internationale en ce domaine. Aussi je pense que le moment est peut être venu d'organiser cette action internationale. L'I.R.P.A. peut sans doute jouer un rôle important en cette matière.

En terminant, je remercie le 3ème Congrès de l'I.R.P.A. d'avoir donné une place de choix à ces problèmes et je souhaite que les participants se fassent, dans leurs pays respectifs, les propagandistes des actions à mener pour parfaire la protection contre les rayonnements non ionisants.

Tableau I  
LIMITES D'EXPOSITION AUX MICROONDES

PAYS	EXPOSITION PERMANENTE	EXPOSITION PENDANT LES HEURES DE TRAVAIL	
		Durée d'exposition $t > 1h$	Durée d'exposition $t < 1h$
	Densité de puissance $W$ en $mW/cm^2$	$W$ en $mW/cm^2$	$W$ en $mW/cm^2$ $t$ en minutes
Etats-Unis	militaires	10	$t = 60 \times \left(\frac{10}{W}\right)^2$ avec $W \leq 100$
	civils	1	si $t > 6$ min.   si $t \leq 6$ min. $W = 10$   $1mW.h/cm^2$ par 6 min.
U.R.S.S.		si $t > 2h/jour$ 0,01 si $t \leq 2h/jour$ 0,1	$W = 1$ avec $t \leq 20$ min. /jour
FRANCE (militaires)	1	10	$t = 60 \times \left(\frac{10}{W}\right)^2$ avec $W \leq 55$
BRITISH MEDICAL COUNCIL		10	

Tableau II

CORNEAL MAXIMUM PERMISSIBLE EXPOSURE LEVELS FOR LASER RADIATION  
 DIRECT ILLUMINATION OR SPECULAR REFLECTION (7 mm pupil )

Laser Type	Q-Switched 1 ns - 1 $\mu$ s Pulsed PRF < 10 per second Energy per pulse $Jcm^{-2}$	Long Pulsed 1 $\mu$ s - 0.1s PRF < 10 per second Energy per pulse $Jcm^{-2}$	Continuous Wave Long-Term Exposure $Wcm^{-2}$
Ruby (0.69 $\mu$ m)	$3 \times 10^{-8}$	$1 \times 10^{-6}$	$4 \times 10^{-7}$
Neodymium (1.06 $\mu$ m)	$2 \times 10^{-7}$	$3 \times 10^{-6}$	$2 \times 10^{-6}$
Helium-Neon (0.63 $\mu$ m)	-	-	$3 \times 10^{-7}$
Argon (0.51 $\mu$ m) (0.48 $\mu$ m)	-	-	$3 \times 10^{-7}$

Laser systems - Code of practice, UK, 1969

Tableau III

Derived Working Limits for Energy Surface Density  
Ultra-violet Radiation (Single pulse)

Wavelength (nm)	Pulse length, t (s)	DWL (skin or eye) (J/cm <sup>2</sup> )
200 - 315	$10^{-2} - 3 \times 10^4$	$3 \times 10^{-3}$
315 - 400	$10^{-2} - 10^3$ $10^{-3} - 3 \times 10^4$	1 $10^{-3} \times t$ ( $10^{-3}$ W/cm <sup>2</sup> )
The total power surface density over both wave-length regions must not exceed 1 W/cm <sup>2</sup> .		

Tableau IV

Derived Working Limits for Energy Surface Density  
 Visible and near infra-red radiation (400 - 1400 nm) (Single pulse)

Eye (7 mm pupil)		Skin (average area 1 mm diameter)	
Pulse length, t (s)	DWL (J/cm <sup>2</sup> )	Pulse length, t (s)	DWL (J/cm <sup>2</sup> )
10 <sup>-9</sup> - 2 x 10 <sup>-5</sup>	5 x 10 <sup>-7</sup>	10 <sup>-9</sup> - 10 <sup>-7</sup>	2 x 10 <sup>-2</sup>
2 x 10 <sup>-5</sup> - 10	1.8 x 10 <sup>-3</sup> x t <sup><math>\frac{3}{4}</math></sup>	10 <sup>-7</sup> - 10	1.1 x t <sup><math>\frac{1}{4}</math></sup>
Examples		Examples	
10 <sup>-4</sup>	1.8 x 10 <sup>-6</sup>	10 <sup>-6</sup>	3.6 x 10 <sup>-2</sup>
10 <sup>-3</sup>	1 x 10 <sup>-5</sup>	10 <sup>-5</sup>	6.4 x 10 <sup>-2</sup>
10 <sup>-2</sup>	5.7 x 10 <sup>-5</sup>	10 <sup>-4</sup>	1.1 x 10 <sup>-1</sup>
10 <sup>-1</sup>	3.2 x 10 <sup>-4</sup>	10 <sup>-3</sup>	2 x 10 <sup>-1</sup>
1	1.8 x 10 <sup>-3</sup>	10 <sup>-2</sup>	3.6 x 10 <sup>-1</sup>
10	1 x 10 <sup>-2</sup>	10 <sup>-1</sup>	6.4 x 10 <sup>-1</sup>
10 - 10 <sup>4</sup>	1 x 10 <sup>-2</sup>	1	1.1
10 <sup>4</sup> - 3 x 10 <sup>4</sup>	10 <sup>-6</sup> x t (10 <sup>-6</sup> W/cm <sup>2</sup> )	10	2
		10 - 3 x 10 <sup>4</sup>	2 x 10 <sup>-1</sup> x t (2 x 10 <sup>-1</sup> W/cm <sup>2</sup> )

Tableau V

Derived Working Limits for Energy Surface Density  
 Infra-red Radiation (1400 nm to 1 mm) (Single pulse)

Pulse length, t (s)	DWL (eye and skin) (J/m <sup>2</sup> )
$10^{-9} - 10^{-7}$	$10^{-2}$
$10^{-7} - 10$	$0.56 \times t^{\frac{1}{4}}$
Examples	
$10^{-6}$	$1.8 \times 10^{-2}$
$10^{-5}$	$3.2 \times 10^{-2}$
$10^{-4}$	$5.6 \times 10^{-2}$
$10^{-3}$	$1 \times 10^{-1}$
$10^{-2}$	$1.8 \times 10^{-1}$
$10^{-1}$	$3.2 \times 10^{-1}$
1	$5.6 \times 10^{-1}$
10	1
$10 - 3 \times 10^4$	$10^{-1} \times t$ ( $10^{-1}$ W/cm <sup>2</sup> )

## Tableau VI

### Actions à mener

- Enquêtes permettant de déterminer effectifs et distribution des populations exposées.
- Etudes épidémiologiques sur groupes de travailleurs exposés pour détermination des effets éventuels à long terme.
- Réunir rapports sur cas d'exposition accidentelle pour meilleure description des troubles cliniques.
- Mise au point de dispositifs de mesure de l'exposition individuelle aux rayonnements non ionisants.
- Détermination de la relation énergie incidente-effet pour les divers organes sensibles aux différentes longueurs d'onde.
- Etude des moyens de protection.
- Harmonisation des grandeurs et unités utilisées pour la mesure de l'exposition et l'expression des limites.
- Harmonisation des normes de sécurité (normes fondamentales et limites dérivées).
- Harmonisation de la signalisation.
- Harmonisation des réglementations nationales (autorisations pour la fabrication, la vente ou l'utilisation).

D'où nécessité d'un organisme international  
qui centralise résultats et élabore recommandations

Figure 1  
SPECTRE ELECTROMAGNETIQUE

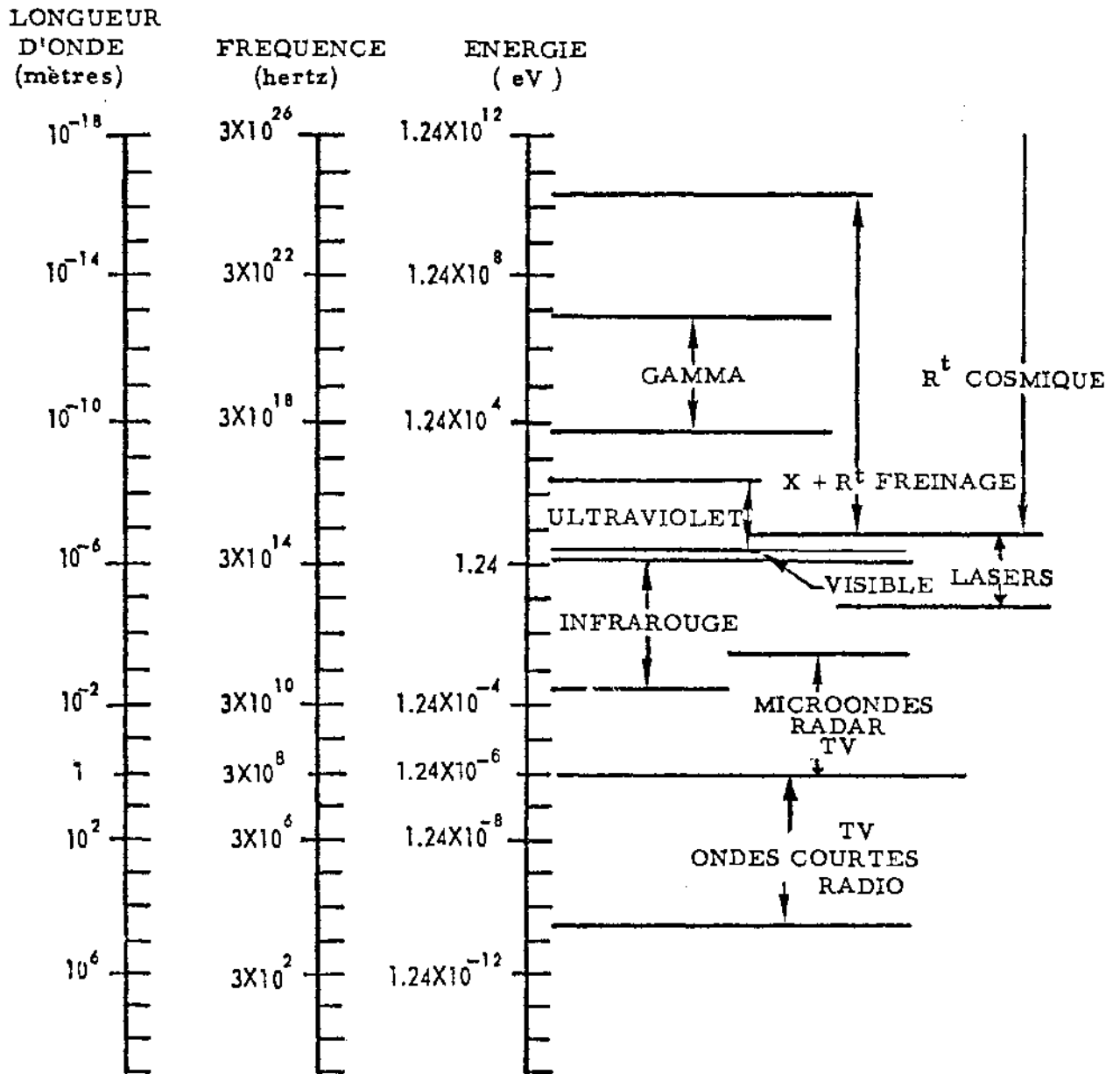




Figure 2

PENETRATION DES ONDES ELECTROMAGNETIQUES

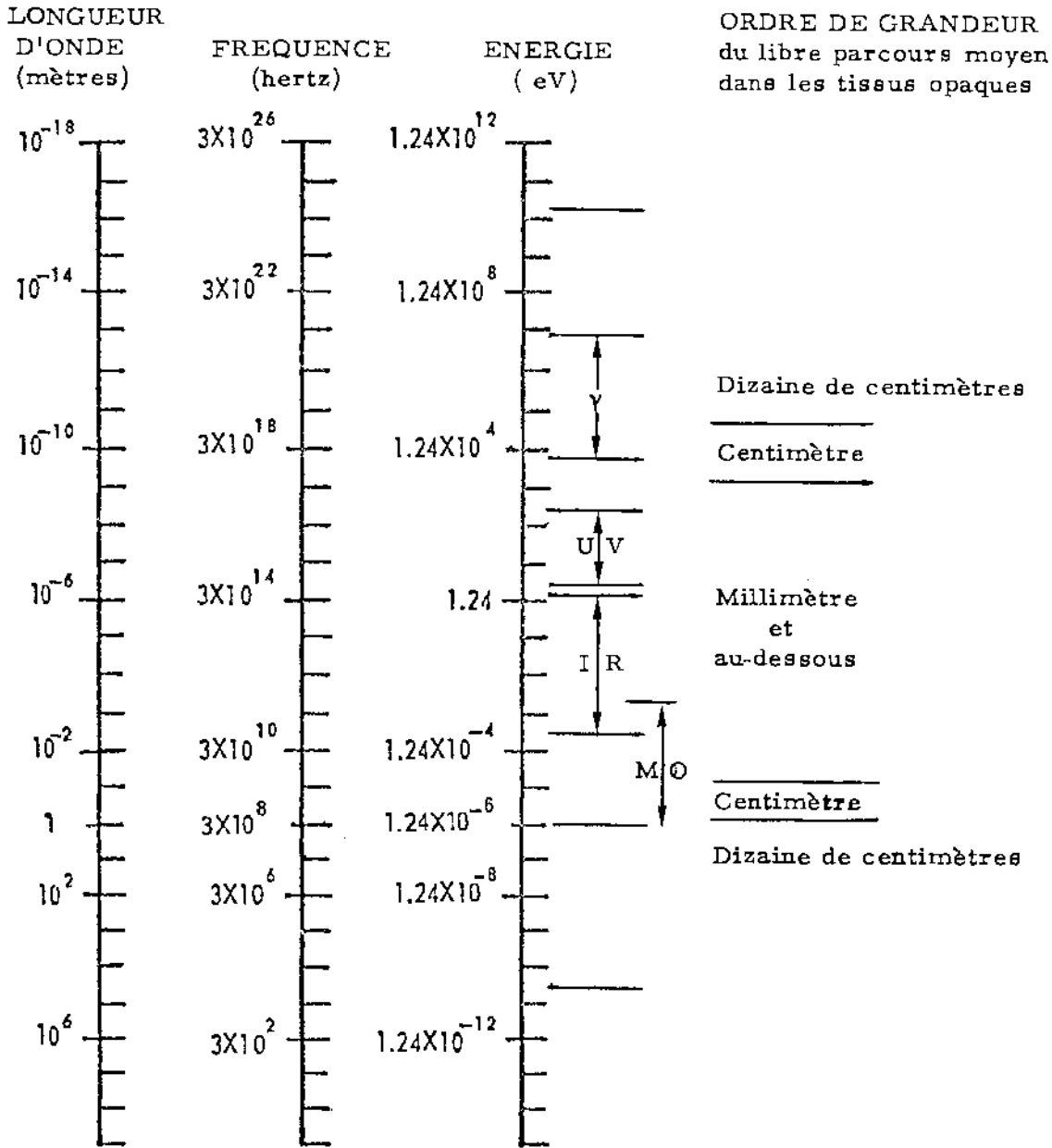
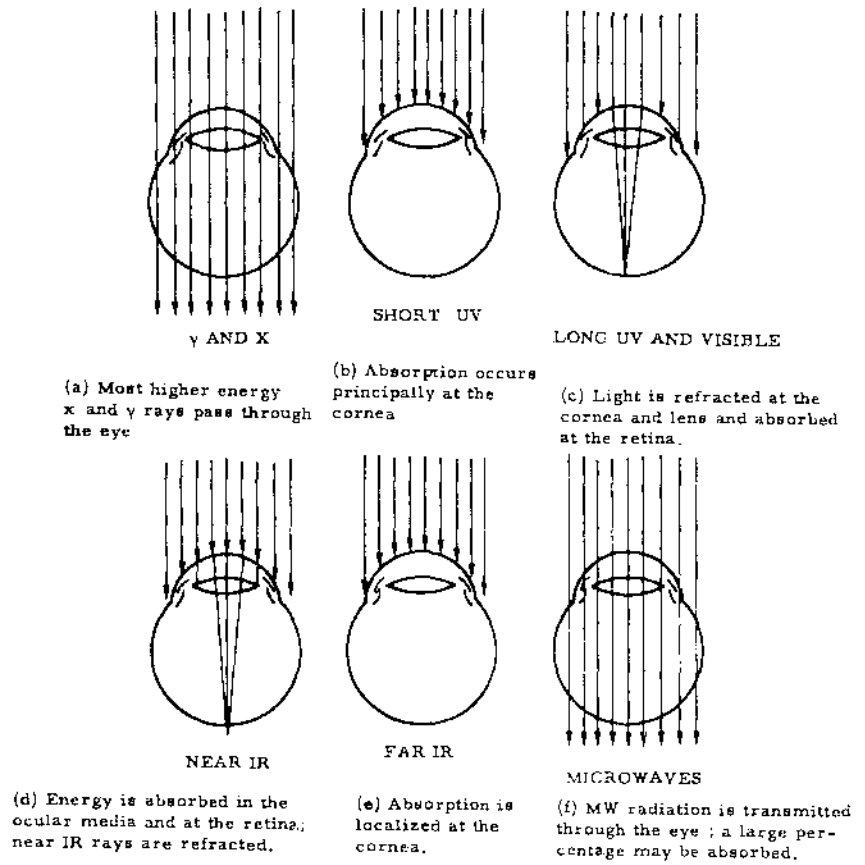


Fig.3

ABSORPTION PROPERTIES OF THE HUMAN EYE FOR ELECTROMAGNETIC RADIATION



from Laser systems - Code of practice, UK, 1969

PUBLIC INFORMATION, LEGAL ASPECTS,  
EDUCATION AND TRAINING

RADIATION PROTECTION TRAINING COURSES AT E.I.R. WÜRENLINGEN, SWITZERLAND

R. Spiess, H. Brunner

School for Radiation Protection  
Health Physics Division  
EIR, Eidg. Institut für Reaktorforschung  
CH-5303 Würenlingen, Switzerland

1. Summary

Due to the increasing applications of ionizing radiations and radioactive materials and required by radiation protection laws a growing number of professions and persons need at least a basic understanding of radiation problems and some practical instruction and training in radiation protection. The vast experience accumulated in nuclear research centers in the field of radiation protection training of employees on all levels is an excellent basis for the development of training programs for most needs. The program offered by the School for Radiation Protection of EIR (Federal Institute for Reactor Research), Würenlingen, comprises four categories :

- A) full-time health physics personnel of all levels
- B) professionally exposed personnel
- C) emergency organization personnel (fire departments, police, army etc.)
- D) students and pupils

Some typical or new courses are described in detail. The importance of such programs for improving the information and education of the public on radiation and safety matters is stressed.

2. The needs for radiation protection training

The needs for some kind of education and training in radiation protection can be grouped as follows :

- a) The persons directly involved and at risk, i.e. the professionally exposed persons in industry, medicine, research etc. are required by law to get a sufficient training. This includes not only the workers but also the responsible supervisors and managers.
- b) Persons that might be directly affected or involved in case of incidents, accidents or catastrophes (police, fire departments, transport organizations, emergency organizations, first aid and medical personnel, authorities, army, civil defense) must have sufficient knowledge and training which enable them to carry out their functions and responsibilities in case of radiation emergencies.
- c) The general public is only indirectly affected or at risk from radiation applications. But for a proper functioning as responsible citizens, opinion- or decision-makers, due to the growing concern for qualities of life and environment, and for political and economical reasons, a wide-spread and sufficient information and education of the public is highly desirable.
- d) As the reservoir for all the categories mentioned above, pupils and students on all levels and in all fields of education, training and instruction should get adequate information and basic education about radiation and safety matters during the regular curricula.

Although radiation protection originated in medicine, in most countries the nuclear research and industrial applications of radiation sources were the first areas where these training necessities were recognized and incorporated from the very beginning in the practical work and in the legal regulations. Prophylactic measures and training, safety and protection not only of the workers but also of the population and the environment were self-evident ways of thinking and operating for nuclear specialists long before the environmental protection "boom" reached politicians and opinion-makers. Today even medicine begins to catch up again with the good examples of the industrial and nuclear fields, and the demands for training of medical personnel on all levels add to the other needs for training of professional personnel and students. Thus there exists a large "training gap" in radiation protection for which the existing educational institutions are inadequately equipped and staffed and often lack the indispensable practical experience.

At present it is most urgent to give at least the legally required minimum of training to all those professionally exposed persons which are working already with radiation sources and have finished their formal professional training long ago. But as a long-range goal the incorporation of a sufficient training into the formal professional training and educational curricula must be attempted.

### 3. The role of nuclear research centers

It is a general tendency in most countries that the "old" established and rather well staffed nuclear research centers have lost a good deal of their original tasks to industry and have to reorientate their programs. A good way to utilize the vast experience accumulated in these centers and their staff is to enlarge the training programs in the nuclear field. Radiation protection is especially well suited for this purpose. Not only have the nuclear centers been among the first to practice radiation protection, but their experiences usually also comprise most fields of radiation uses, possibly with the exception of some medical applications. Nuclear centers offer the unique advantage that the teachers and instructors can work part-time in practical radiation protection or in research and development and thus keep up-to-date in the field and abreast with the latest developments, but also in close contact to the field requirements of practical health physics. Teachers in the purely educational institutions are much more liable to easily lose the contact with practice. The training programs provide also a refreshing and stimulating feedback on the research and development programs of a research center's health physics division and help to direct those towards the real needs which otherwise might be more difficult to recognize. For the individual health physicist who in his part-time function as instructor or teacher has to present his specialty in an easily understandable way to laymen, the healthy effects of such experiences should be stressed, for he is each time challenged to reconsider, explain and defend what he does and why.

### 4. History of radiation protection training at EIR

The Health Physics Division had to deal with training problems from its very beginning in 1957 because its entire staff had to be trained on site or by participating in the few international courses available at that time which usually did not concentrate on health physics alone but on nuclear science in general. The first health physics technicians got an informal and individual training from the health physicists, but after a few years, at the beginning of the Sixties, a training group with a full-time instructor was created. Members of the division collaborated also with outside course activities by authorities and educational institutions. By principle even the "full-time" instructor was required to do part-time work either in routine or development in order to keep in contact with practice. With increasing training activities he was assisted by part-time instructors for practical exercises, recruited from the experienced health physics supervisors and technicians. The advent of nuclear energy created additional needs for training of health physics and reactor operating personnel. Between 1956 and 1971 some

2700 persons of various professions and levels received a short or extended radiation protection training from or with the cooperation of our division.

In connection with the modification of the institute's program which particularly emphasized training as one of the three main pillars, the training program in radiation protection was reshaped and intensified, creating a frame within which most of the present and future needs for radiation protection training in our country can be fitted or supported. This led in 1972 to the reorganization of the training group under the new name of "EIR School for Radiation Protection", still as a part of the Health Physics Division. A large variety of training courses are in various stages of realization, some operational for a long time, others in the experimental phase, still others in the preparatory stages. The number of participants in the training program increased to about 570 in 1972 and further to about 700 in the first half of 1973.

#### 5. Organization of the EIR School for Radiation Protection

The head of the school is a full-time instructor with a previous training as a health physics technician and an engineering school degree equivalent to a B.A. According to our principles he has also a function in routine health physics as head of the instrumentation and calibration group. He is responsible for the program of the school, the preparation of new courses and he teaches part of the theoretical topics. As assistant instructors for some theoretical teaching and most of the practical training he disposes of part of the working capacity of our health physics supervisors and some suitable technicians. This guarantees a practical instruction as close as possible to our practical experiences. The same supervisors and technicians spend part of their time assisting Swiss radiation protection authorities in the inspection of radiation protection conditions in hospitals, research laboratories, educational institutions and nuclear installations, including power stations, and cooperate in special protection problems outside the institute. Thus they have a broad experience and view to draw from for the training, but they get also useful feedbacks from the training and from discussions with course participants for their own work. Last but not least this brings our division in close, mutually interesting and beneficial contacts to almost all authorities, industries, institutions etc. which have any kind of radiation problems in our country. This will facilitate the introduction of similar viewpoints, philosophies and practices in all health physics applications.

#### 6. Preparations for new courses

Before a new course can be given, many months of preparations are necessary. As a first step a concept is worked out giving the goals of the course, the topics to be treated and instructed, possible examinations, qualifications of participants, duration etc. This concept is submitted to authorities and interested parties for detailed discussion. Sometimes this requires that the radiation protection philosophy for a certain profession or application must first be developed. If possible we ask the responsible authority to approve and recognize the course officially as a sufficient introduction or training for professionally exposed persons or for the competence in radiation protection required by radiation protection regulation. Next the detailed material for the course is prepared, such as course texts, exercises, experiments, demonstrations, practical training, visual aids etc. and all participating instructors are familiarized with the course. The course is announced and propagated as early as possible. Up to now the response rather surpassed our most optimistic expectations. Once a course has been given for the first time, the propaganda made by the participants creates additional requests for repetition of the course or for new or modified types.

#### 7. Execution of the courses

The theoretical instruction for the course types of similar levels has been standardized as far as feasible and split up into "building blocks" or packages

of topics which can be combined into any kind of course schedule or combination of subjects as desired, without having to work out each course entirely from the beginning. Only special topics required for certain courses need to be prepared individually, mostly for advanced courses that are not given all too frequently. The course texts are printed in large numbers, but on loose sheets, and are assembled into the requested combination and number of course textbooks for each course given. Changes and amendments of the texts are easy to make without delay or waste of outdated complete textbooks. Economics and flexibility are both optimized with this method.

In order to give all participants an efficient training in a minimum of time, courses with practical instruction are limited to 15 participants (with the only exception of the one-day monitoring course for army and civil defense where up to eight groups of ten men each can be handled in parallel). For the practical training the participants are further divided into two smaller groups.

Up-to-date methods are applied for instruction. The purely audiovisual method has been replaced by "simulation", which stands for "listen - look - touch - practice". The only purely audiovisual course is the short general introduction for new employees of the institute (D.1 Type D). Modern aids such as TV cameras and video recorder, overhead projectors, movies and slides are used. Detailed experiments, also simple, fundamental ones, and standard practical exercises facilitate the understanding of the often rather abstract subjects. We follow the development of modern didactic methods very closely and are ready to apply whatever will improve our courses. We cooperate with a new working group of the Fachverband für Strahlenschutz which has begun to treat problems of radiation protection training on all levels and which, as a first result, has collected an inventory of textbooks, training aids and training programs.

### 8. The program of the school

Four categories of training courses are offered :

- A) for full-time health physics personnel on all levels, from assistant or technician to supervisor, health physics engineer or health physicist. These are people from other technical professions which have to be retrained for health physics.
- B) radiation protection training for professionally exposed persons which have already completed their professional training or are further trained for a nuclear occupation such as reactor operator, radiochemistry laboratory technician etc.
- C) Personnel of emergency organizations, police, firemen, army, civil defense.
- D) Introductions to radiation protection fundamentals and practice for pupils, students and trainees in various types of schools and training programs.

All the above categories are further differentiated according to the level and previous training or experience of the participants, the type of their present or future occupation, the degree of competence or responsibility required etc. Appendix A lists the course program offered at present, while in the next section and in Appendix B four typical or novel courses are described in more detail.

### 9. Some examples of courses

#### 9.1. Training of health physics technicians (A.1, A.2)

If candidates for health physics technicians have had no previous training and experience in radiation protection, they have to begin at the bottom of the program with an introductory course. An extended version of this course is also given to health physics assistants, a category of auxiliary personnel, usually without other formal professional training, which we employ for simpler routine tasks such as wipe tests, decontaminations etc. The candidates for technicians, at least those for our institute, have been selected in cooperation with the Institute for Applied Psychology, Zürich. There they undergo a special aptitude test developed by a psychologist who had been one of our first health physics tech-

nicians before he began to study psychology.

The introductory course gives the candidates a first practical idea of the type of work while it allows us to judge the aptitude of the candidates and to employ them as assistants for some time, if for other reasons the following parts of the training must be delayed for some weeks. The 2 weeks' introductory course A.1 is followed by the 12 weeks' basic course A.2, divided into six basic and practical parts (details see Appendix B). The course A.2 is terminated by examinations, written and oral ones in theoretical knowledge, and a practical examination consisting of the preparation and execution of a routine health physics operation. The successful completion of these examinations qualifies the man for employment as health physics technician and for the advanced course A.2 of 4 weeks duration. He will usually take the advanced course after some weeks or months of practical experience in routine operations, and the course familiarizes him further with special monitoring techniques and the interpretation and application of the radiation protection regulations.

How far a technician will climb on the ladder of standardized careers in health physics, provided he is employed by EIR, depends on the results of the practical experience, additional training and examinations, and of course his personal qualifications, professional accomplishments etc. The chances to follow an interesting career that in some cases may lead up to the level of an engineer is of great help in finding and retaining good personnel. The flexibility of our system allows to make optimal use of each man's qualities and to offer him a job which fits his interests and capabilities without becoming boring or stagnant.

#### 9.2. Radiation protection training for mechanics and workmen (B.3)

This course, whose details are given in Appendix B, was originally started for mechanics and other workshop personnel who only infrequently had to do with contaminated or activated components from reactors or laboratories. The start of the first nuclear power stations in the country and the experiences during the first shut-downs quickly showed the necessity for training of all the operating and maintenance personnel in this type of course, and its content was accordingly adjusted. The course lasts one week and aims at giving the participants a practical understanding of radiation protection, the ability to work properly in controlled zones and under elevated levels of external radiation and contamination in such a way that they can protect themselves and do not endanger their coworkers. The use of protective clothing, of simple instruments for working place monitoring, and the reaction in case of incidents are instructed, thus enabling them to carry out routine tasks and repairs without direct control by a health physics technician.

#### 9.3. Radiation monitoring course for police and firemen (C.1 Type B)

This course type originated from concerns that grew within the fire department of Zürich airport after a crash landing of a foreign passenger plane, reports on the Palomares incident, and after the crash of another plane in Eastern Europe whereby a radioisotope cargo got lost. The firemen's problem was whether rescue operations for passengers might have to be stopped or delayed and could present an unacceptable risk for the firemen if radioactive cargoes were on board. The more general problem of what to do in case of a transport accident or a fire involving radioactive material applies also to police, ordinary fire departments of cities or industries, transport personnel etc. The first set of courses was given with excellent results for the fire department of Zürich airport, where the one week course will be followed at least once a year by some practical exercises and repetition, supplemented by internal repetition as part of the routine training of the firemen. The first part of the course aimed at giving an idea and a "feeling" for radiation dangers and relative risks. The institute's isotope production division provides excellent and realistic illustrative material and demonstrations. Monitoring of radiation and contamination and emergency measures form the main practical part of the course. From the first participants rumors spread quickly to other

fire departments and police, and it looks as if this course might become a frequent feature of our program.

#### 9.4. Radiation monitoring exercise for army and civil defense (C.2)

Army and civil defense have excellent and efficient training programs for part-time and full-time specialists in nuclear defense (AC) and monitoring. The training of AC officers is handled in central courses by professional instructors, while the soldiers and corporals (several in each unit) are trained in field courses by the AC protection officers of the regiments and divisions. Civil defense operates on similar principles. Training programs and material are excellent and efficient, but the army lacked possibilities for practical training with real radiation sources under simulated field conditions. Following a request and developing ideas of an AC officer of a division we improvised a first exercise with good success. In close cooperation with army instructors and AC officers we created a one day monitoring exercise with practical demonstrations on radiation levels and shielding, contamination and decontamination, and monitoring of a "fallout" area under field conditions. The four exercises of about one hour each are executed under the direction of the AC officer of a regiment and his staff and only supervised by our technicians.

The second part consists of a visit to the institute, a movie on nuclear energy, informations and discussions on radiation protection in civil applications, environmental effects of nuclear energy etc., thus providing some basic information useful for civil life and an opportunity to discuss problems that play quite some role in politics and newsmedia at present. As we give this course to several hundred army and civil defense personnel each year, with an increasing frequency, this may develop into an effective contribution to a better information of the public, because all these people return to civil life after some days or weeks of military or civil defense service and come from virtually all regions, social and professional groups of the country.

The individual doses received during the exercise are registered by direct reading dosimeters and amount in the average to a few millirem. During the information part the significance of these doses is explained and related to doses causing acute effects, to the natural background doses and the doses expected from radiation applications and nuclear energy. Such a personal experience helps to put radiation in proper perspective for the participants of the courses, they have also got the experimental proof for the efficiency of the protective measures. Results are an increased confidence, diminished anxieties about radiation hazards, and a better basis for rational judgements.

#### 10. Conclusions

Duration, topics and content of our courses are not yet in all cases what we judge optimal, sufficient or necessary, but they show what can be realized and required under present circumstances in various fields and professions with the acceptance and support of authorities, employers and participants, and on which lines a further development and improvement will be possible.

We hope to have shown how nuclear research centers with relatively limited means can provide valuable training opportunities in various fields, cover a good part of the training needs in radiation protection, and contribute efficiently to a better information and understanding of radiation protection and nuclear safety problems in professional groups and larger segments of the public. This may improve the chances for survival of such centers and their health physics staff and provide interesting and satisfactory opportunities for health physicists and the future development of health physics.

\*\*\*



Appendix A

List of courses offered by the EIR School for Radiation Protection

A) Full-time health physics personnel

A.1. Radiation protection course for health physics assistants

duration : 4 weeks for operations assistants, 2 weeks for technician candidates.  
participants : full-time auxiliary health physics personnel  
remarks : required introductory course (2 weeks) for candidates for health physics technicians who will continue with course A.2.

A.2. Radiation protection course for health physics technicians

Basic course A.2 :

duration : 12 weeks  
requirements: Course A.1 passed, former professional training in technical fields (mechanics etc).  
participants: candidates for full-time health physics technicians  
examinations: theory written and oral, practice.

Advanced course A.2 :

duration : 4 weeks  
requirements: courses A.1 and Basic A.2 + examinations passed  
participants: health physics technicians  
remarks : for details see section 9.1 and Appendix 5.

A.3. Radiation protection course for health physics supervisors

duration : 12 weeks  
requirements: examined health physics technician with several years of practical experience, qualifications for chief function,  
participants: health physics technicians  
examinations: theory written and oral. Practice: complete evaluation, preparation and execution of a large health physics operation.

A.4. Radiation protection course for health physics engineers

duration : 8 weeks  
requirements: diploma of a higher technical institution (engineering school) + autodidactic study of courses A.2 and A.3, if possible during stage at our health physics division.  
participants: future heads of operational health physics sections in nuclear power stations, laboratories or authority inspection groups  
examinations : theory written and oral, practice.

A.5. Radiation protection repetition course for health physics technicians

duration : 1 week

A.6. Radiation protection repetition course for health physics supervisors

duration : 1 week

B) Professionally exposed persons

B.1. Radiation protection course for radiochemistry laboratory technicians

Type A : Basic course (short course)

duration : 1 week

participants: laboratory technicians and auxiliary personnel

Type B : Advanced course

duration : 2 weeks

participants: laboratory supervisors and technicians

B.2. Radiation protection course for factory-inspectors

duration : 3 weeks

participants: cantonal and federal factory-inspectors or persons responsible for radiation protection and industrial hygiene in industry

B.3. Radiation protection course for mechanics and workmen

duration : 1 week

participants: professionally exposed workers employed for assembly, maintenance, repairs, shut-down operations of nuclear components and installations.

remarks : for details see section 9.2 and Appendix B

B.4. Radiation protection course for technical X-ray personnel

duration : 1 week

requirements: fundamentals (physics and technology) of X-ray equipment

participants: sales-, repair-, assembly- and operating personnel of X-ray equipment for industrial or medical applications

C) Emergency organization personnel

C.1. Radiation protection monitoring course

Type A : EIR emergency organization

duration : 2 days

participants: EIR emergency organization members

Type B : Professional emergency teams

duration : 1 week

participants: fire department chiefs and officers, professional firemen of airports and cities, police etc.

remarks : for details see section 9.3 and Appendix B.

Type C : Enclosed radiation sources (handling of incidents)

duration : 2 days

participants: workers who operate or are responsible for enclosed radiation sources

Type D : fire-fighting in radiochemistry laboratories

duration : 1 day

participants: voluntary and industrial firemen

C.2. Radiation monitoring exercise for army and civil defense

duration : 1 day ( 8 hrs)

requirements: formal military or CD training in AC defense

participants: radiation monitoring and protection personnel and officers of army and civil defense

remarks : for details see section 9.4 and Appendix B

D) Introductory courses of general nature

D.1. Radiation protection introduction

Type A : professionally exposed personnel

duration : 2 days

participants: new employees to become professionally exposed

Type B : Educational institutions

duration : 3 hrs

participants: students of higher educational institutions

Type C : Hospital personnel

duration : 3 hrs

participants: professionally exposed personnel of hospitals (including M.Ds)

Type D : new EIR employees

duration : 1 hr

participants: all new employees of EIR, whether professionally exposed or not, including all auxiliary or temporary personnel

E) Radiation protection as part of nuclear training courses

E.1. Radiation protection course for reactor operators

duration : 2 weeks

requirements: complete training as reactor operator in EIR Reactor School

participants: candidates for operators licence

E.2. Radiation protection course for reactor shift supervisors

duration : 1 week

requirements: licensed and experienced reactor operator

participants: candidates for shift supervisor examination (EIR Reactor School)

remarks : E.1 and E.2. are limited to practical instruction, the theoretical training is given by the Reactor School

E.3. Radiation protection course for nuclear engineers

duration : 1 week

requirements: nuclear engineering lecture program

participants: students and graduates of engineering schools

Additional radiation protection courses for incorporation into other curricula of higher education or professional training in science, technology and in the medical field are being discussed and prepared.

\*\*\*

Appendix B

Examples and details of some courses

1) Courses A.1 and A.2 : Training of health physics technicians

A.1. Introductory course (2 weeks)

aim : Review of the fundamentals and basic philosophy of radiation protection and of the routine tasks of a HP technician

organization: 14 lectures with short preparatory theories and main emphasis on practical demonstration and exercise of the routine tasks

topics: - fundamentals: radioactivity / radiation, sources, hazards / basic principles of radiation protection / radiation monitoring, doses, dosimetry / contamination, decontamination / waste problems.

- practical training: marking of controlled zones / radiation monitoring

instruments and dosimeters / protective clothing / contamination monitoring and decontamination / waste collection in controlled zones / behavior in case of incidents.

A.2. Basic course (12 weeks)

aim : complete training of health physics technicians. Knowledge of fundamental physics and technology required for health physics. Mastery of all routine tasks and methods for working place control

organization: 6 parts: 3 basic, 3 practical. The fundamental lectures are illustrated by experiments. Exercise of simple calculations. Main emphasis on mastery of radiation monitoring methods.

topics: Basic instruction 1 : fundamentals of nuclear physics (2 weeks)  
composition of matter / radioactivity / radiations / radiation interactions with matter / dose definitions / shielding

Basic instruction 2 : radiation monitoring methods ( 2 weeks)  
electrical and electronic fundamentals / principles of measuring/ principles of radiation detection / ionization chambers / proportional counters / GM counters / solid state detectors / scintillation detectors / chemical detectors / detection and monitoring of  $\alpha$ ,  $\beta$ , X,  $\gamma$ , n.

Basic instruction 3 : hazards of ionizing radiations to man (3 weeks)  
radiation exposures of man / biological effects of ionizing radiations / external exposures / internal exposures / maximum permissible doses and derived working levels .

Practical instruction 1 : health physics at nuclear reactors (1 week)  
physics, technology of nuclear reactors / reactors as radiation sources and their hazards / risks of nuclear reactors.

Practical instruction 2 : HP in radiochemistry laboratories (1 week)  
fundamentals of radiochemistry / installations and organization of radiochemistry labs / types of work in a laboratory / radiation hazards and risks in radiochemistry laboratories.

Practical instruction 3 : operational health physics methods (3 weeks)  
routine methods (monitoring etc) / methods as specified by HP operational manuals / individual handling of routine operations and problems at reactors and in laboratories, under supervision of an experienced HP technician.

examinations : Theory and fundamentals : written and oral  
Practice : complete health physics control of a routine operation in a controlled zone. Several experts from the school, the HP division or other EIR divisions or from authorities are present.  
The examinations are formally acknowledged with a diploma.

A.2. Advanced course (4 weeks)

aim : repetition and advanced treatment of basic course A.2 topics. Special monitoring methods. Interpretation and application of the federal radiation protection regulations for special cases.

organization : introduction of new topics followed by discussion of practical situations, aimed at integration of theory and practice.

topics : those of basic course A.2 / air monitoring / background compensation / activity measurements / radionuclide identification and analysis / spectrometry / DWL derivation / practical problems.

remarks : for the technicians of the SIM accelerator the basic course A.2 was supplemented by an additional practical course on accelerator problems.

\*

2) Course B.3. Radiation protection for mechanics and workmen ( 1 week)

- aim : See section 9.2. Ability to handle practical radiation protection problems when working in controlled zones
- organization: as course A.1.
- topics : - fundamentals : short version of topics of course A.1. / evaluation of hazards and protective measures when handling and machining contaminated or activated objects.
- practical : monitoring instruments and methods for external radiation and surface contamination / behavior in controlled zones / installation of temporary zones / protective clothing / active working methods / decontamination of persons and material / waste handling/ transport regulations / practical exercises in active workshop.

3) Course C.1 Type B : Radiation protection monitoring for emergency teams(1 week)

- aim : See section 9.3. Mastery of handling the first emergency actions at the site of a transport or other radiation incident. Fencing off the site, cooperation with special teams, minimizing of risks for public and environment, self-protection during rescue etc.
- organization: as course A.1.
- topics : fundamentals : as course B.3 with main emphasis on emergency situation
- practical : monitoring instruments and methods / behavior under high radiation and contamination levels / protective clothing / practical exercises of simulated accident situations.

4) Course C.2: Radiation monitoring exercise for army and civil defense (1 day)

- aim : Demonstration and realistic, but separate exercises of problems occurring in a fallout situation. Information on civil radiation protection and nuclear safety.
- organization: groups of max. 10 men, rotate between 4 posts for 4 hours, second part (visit etc) four or all groups together.
- topics : - post A : decontamination as a "household" problem. Contamination and coarse decontamination of military clothing with simulated fallout ( $Tc-99m$  labelled  $Al_2O_3$  grains with diameters of 70 - 150  $\mu m$ ).
- post B : liquid contamination and decontamination of shoes and hand. Spreading of liquid contaminant ( $Tc-99m$  labelled glycol), decontamination with water, soap and brush.
  - post C : Shielding factors and inverse square law. A  $Cs-137$  source in a rectangle of walls of earth, wood and concrete, fourth side open, is monitored from all directions at various distances.
  - post D : Radiation monitoring of a simulated fallout field. On an area of 2000  $m^2$  a reduced scale model (1:80) of routes for monitoring teams is laid out, spiked with camouflaged  $Cs-137$  sources of various activities, simulating an extended contamination. The 2 man teams with ordnance equipment have to monitor a total of 41 points, calculate H+1 hr normalized dose rates and draw the contamination map on the real topographical map, control personnel doses etc.
  - Part II : Introduction to civil radiation protection and regulations, movie on nuclear energy, visit to EIR reactors with demonstration of operational health physics, environmental monitoring, waste treatment, protective clothing etc. Discussions according to particular interests.

\*\*\*\*\*

## PROJECT TRAINING FOR GRADUATE STUDENTS IN HEALTH PHYSICS

J.R.A. Lakey

A.V. Yorke

Royal Naval College, Greenwich  
London SE10, England

### ABSTRACT

The Royal Naval College has conducted graduate training in health physics for over 10 years and has developed a system of project training which gives students practical experience in the context of the formal theoretical course. The purpose of project training is to provide realistic radiological protection problem solving at a time when the student is consolidating his theoretical knowledge.

This paper describes the procedures used to introduce, supervise, and assess these projects.

The projects are initiated early in the course by presentation of a package which includes a statement of the aims of the project, copies of relevant papers, apparatus manuals, radioactive source calibrations and guide lines for progression of the project. The student is required to write a report on his project which is presented orally and staff appraisal takes account of all aspects of the students project work.

The paper evaluates the success of this approach against the cost in staff and equipment resources required to achieve realistic and effective training.

### INTRODUCTION

The Royal Naval College, Greenwich, has conducted graduate courses in health physics for over 10 years in association with other specialist post-graduate courses. The Department of Nuclear Science and Technology, which is responsible for these courses, was founded in 1959 to provide a centre for Naval Nuclear Education and Training of officers and now offers over 18 courses per year to a total of about 250 students. All these courses contain an element of health physics. The necessity for health physics in the training of reactor engineers is already well recognised<sup>2</sup> since they must understand the hazards of radiation and, as reactor operating personnel, they will receive the largest doses during reactor plant down time for repair or preventative maintenance. The experience on which this paper is based was gained over the last 10 years in two specific graduate courses, one of 12 weeks duration and the other 24 weeks.

### GRADUATE COURSES

The first course, the Nuclear Radiation Protection Course (NRPC) has

already been reported<sup>3</sup> but more recently another graduate course, the Nuclear Reactor Course (NRC), has been extended to include radiological protection as an essential qualification. The NRPC is recognised by the British Institute of Physics and the successful NRC candidate also receives the Postgraduate Diploma of the Council for National Academic Awards.

#### DEPARTMENT FACILITIES

The facilities available for training in the Department of Nuclear Science and Technology include the 10 kW training and research reactor JASON<sup>4</sup> which was the first to be installed in any educational establishment in the United Kingdom. This has given the staff considerable first hand experience and set a pattern in terms of safety documentation, procedures and applications to other Universities. The Reactor is used to bridge the gap between simulator training and full power reactor training and provides a source of radiation which is used in many supporting training experiments and projects. In particular it provides a realistic environment for students to learn how to handle, survey and control sources of ionising radiation. The reactor is used for 49% of the time for student training and diverges to power over 480 times per year. Over 2000 students have been trained and currently the Department provides 8 different types of courses. The research utilisation of the reactor includes several applications of activation analysis and reactor dynamics in addition to specific health physics studies such as reactor shielding<sup>5</sup> and the study of radioactive aerosols<sup>6</sup> and some dosimetry studies.

#### PROJECT TRAINING

The interdisciplinary nature of environmental health engineering has been emphasised by the World Health Organisation<sup>7</sup> in their booklet on The Education and Training of Environmental Health Engineers, which states that many disciplines may be required for the solution of complex environmental health problems.

This generalisation is specially true in health physics; team operation is the rule and the individual member must be familiar with the vocabulary, techniques and goals of other members of the team.

This paper describes an approach to graduate training in health physics which is designed to develop the student's awareness of other disciplines, and to give him practice at problem solving under realistic conditions. The two courses on which the experience was gained are being reviewed by objective training analysis which requires the definition of an Operational Performance Standard. This is translated into a Training Performance Standard which provides the basis for the detailed course design. All this demands effective feedback on performance of past students and also of the reactor plants on which they have worked. The replacement of conventional set practical sessions by a smaller number of set practicals and project work has emphasised the need for careful attention to the objectives of the course. The projects themselves must be well organised to ensure full benefit for the student and effective appraisal of his success.

#### PURPOSE OF PROJECT TRAINING

#### DEFINITION OF A TRAINING PROJECT

A training project is a supervised task set by the students' tutor to meet some objectives of the course. A good project must be interesting to the student but limited in scope so that it can be completed within the time allocated. The project must be realistic to permit the student to recognise

a 'real problem'; novelty and relevance provides the necessary motivation. To achieve these conflicting features the development of training projects relies to a large extent on feedback from operational establishments and spin off from departmental research projects.

The purpose of project training must be examined in relation to the overall objectives of the course. A common feature in the objectives of health physics courses is the achievement of three important attributes which the successful student must possess. The student must:

- a. have a sound knowledge of the process which produces the hazard,
- b. understand the hazards of radiation,
- c. have sufficient appreciation of practical problems to be rapidly accepted into the operating team.

The development of these attributes is an important objective of the course but it can not be met entirely by classroom instruction, especially when the students themselves are practical men. Conventional practical work can be designed to re-inforce classroom instruction or to help students familiarise themselves with equipment and techniques, but it may stultify the students interest. When practical work has to be allocated in short periods of a few hours at one time, the student will rarely have the opportunity to use an interdisciplinary approach and the stereotyped exercise limits his scope for problem solving. On the other hand, set practicals are straight forward to administer, and it is comparatively easy to assess the students performance against that of his colleagues because the work expected is identical.

#### PROJECT OBJECTIVES

The approach described in this paper replaces most of the set practical sessions by more broadly specified projects designed for the following purposes:

- (1) To apply the student's theoretical knowledge acquired from the course work to problems associated with the operational situation.
- (2) To give direct experience of relevant health physics practice.
- (3) To emphasise the interdisciplinary nature of health physics.
- (4) To assess the student's ability to solve problems under realistic conditions and his ability to communicate his observations and recommendations.

The first two objectives replace the set practical but added motivation can be imparted when the project contains an element of novelty. The third objective requires an input from other disciplines such as reactor physics, reactor engineering, chemistry, metallurgy and radiobiology. This is important to ensure that the student is made aware of the relevance of his work to overall plant safety. The last objective reveals the special advantage of project training and, to achieve it, the class have to share experience gained on the individual projects by participation in a formal presentation of the project reports.

#### PROJECT MANAGEMENT

The introduction of project work and its subsequent extension to become a significant proportion of Course time necessitates the designation of a Project Manager. The Project Manager is responsible for coordinating individual project supervisors to ensure the satisfactory progress of the projects. These supervisors are required to carry out the following tasks:-



- (1) The production of the project outline (in association with the Project Manager).
- (2) The day to day supervision of the project.
- (3) The timely presentation by the student of a project report and the provision of guidance as to the required standard.
- (4) The assessment of the report in conjunction with at least one other member of staff.

#### PROJECT PACKAGE

The project work described in this paper is given to students undergoing relatively intensive training in which the duration of projects is strictly controlled. It is therefore essential that the objective and project outline are clearly defined. The use of a 'project package' has been developed in an attempt to maximise the benefit to the student.

The project package is required to:

- (i) Provide the student with sufficient information on which to make his choice of project.
- (ii) Ensure that sufficient staff work has been undertaken to permit the completion of a worthwhile project.
- (iii) Enable a comparison of the project proposals to be made in terms of the level of the work involved.
- (iv) Provide the Project Supervisor with the basis for monitoring the progress of the project.

The project package includes:

- (i) The background information necessary to place the project in context.
- (ii) The objective of the project.
- (iii) The schedule of apparatus to be made available and the apparatus manuals.
- (iv) The guide lines for initiation of the project.
- (v) Selected reference material.

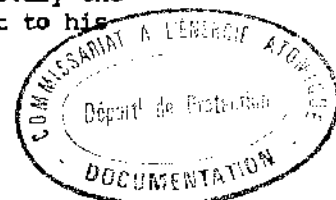
Having made his choice, the student is provided with sufficient information to commence some basic reading and planning before any further discussions are held with the supervisor. In this way the student is able to take some part in the initiation of a project. A typical project package is summarised in Appendix 1.

#### PROJECT SELECTION

Three factors are considered in the selection of the individual projects:

- (1) The previous experience of the individual student;
- (2) the future task in which student will be engaged;
- (3) the student's performance.

It is sometimes possible to remedy obvious deficiency in the student's past experience by the selection of an appropriate project. Alternatively the student's best interests may be served by completing a project relevant to his



future commitment. The student is encouraged to discuss the various projects with the supervisors so that the selection also reflects his personal interests.

#### PROJECT SUPERVISION

The responsibilities of the project supervisor would appear to diminish once the project is launched since the student has sufficient background information. Experience has shown that the supervisor must act in an advisory capacity, reviewing progress at intervals determined by the duration of the project. In this context the student-supervisor relationship is important, and an informal approach has been found to provide the ease of communication which is an essential part of project work.

In the event of a major equipment failure, or a particularly interesting unforeseen development, the supervisor may redirect the project to capitalise on the situation. The project work culminates in a formal report and the supervisor is required to review the draft report and make constructive suggestions and query any doubtful aspect of the student's work.

#### PROJECT APPRAISAL

Within one course all the projects may have different main topics and therefore different supervisors. Although the expected work content can be assessed by the Projects Manager, the problems arising in the execution of the project may require a different work content and the demands of the individual supervisors cannot be standardised. This situation may generate some difficulties in achieving a fair comparison of each project. The supervisor is therefore made responsible for the preparation of a written summary of the report which takes account of these variable factors.

The project which carries up to one third of the course marks is finally evaluated under the following three headings:-

- a. Methodology and Practical Work. The orderliness of the approach adopted by the student and the design and execution of the project is examined to establish his penetration of the problem.
- b. Written Report. This is examined to provide further appreciation of the student's reasoning powers and orderliness in presentation.
- c. Oral Presentation. This reveals the student's ability to exercise judgement in the selection of the important aspects of the work and his performance in answering questions on the project demonstrates the depth of knowledge and comprehension. Since all the Course members and the examiners are present at the oral presentation, it serves to acquaint the other students with the subject and helps to share particular lessons that have been learnt.

#### EVALUATION OF PROJECT WORK

#### STAFF RESOURCES

At first sight the project approach to training appears to be more costly in both time and resources than formal practical work. This depends on the size of the course and the degree of utilisation of the laboratory equipment. For example, 6 groups of students could perform a set practical either (a) simultaneously and therefore requiring 6 sets of equipment, or (b) in three separate sessions requiring 2 sets. The solution adopted is usually constrained by the overall course programme, which determines the timing of the practical sessions. Since the RNC courses involve practical work in several topics it

has been policy to arrange several different practical sessions in parallel, the students completing each experiment in the various laboratories. Hence the laboratory utilisation achieved is dependent on the size of the Course, but on the average is equivalent to 3 sessions per experiment per course. This policy has ensured that, with the exception of the very low cost equipment, only a limited number of sets of equipment are required.

The development, documentation and updating of the conventional practical training also requires considerable resources and Tables 1 and 2 compare for the same allocation of practical time, the total man-hours of staff time for formal practical and project work. The Tables are based on 8 students on the course with the breakdown of laboratory work being typical, rather than specific to any one course: development costs are calculated on the assumption of a review of all experiments after every 2 to 3 courses.

TABLE 1. CONVENTIONAL PRACTICAL

TASK	STAFF HOURS	SESSIONS	NUMBER OF PRACTICALS	TOTAL TIME HOURS
Administration	20	1	-	20
Development	3	20	-	60
Basic Radiation Protection	6	2	10	120
Basic Radiation Physics	6	2	4	48
Basic Reactor Physics	8	2	4	64
Simulator	4	2	2	16
Examination of Practical work	1/5	8	20	32
TOTAL TIME:				360 hours

The calculation for the project work is based on 8 students each undertaking a different project, with the support of some basic introductory experiments and is chosen to illustrate the maximum staff effort. Some formal practical work must be included to ensure that the student is familiar with the basic radiation laboratory procedures and techniques. These costs could be reduced by combining students into groups of 2 or more for each project but this removes some of the advantage of project training.

TABLE 2. PROJECT WORK

TASK	STAFF HOURS	SESSIONS	NUMBER OF PRACTICALS	TOTAL TIME HOURS
Project Administration	3	1	8	24
Development and Consultation	2	1	6	12
Planning and Preparation	10	1	8	80
Supervision	15	1	8	120
Assessment	4	1	8	32
Presentation	3	1	8	24
Basic Radiation Protection	6	2	2	24
Basic Radiation Physics	6	2	2	24
Basic Reactor Physics	8	2	2	32
Examination of Practical work	1/5	2	6	3
TOTAL TIME:				375 hours

These figures which are based on several years experience demonstrate that the cost in terms of man hours differs little between the two alternative schemes. In the planning and preparation of projects the specialised experience of the supervisor is used and this is more stimulating for him than the

development of set practicals to achieve more limited objectives. A survey of the equipment resources required for mounting the work described in Table 2 has shown that as a result of the extensive use of project equipment in research and the flexibility in planning a reduction of 25% in the capital equipment cost was possible compared to conventional practicals.

#### COMMENTS ON THE STUDENTS RESPONSE

##### STUDENT REACTION

The total time allocated for participation in practical work in set practicals and in projects is the same. However, the student reaction to the laboratory work in each scheme is noticeably different.

The students on intensive courses will quickly form opinions on the relevance and necessity of the practical work and will reject unnecessary duplication in the presentation of the material. Set practicals with the usual close relation to the lecture material are frequently rejected as repetitive. In contrast projects have stimulated student interest and participation and help to ensure that the objectivity of the course stands up to close scrutiny by the students. One measure of the success of the project is the amount of additional time a student may be prepared to devote to the work; in fact it is common for the supervisor to have to ensure that the student does not spend an excessive amount of time on the project.

##### STUDENT ATTAINMENT

The effectiveness of any instructional technique in attaining some part of the course objective cannot easily be objectively assessed and subjective assessments tend to vary widely. The ultimate test is to follow up the students when they have moved on to their operational role. If the staff effort is available the students are interviewed in their work area at least one year after completing the course. Students do appear to move smoothly into their operational task, in some cases continuing to follow up the project topic as a centre of interest in their new job. This itself is a convincing demonstration of the value of this approach to the student.

#### CONCLUSIONS

The procedures described in this paper are offered as a product of systematic course design which could be applied to other graduate courses in health physics. The two courses on which this work has been developed are relatively long - 12 weeks and 24 weeks respectively - but it is considered that the method can be applied with success to shorter courses. The reliability of the apparatus used and the provision of guidelines for the project becomes important if the student is not to waste valuable time coping with instrument faults or re-discovering relatively unimportant information. In all cases the supervisor must ensure that there is sufficient scope for problem solving and that the project does not relapse into meaningless collection of data. The experience and judgement of training staff are taxed more heavily in the project supervision than the more passive role of monitoring a set practical. In many cases the student gains his first rigorous experience in scientific communication when he prepares his project report and presents it to his colleagues and the examiner. It is probably this increased demand on the supervisor and student which ensures the success of this approach to health physics training.

PROJECT PACKAGE

The package contains the following main items:-

1. Objective of Project
2. Background information
3. Equipment and Services supplied
4. Guide lines for progress
5. Literature references and Instrument Manuals

To illustrate this the following is an abbreviated package for a Thermoluminescent Dosimetry Project:-

1. Objective: To investigate factors affecting the precision of TLD-700 for dosimetry in low level photon and neutron fields.

2. Background: The operation of a nuclear reactor involves the staff in radiation exposure to mixed photon and neutron radiation fields. Whilst the radiation levels may be low compared to the legal limits there are both ethical and practical reasons for aiming at high precision in these measurements. For example, the success of a shield design may be judged by the man rem accumulated by the operating staff or a new reactor plant may be surveyed by short duration exposures of TL dosimeters.

3. Equipment and Services Supplied:

- i. Access to the research reactor
- ii. One Dynatron TLD reader modified to give a graphical display of glow curves
- iii. Annealing ovens
- iv. Dispensing equipment for powder, extruded chip and disc forms of TLD

4. Guidelines:

- i. Delineate radiation fields to be studied taking account of operational conditions on a power reactor against the closest approach available on the research reactor.
- ii. Gain familiarisation with TLD equipment by trial runs on irradiated samples
- iii. Assess the number of tests feasible in the time available and schedule test points
- iv. Execute selected measurements, read and analyse data for precision
- v. Run subsidiary experiments after discussing (iv) with Supervisor
- vi. Write draft report

5. Literature References and Instrument Manuals:

References and manuals are revised and amended for each project.

## REFERENCES

1. EDWARDS J.  
Courses in Nuclear Science and Technology at the Royal Naval College  
Symposium on the Education and Training of Engineers in the Nuclear  
Industry. The Institution of Mechanical Engineers, 5 December 1968.  
Proceedings 1968-69. Volume 183 Part 3F page 12.
2. ELLIOTT Jr N.S.  
The Present and Future Training of Nuclear Engineers.  
Babcock and Wilcox, p 401. Nuclear Engineering Internal May 1973
3. LAKEY J.R.A.  
Training in Radiological Protection for Reactor Health Physicists.  
International Radiation Protection Association Congress, Brighton,  
England, 3-8 May 1970.
4. DUNTON P.J., EDWARDS J.E., LAKEY J.R.A., et al  
Utilisation of British University Research Reactors.  
Fourth UN International Conference on the Peaceful Uses of Atomic Energy.  
6-16 September 1971. Ref A/CONF. 49/P/485
5. LAKEY J.R.A., MARTIN A.  
A monitoring system for the assessment of reactor shield performance.  
IAEA Symposium on New Developments in Physical and Biological Radiation  
Detectors. (Nov 1970) Vienna.
6. LAKEY J.R.A.  
Control of Airborne Contamination arising from the operation of the  
JASON Research and Training Reactor.  
International Symposium on the Radiological Protection of the Worker  
by the Design and Control of his Environment.  
Soc for Radiation Protection, Bournemouth (April 1966).
7. LOGAN J.A. (Editor)  
The Education and Training of Engineers for Environmental Health 1970.  
World Health Organisation, Geneva.

THE LAW AND LOW LEVEL RADIATION  
Charles F. Eason and Natalie Y. St. Denis  
U. S. Atomic Energy Commission  
Washington, D.C.

Abstract

One of the responsibilities of the health physicist is to recommend and enforce radiation protection standards based upon recognized occupational radiation exposure guides. It follows, therefore, that the health physicist, because of his background and technical knowledge, will play a key role in evaluating a claim allegedly arising out of exposure to ionizing radiation.

It is the purpose of this paper to examine a number of latent radiation injury cases with particular emphasis on the kinds of radiation records offered in evidence, the nature of the expert testimony by both the health physicist and the medical expert and the conclusions of the court or Board in the final adjudication of the claim. The paper will also explore the views of those who hold that the present legal system in the United States is not appropriate for the handling of latent injury claims.

Introduction

It is well known that in spite of the highly successful efforts of those engaged in the field of radiological health, radiation workers run a risk of being exposed to some degree of radiation in the course of their employment--however small that risk may be. Radiation workers may also develop certain diseases that are known to be caused by radiation but which also develop spontaneously in the absence of radiation exposure. What happens in the United States when a radiation worker sues for compensation on the basis that the disease from which he is suffering was incurred in the course of his employment?

In cases involving substantial exposure, recovery is almost always assured. However, in cases involving delayed injury, the claimant rarely recovers. Should he?

The purpose of this paper is to examine this question.

In order to understand the reasons why so few cases involving low exposures and delayed occupational injuries are compensated, one must appreciate that it is first necessary under our legal system to determine whether the exposure "caused" the injury. It is also necessary to draw a distinction between the two categories of radiation injury cases: (1) Claims involving acute effects which appear immediately or within a short period after a very large exposure; and (2) Claims involving latent effects which do not manifest themselves until years later. In the first instance they offer the litigant, the expert

witnesses and the courts little difficulty. Dosimetry can be reconstructed, and the symptoms or illnesses are usually characteristic. Because the nexus between the pathology and the disease is obvious, causal relation is relatively simple to establish. It is the second category of cases that pose problems of causation. Why?

#### Causation

Simply stated, it is because the proof of causal connection in a claim for injury, if we accept the medical notion of the meaning of the term, is, in most instances, extremely difficult to establish.

In a radiation claim both the physician and the court must confront the problem of causation. When the medical expert is asked to testify concerning the cause of the claimant's pathology it frequently develops that his testimony is so couched with reservation that the court or board has no alternative but to make the medical determination on its own. The reason for this hesitancy or inability on the part of the medical profession to find causation lies in its notion of the meaning of causal connection. Doctors define causation in a special sense. They prefer to base their conclusions on statistical studies of the relationship between a suspected causative factor and the disease.<sup>1</sup> In 1969 an outstanding authority on workmen's compensation pointed out that longitudinal studies, using an exposed population group and one or more control groups are a principal device for testing causal theories involving human pathology and that longitudinal studies yield information concerning probabilities of causal nexus in a population. But the legal problem, always, is to determine causal nexus in each individual case. "The unspoken medical assumption is that causation in a legal proceeding is a pure question of scientific fact!"<sup>2</sup> However, in the absence of a suitable test which could be used to establish the dose-response relationship at low range, the medical expert is willing to assume for purposes of conservatism that no threshold exists and that linear build-up may possibly result in adverse biological response. But when the etiology of a disease remains unknown, the medical expert will not under most circumstances say that the exposure "probably caused the disease." The reasons are crystal clear. At the present state of scientific knowledge it is simply not possible to relate individual response to disease, nor does the solution appear imminent. Some fourteen years ago a view was expressed before a Congressional Committee that "The more that is discovered about the complex etiology of disease . . . the less it appears possible to identify causality, and the more we grow dependent upon vague and arbitrary interpretations, with inevitable inequities . . ."<sup>3</sup>

When an employee develops a disease which conceivably could have resulted from occupational exposure, should the economic loss fall on the employee or should it be shifted to the employer? The courts in the United States in many instances have taken a rather juristic view in response to this question. It is clear that the courts need not be bound by medical notions of causation for medical evidence indicating a distinct possibility of a relationship between the job and the disease, while insufficient to support a finding of causation in the medical sense, may warrant a finding of causation in the legal sense. Our workmen's compensation laws in the United States typically include a mandate that they should be liberally construed to protect the employee. This being the case, decisions not to compensate where the exposures are small but, nevertheless, the employee is suffering from a radiation connected disease, may very well be inconsistent with the policy and purpose of our workmen's compensation laws.

Unfortunately most courts take the position that they cannot deviate from the requirement of medical probability. For example, in a recent Texas Supreme Court case<sup>4</sup> in which the claimant was denied compensation, the court found



that the evidence did not indicate the existence of "a reasonable medical probability" of causal connection between petitioner's cancer and radiation but merely "the possibility" of such a connection. In drawing what it termed a logical distinction between a "reasonable medical probability" and a "medical possibility" the court said, ". . . a possibility becomes 'probable' when in the absence of other reasonable causal explanations it becomes more likely than not that the injury was a result of its action."

However, the opinion of the dissenting judge is worthy of note. He found that the expert medical testimony indicated that the etiology of cancer is really unknown, that the claimant's cancer could have been caused by radiation, but that there was no way to determine the cause of a particular cancer; that it is possible for a person exposed to radiation over a long period of time to develop cancer, but that it could not be stated how much exposure would be required; that any radioactive material can conceivably cause cancer on prolonged exposure; that anyone exposed to certain amounts of radiation has a higher than normal risk of developing malignant changes in the body tissues but that in this particular situation a diagnosis of probability either way could not be made.

Further, he rejected the weight the court placed upon the medical opinion evidence of experts who refused to testify that the cancer was "probably" caused by the radioactivity to which the petitioner was exposed. In his dissenting opinion he stated, "We are not to isolate the testimony of the doctors, but must determine the effect of such testimony upon other evidentiary proof in the case."

This statement was followed up with a reference to the spirit of workmen's compensation statutes and the need for liberal interpretation. He admonished the court for apparently forgetting "for the moment" the purpose of the Texas Workmen's Compensation Act. He stated that to hold, as the court held, that the evidence as a whole, which the jury considered in reaching its conclusion, did not meet the standard of proximate causation with sufficient certainty to impose liability upon the insurance carrier of claimant's employer, "is to effectively remove injuries which require medical testimony to substantiate causation from the common law of tort."

#### Health Physics Testimony

Now when it comes to dealing with roentgens, rads and rems and the recommendations of the various standard making bodies, we find that this is the field of expertise of the Health Physicist. It thus follows that because of his background and technical knowledge and the complicated array of terminology which finds its way into a radiation claim, the health physicist will play a key role in evaluating an individual's radiation exposure. The courts cannot adjudicate and the medical expert cannot opine until the source, duration and amount of exposure is known. In fact, the health physics experience of the injured claimant is one of the most important factors in any claim involving radiation injury and the health physics testimony and evidence is of extreme importance because it protects medical experts by assuring them that their testimony and opinions are predicated on the most probable exposure.<sup>5</sup> Furthermore, the health physicist supplies and interprets available radiation records of claimants' external and internal occupational exposure (e.g., film badge and other types of dosimeter records, whole body counter records, records of bioassay data and interpretation, etc.) and he supplies estimates of exposure in the absence of radiation records and interprets other records relating to the claimant's exposure (e.g., records of work orientation and training, radiation and contamination survey reports, records relating to the radiation status of the claimant's work area and records relating to the employer's radiation protection program.)

Although it is the considered view of many lawyers familiar with radiation litigation that "records are the most important item in establishing dose from a legal standpoint;"<sup>6</sup> there are still members of the health physics establishment who, I regret to say, maintain that the record developed on a day-to-day basis to assure the safety of the radiation worker has little value 15 to 20 years later when that very same record is introduced in a workmen's compensation proceeding as evidence of exposure. However, it is apparent from a reading of the cases that not all health physicists agree with this position for they have on many occasions furnished courts and boards with convincing evidence of the claimant's exposure from the radiation exposure records in their possession.

For example, in almost all of the claims containing allegations of radiation injury, radiation records (e.g., dosimetry, bioassay and other records related to the claimant's exposure) are not only supplied by the health physicist but are examined and referred to by the health physicist and the medical expert in deciding causation.

By way of illustration, in a case involving a blood disorder a health physicist testified that records showed the premises where the claimant worked were found to meet the standards of radiation protection as recommended by the National Committee on Radiation Protection. He also testified that film badge reports indicated all exposures were below permissible dose. The radiation expert in the case noted that after reviewing the case file, film badge readings and the physicist's survey of the environment, it seemed apparent that claimant's exposure had been at a relatively low level which would not be expected to give rise to incapacitating bodily injury. Compensation was denied.

In another case involving exposure to radioactive tracers for a five-year period, a medical expert stated, after review of the records:

"Beginning with the exposures received, we find that these are well documented and do not appear excessive . . . . Dosimetry appears to have been reliable and film badge and monitoring reports indicate that the decedent's exposures were well below those considered maximum permissible. . . ." Compensation denied.<sup>8</sup>

The most popular argument advanced by those who discount the value of radiation records is the unreliability of monitoring devices to record low exposures. Yet from my own experience I know that a properly organized health protection program can and does furnish data which provides a pretty good estimate of the maximum exposure which the individual worker could have received in the course of his employment. In fact, there is testimony from the medical establishment that negates the view that records have no value. In a recent case in which the claimant wore no film badge a medical doctor noted: ". . . [claimant] wore no protective badge which would have adequately monitored his X-ray exposures." Compensation denied.<sup>9</sup>

While recorded exposures are valuable evidence in a radiation claim, I believe you will agree that it is important that the courts not give undue weight to evidence of exposure in recorded form at the expense of other evidence of exposure. For example, in the Texas case which I cited earlier it appears that the court may have relied too heavily on the film badge analysis alone in denying the claim of a radiation worker while disregarding other estimated evidence of exposure.

In this case the decedent was engaged in handling, assembling and disassembling nuclear materials and weapons for approximately four years. For a two-year period while "handling" the materials, he was not issued a film badge or

or protective clothing. For the other two-year periods, he was issued film badges and protective clothing. Badge analysis revealed exposure on two occasions as 36 millirems, although this was determined to be only a fraction of his total exposure since the badge was being worn under protective clothing. The amount of exposure was not known but estimated to be greatly in excess of 36 mr. In addition, evidence showed that petitioner was on one occasion in an "incident" area but the proximity of petitioner to the location of the "incident" or number of rems to which he was, in fact, subjected was not known. The protective badge worn by a fellow worker also in the "incident" area showed 6,500 millirems of radiation. Evidence further showed that, for two years petitioner was exposed to "radiation leaks" from material handled, but the amount of radiation to which he was subjected was not known since he was issued no measuring device.

In addressing itself to the petitioner's contention that "the whole evidence" of this case did create a reasonable medical probability, the court agreed that reasonable medical probability can be based upon "the whole evidence." However, the court could not agree that such evidence was before them inasmuch as the extent of any radiation beyond the relatively safe dosage of 30 mr was unknown.<sup>10</sup>

In another case it was the absence of recorded evidence which appears to have influenced the U. S. Veterans Administration in denying compensation to a veteran who was assigned as an X-ray technician from 1953 to 1954 and developed acute lymphocytic leukemia in 1969. He wore no film badge and there were no records of his work environment. In spite of health physics testimony that there was a 50% to 80% chance that occupational exposure caused his death, the Board pointed out that the evidence of record did not indicate that the veteran received "excessive radiation" during service.<sup>11</sup>

In a 1961 Federal Workmen's Compensation Appeals Board Decision, the Board stated that because of the absence of a film badge during one period of the claimant's exposure, it was impossible to determine whether there was a significant exposure to radiation. Compensation denied.<sup>12</sup>

#### Total Evidence

From the point of view of the lawyer, the radiation record can never be too extensive. Accordingly, he will want to know of his client's total environmental exposure--occupational and non-occupational.

In a radiation claim a statistical game of possibilities and probabilities can, and in many cases does, greatly influence the result in a workmen's compensation case and, accordingly, the availability, accuracy and adequacy of exposure data, including prior medical as well as industrial exposure, takes on great legal significance.<sup>13</sup>

By way of example, take the case of a young man who developed acute leukemia after an exposure of a little more than 5 rem during a four-month period. A physician, knowledgeable in the effects of radiation, became acquainted with the case and noted that the man had received an indeterminate but apparently large amount of therapeutic radiation as a child. Keeping this in mind, and the fact that there is usually no decrease in the potential to induce leukemia by a long interval from the time of a first dose to the time of a second additional dose, the doctor felt that the worker's leukemia, if not caused, was at least aggravated and precipitated by his low occupational exposure. Compensation was granted.<sup>14</sup>

### The Aggravation or Acceleration Theory

The concept of aggravation appears to offer an alternative for the medical expert who seeks to establish medical probability in the face of low occupational exposures. In one case, a veteran who had been involved in nuclear testing while in the service was then employed teaching radiological safety as a civilian. He claimed that his leukemia was a result of exposure received during six years of civilian employment. Service connected exposures were unrecorded and civilian exposures were low. However, it was noted that there were certain instances of exposures which could well have been "over permissible limits." A radiologist found that the final monitored exposure could well have been an aggravating factor and, although the degree of claimant's exposures were conjectural, the leukemia could be considered the probable result of his occupational exposure. Compensation was granted.<sup>15</sup>

In another case, the claimant had a history of working in a microwave environment. After a latent period of many years he worked intimately with a weak ionizing radiation source. A board certified radiologist found that the low exposure to ionizing radiation resulted in a reactivation and/or acceleration of a dormant cataract and pointed out that without prior sensitization of the lense by exposure to microwave radiation, the radiation from the electron microscope would not have adversely affected the claimant. Compensation was granted.<sup>16</sup>

In yet another case, claimant was employed as a medical radiology and X-ray technician for approximately eight years. He developed leukopenia. Evidence showed that radiation protection practices were good and records showed exposures were low. The Bureau's Medical Director supported a causal relationship by aggravation from chloromycetin, a potent antibiotic with a known side effect of bone marrow depression.<sup>17</sup>

In a 1971 decision a civilian X-ray technician was granted compensation for chronic myelogenous leukemia. Evidence showed he had been exposed to low cumulative exposures for 20 years in the course of employment; that radiation protection was good; that he had service-connected exposure for a period of one year at the age of 18; that during three months of training while in the service he was constantly exposed to X-ray without benefit of safety equipment or protective measures to avoid exposure. The medical opinion indicated that there was aggravation of previous pathology. The claim was allowed for leukemia due to radiation exposure.<sup>18</sup>

Occasionally an award is made even when occupational exposure is low and with no need to resort to the theory of aggravation. For example, in one case a medical radiology technician employed in that capacity from 1957 until 1961 was isolated from further ionizing radiation in 1961 as the result of blood tests and the industrial medical officer's opinion that claimant had apparently reached his "personal level of tolerance." He developed leukopenia in 1966. Radiation records revealed no excessive exposure on film badge and personal pocket dosimeter. Work was performed using the accepted precautions of lead screens and aprons. It was established that claimant used reasonable care and had not been exposed to the direct X-ray beam at any time. However a radiology specialist attributed claimant's blood disorder to "incidental radiation effects." Compensation granted.<sup>19</sup>

In another case a 36 year old physicist at a radiation laboratory developed cataracts in both eyes. In his work around accelerators from 1950 until 1962 film badge exposure showed only 0.61 R. An ophthalmologist testified that claimant had radiation cataracts. Another doctor stated that claimant's cataracts were of the location and appearance associated with radiation cataracts; that while these cataracts can occur without radiation and while

claimant's record of exposure was very low, in view of claimant's work and age group the situation was "highly suggestive." Compensation granted.<sup>20</sup>

#### Radiation Protection Standards

The health physics profession readily admits that there is no such thing as known radiation safety; by that I mean some level of radiation exposure below which there is no biological effect whatever.<sup>21</sup> In short, frank admission is made that total protection against harm from man-made radiation would require a health standard of zero exposure; that radiation protection standards are not merely technical, that they are established through a balancing of risk versus economic and social benefit. Safety standards do not take into account the physical difference among individuals. Even though for safety guide purposes use is made of a "standard man" concept to determine the mass and effective radius of the critical organs of the body, when it comes to an individual radiation claim, the claimant's dose-response can hardly be considered standard. Yet a review of the cases shows that in a substantial number of claims permissible levels of exposure are used as indices of safety when deciding the issue of causation.

For example, a radiologist noted that the claimant's exposures "were in fact considerably in excess of the maximum permissible dose." He concluded the claimant's exposure probably caused his death from lymphosarcoma. Compensation granted.<sup>22</sup>

In still another case the medical expert noted that there was no contamination of the claimant's working environment "above permissible limits." Compensation denied.<sup>23</sup>

In another case a health physicist testified that the premises where claimant worked were found to meet the standards of radiation protection as recommended by the National Committee on Radiation Protection. The health physicist also testified that the film badge reports indicated that all exposures were "well below the maximum permissible dose." Compensation denied.<sup>24</sup>

In all of the claims referred to available film badge and other radiation records relating to claimant's exposure were introduced into evidence. Health physicists referred to records of exposure and related them to protection standards. What was the purpose of such testimony if not to imply safety or lack thereof? It has been said that there is a general tendency among laymen to assume that any exposure in excess of the various permissible levels and standards for any period whatever can be equated with proof of medical causation,<sup>25</sup> but since protection standards were never intended as indicators of absolute safety their use in the courtroom should be carefully scrutinized.

#### Alternative Proposals

At this point I believe that the problems inherent in our present legal system, as it is applied to low-level radiation claims, are abundantly clear. However, it is still the majority view of the legal establishment that the established principles of common law torts should continue to be employed in cases of delayed injury from radiation exposure.

Is there another route?

Professor Samuel B. Estep of the University of Michigan Law School has, over the years, suggested a somewhat novel approach to the problem. He would award compensation simply for the increased susceptibility to possible future disease. The uncontrollable factors which limit the accuracy of biological measurement by physical dosimeter readings seem essentially the basis for Estep to suggest

establishment of a "Contingent Liability Fund"<sup>26</sup> which would provide benefits to a radiation injured claimant regardless of his failure to show a causal relationship between the exposure and the injury. The proposed fund would consist of contributions by both the employee and the employer, the respective contributions to reflect both the "spontaneous" risk of leukemia and that due to the occupational exposure. In the event that the employee does develop leukemia, he is awarded a fixed amount of compensation without the necessity of adjudicating the causal relationship to occupational exposure. "Not only would such a scheme avoid the necessity for arbitrary adjudication, it would also avoid the expensive costs of administration. This would be of benefit to the worker, the employer and society as a whole."<sup>27</sup>

The Estep approach is somewhat akin to the concept of national health insurance. There are those who point out that in Great Britain no man, woman or child need for any reason fall below a minimum standard of life. By a combination of insurance schemes, a worker who comes down with a disease, occupational or otherwise, is assured of full medical treatment and weekly benefits during the course of his illness.<sup>28</sup>

Some ten years ago, Dr. Herman Somers in testifying before a committee of the United States Congress stated that:

"The evidence has been mounting for some time that the problems rising out of the scientific and technological revolution of our day are of a character which may not be capable of resolution within the traditional workmen's compensation design. The central question which we must ultimately face is whether or not, in the second half of the Twentieth Century, it will remain feasible, let alone justifiable, to operate a social insurance program on the old premise that a reasonably clear demarcation can be made between occupational and non-occupational disability."<sup>29</sup>

Lastly, for those who reject the insurance approach, a statutory prima facie presumption in favor of the claimant has been suggested. The burden of proof would then be upon the employer to show that radiation exposure was not the cause of the claimant's disease. It is my guess that the employer may have just as much difficulty in proving no causal connection as the plaintiff now has in proving causal nexus. New York has adopted such a law. In a recent New York case<sup>30</sup> the employee, a theoretical physicist, died from acute myeloblastic leukemias. In affirming an award the Court said:

"The record discloses that decedent was exposed to radiation for a substantial part of two periods and also at other times in various amounts. The testimony of the medical experts is emphatic that there is really no 'threshold' or 'safe' dosage of radiation because at the present stage of scientific knowledge it cannot be ascertained exactly what effects radiation has on the human body. It is also admitted that each individual reacts differently to exposure to radiation. The award is supported by substantial evidence and by the presumptions [N.Y. Workmen's Compensation Law §§ 3(2),47] . . . especially so in view of decedent's good health prior to his employment."

Another example of the presumption concept can be found in the Federal Coal Mine Health and Safety Act of 1969<sup>31</sup> which provides for certain presumptions in favor of the claimant in pneumoconiosis claims where it is found that the miner was employed for ten years or more in underground coal mines.

Congress has in the past considered legislation which would have instituted a Federal workmen's compensation program for employees exposed in their employment to "radioactive materials." The Price-Zelenko bill<sup>32</sup> would have established a presumption of causation in favor of any employee who (a) received an exposure in excess of the limits set by a Federal agency and (b) developed any ordinary disease which the United States Public Health Service certified can be induced by exposure to radioactive material.

During hearings on the bill there was strong opposition and rightly so to the proposal because a presumption of medical injury would be based on some arbitrary maximum permissible dose limit.

However, it has been stated that a statutory prima facie presumption in favor of the claimant would not shift significantly the percentage of cases in which the claimant would be upheld.<sup>33</sup> The solution would be for the Courts to apply the laws of negligence, of product liability and of workmen's compensation in the growing field of radiation hazards in a manner which supports preference for the plaintiff when causal relationship, though not clearly established, is clearly possible. The cases involving low exposures are relatively few. If, as has been said, ionizing radiation "is the most studied, best understood and most wisely used agent,"<sup>34</sup> the cases will continue to be few. Thus compensating a few individuals who have been exposed to levels of radiation which may have "possibly" caused their disease will not establish radiation as a hazard worse than it is at the present time. If, in fact, the hazard is miniscule, it will remain miniscule except for the injured worker.

Those of you who have followed the course of this paper have reason to wonder as to the proper solution for the handling of injury claims involving low level exposures to radiation. There is no easy answer to this question. When a court of law is attempting to determine the cause of a claimant's pathological condition in a workmen's compensation case, the court is faced not only with the question of scientific etiology but with a policy problem as well; namely, whether under all the circumstances it is fair to shift the economic consequences of the pathological condition from the claimant to the employer. Some of the techniques I have described today would do this very thing, but until more research is done and we better understand biological response to radiation, a great deal of inter-disciplinary concern and effort must go into solving the problem of the worker who allegedly suffers disease and death from low exposures. IRPA, with so many qualified persons from all over the world professionally engaged and actively interested in radiation protection, can contribute significantly toward a solution of this problem.

#### References

1. O'Toole, Dean Thomas J., Medical Radiation Information for Litigation, Proceedings of a Conference at Baylor University College of Medicine, Houston, Texas, Pub. 1969, U.S. Department of Health, Education and Welfare, p. 259.
2. Id.
3. Somers, Dr. Herman M., Chairman and Professor of Political Science, Haverford College, Haverford, Pa. Hazards and Workmen's Compensation, Hearings Before the Subcommittee on Research and Development, Joint Committee on Atomic Energy, March, 1959, p. 514.
4. Employers Mutual Liability Insurance Company of Wisconsin v. Parker, 440 S.W. 2d 43 (1969).

5. Forgotson, Edward, M.D., LLB, University of Michigan, "The Significance of Health Physics Testimony in Radiation Injury Litigation," Health Physics, Vol. 9, Pergammon Press, 1963, pp. 741-2.
6. Id.
7. Studies in Workmen's Compensation and Radiation Injury, Vol. V, U.S. Atomic Energy Commission, 1969, Case No. 9, p. 30.
8. Id., Case No. 4, p. 20.
9. Id., Vol. VI, 1972, Case No. 1, p. 16.
10. Supra footnote 4.
11. Supra footnote 9.
12. In re Baker, Decisions of Employees' Compensation Appeals Board, Vol. 12, 1961, p. 368.
13. Tinsley, Thomas A., Director, Bureau of Employees' Compensation, U.S. Department of Labor, Medical Radiation Information for Litigation, Proceedings of a Conference at Baylor University College of Medicine, Houston, Texas, Pub. 1969, U.S. Department of Health, Education and Welfare, p. 192.
14. Id., Case "B", p. 198.
15. Supra footnote 7, Case No. 3, pp. 16-17.
16. Id., Case No. 24, p. 62.
17. Workmen's Compensation Claim No. A15-10573, Office of Federal Employees' Compensation, U.S. Department of Labor.
18. Workmen's Compensation Claim No. ODPE4544, Ohio Bureau of Employees' Compensation.
19. Supra footnote 7, Case No. 8, pp. 28-9.
20. Supra footnote 9, Case No. 50, pp. 161-2.
21. Taylor, Dr. Lauriston, "Standards for Radiation Protection," Health Physics Society Newsletter, March 1973, p. 13.
22. Supra footnote 7, Case No. 2, p. 15.
23. Id., Case No. 6, p. 25.
24. Id., Case No. 9, p. 30.
25. Supra footnote 5, p. 742.
26. Estep, Samuel D., "Radiation Injuries and Statistics," Michigan Law Review, 1960, pp. 259, 281-398.
27. Sagan, Leonard A., M.D., "Radiological Problems Associated with Adjudication of Workmen's Compensation Claims," Journal of Occupational Medicine, Vol. II, No. 6, June, 1969, p. 339.



28. Social Security in Britain, Central Office of Information Reference Pamphlet 90, Her Majesty's Stationary Office, London, 1970.
29. Supra footnote 3, p. 513.
30. Besner v. Walter Kidde Laboratories, 24 AD 2d 1045, 237 N.Y.S. 2d 585 (1963), 265 N.Y.S. 2d 312 (1965).
31. Public Law 91-173; 83 Stat. 742.
32. H.R. 1267 and H.R. 2731, 1961.
33. O'Toole, Dean Thomas J., "The Incident, Nature and Adjudication of Workmen's Compensation Claims Involving Radiation Exposure and Delayed Injury," Vol. II, Studies in Workmen's Compensation and Delayed Injury, The Department of Labor and The Atomic Energy Commission, 1965.
34. Supra footnote 21.

RADIATION PROTECTION INFORMATION AND EDUCATION FOR THE PUBLIC :  
MISTAKES AND LESSONS , STRATEGIES AND TACTICS.

H. Brunner, dipl.Phys.ETH  
Secretary, Fachverband für Strahlenschutz  
Assistant head, Health Physics Division  
E I R, Eng. Institut für Reaktorforschung  
CH-5303 Würenlingen, Switzerland

1. Summary

Radiation protection and nuclear safety have become major targets for attacks by critics and opponents of many sorts. We must try to improve our methods for coping with such challenges and avoid to waste too much of our time or forces. By learning from past mistakes and analysis of the situation appropriate tactics and strategies can be developed that take care of the immediate needs and set some long-range goals for improved information and education of the public. Characteristics and problems of using or cooperating with news media are discussed and some proposals for actions by IRPA and its affiliated societies are presented.

The following ideas and proposals are the personal views of the author.

2. Problems and types of confrontations with the public

Within the last few years radiation protection and nuclear safety have suddenly become major targets for attacks by critics and opponents of many sorts. The types and motives of such attacks have been discussed a great deal and shall not be repeated here. We all agree that the reproaches are not justified, neither absolutely nor in perspective to what happens in other protection and safety areas. But there exists an obvious information gap in the public, and at least part of the blame for this falls on us. Complaining does not liberate us from these problems. The defense against the uninterrupted, often exaggerated or even stupid, but nevertheless effective attacks consumes more and more valuable time of authorities and experts without apparently leading to a quick success or visible end of such debates. The fact, that even government agencies or parliamentary committees are not ashamed of inviting self-proclaimed so-called "independent experts" and professional opponents on the same conditions as the real qualified experts shows how far the confusion about who is right or wrong has gone already. One is sometimes reminded of two sinister manifestations of the Middle Age, the Inquisition and the Crusades, which both were due to the agitation of fanatics appealing to idealistic motives and fear, and which resulted in terror, retardation of progress, defeat and failures. Some countries had to bear the consequences for centuries. Frustration, anger, resignation, exhaustion of forces and neglect of main and long term tasks may be the undesirable results for our profession if we do not try to improve our methods of coping with such challenges or if we waste all our forces for short-lived emergency actions at the expense of a well planned long-range program. We must learn from past mistakes, analyze the situation and develop appropriate tactics and plans.

We have to deal with various characters, types and forms of confrontations, communications and contacts between experts and the public. The character of a

confrontation can be voluntary and offensive, when we have taken the initiative, set the goals and selected form, place and time, or it can be involuntary and defensive, when somebody else has invited, challenged or attacked us on his own terms.

We can assume the following coarse types of contact :

- A) Public performances: information, discussion or contradictory meetings; hearings; panel discussions; press conferences (sometimes with radio and TV coverage); seminars and symposia etc.
- B) Mass media : press; radio; television;
- C) Publications: books; specialized periodicals; laws, regulations, standards ; in the future audiovisual items such as TV cassettes;
- D) Educational programs

Some aspects of each type with regard to our problems are summarized below, and some of them are discussed in more detail in Appendix B.

A) Public performances have more frequently an involuntary character, being organized by local political parties, societies, opponent groups, utilities or authorities. The audience is restricted to some hundred persons at most (radio/TV coverage excluded) and it may often be selected or biased. The time available is always too short, be it for lectures and statements or for discussion. The atmosphere is often unfavorable, unfriendly or outright unfair. Success or failure are very much dependent on the chairman or moderator and on the show talents of the active participants. Technical troubles with microphones and other inadequacies create additional problems. Meetings are often invaded by outside organized opponent groups which try to monopolize the performance. Biased reports by mass media may present a completely distorted picture of the event to a much larger audience. Such public performances have mostly only short-lived effects and are rather negative and unsatisfactory to anybody really concerned apart from "showstars" such as politicians or professional opponents. The level of the discussions is either primitive or far above the comprehension of the audience. To sum it up : many public events are simply alibi-functions or much fuss about nothing. We should try to avoid them or restrict them to the voluntary variety.

B) Mass media have their own rules and characteristics that differ locally and nationally and that have to be known precisely if any efficiency is attempted. To some extent also many politicians and other public opinion-makers have to be handled similarly. Mass media are primarily interested in "news", i.e. new informations, sensations, stories, scandals etc. not yet published or diffused by other mass media. The main quality of such news must be to make headlines, to increase the circulation or audience. With some forms of mass media almost no limits of truth, morale or respect exist, only a sharp drop in circulation or rating can stop them. "Bad news is good news" for news media and much more interesting than dry facts. To be first to publish a sensation or information is much more important than to inquire or check the validity before publication. The correction of wrong informations is usually left to those involved or concerned, but does only rarely get the same amount of publicity or prominent place as the original uncorrect feature. The public or audience is large and of more or less unknown composition. Usually the confrontation will have an involuntary character. We have to expect biased, exaggerated or distorted presentations, even some sort of "censorship" by publishers, editors or producers with the easy excuse of lack of space or time. There is usually no long-lasting interest in a special topic with the exception of some outstanding newspapers or periodicals that have specialized editors or writers, or of some "engaged" publications or reporters. Most newsmen have to deal with a large variety of topics without having time or interest to acquire a deeper knowledge. Few discussions or direct contacts with the public are possible; only a small percentage of letters to the editor are published, and no news medium likes to admit a mistake. With some notable exceptions the affects of news media

presentations last only a few days. Voluntary, active cooperation with news media is only possible if good personal relations to editors or producers can be established. The "target" public or audience is quite different for each medium, newspaper or program feature, and a "market and media research" similar to the one done by advertising agencies is necessary for optimal efficiency.

C) Publications is used here as a collective term for all kinds of printed or otherwise duplicated material that are available and distributed to the public over a rather long or even unlimited period (e.g. through libraries). They can be used and reused at any time by laymen or experts, individuals and groups, in one or several countries at any not predetermined times. They may be copied, referred to, cited, summarized, often also discussed in mass media or special periodicals. A great advantage is ample space for detailed treatment of a topic. The size of the public is unknown. There is no direct mutual contact between author and readers and no direct, immediate discussion. Misuse is possible without chances for clarifications or corrections. But many characteristics and rules are similar to those of mass media, and equivalent precautions are necessary for good results.

D) Educational programs of all types and on all levels are probably the most efficient contacts with long-lasting effects, extended and repeated interactions between specialists and the public. But good programs need detailed, time-consuming preparations, and their efficiency depends to a great deal on the pedagogic qualities of the lecturers or teachers. Such programs allow sufficient time for a thorough presentation of the material, explanations, repetitions and discussions, but also for the creation of an atmosphere of confidence between author and public which facilitates the implantation and acceptance of the information. The audience will be small, except for some basic courses or radio/TV educational series, but if regular teachers are selected as the first targets and the topic can be incorporated in various curricula and professional training programs, the repetition over many years will increase the size of the public and slowly build up a useful and solid foundation of basic knowledge.

The present situation challenges us mostly with involuntary confrontations of the types A and B. In order to improve our position and win back the initiative we need a long-range program for the conquest of types C and D which will produce a feedback on the mass media and the general public. Public performances should be reduced to the indispensable minimum, i.e. voluntary events such as press conferences, information meetings and symposia in a quiet, fair and matter-of-fact atmosphere with an audience that knows at least some basic facts. But most of our present activities are fire-fighting emergency actions due to unexpected attacks. We struggle to hold our positions, and many defensive counterattacks suffer from a lack of time and preparation and may make things even worse.

### 3. Some typical mistakes

As the mistakes during public confrontations in the nuclear field have been discussed frequently, I shall only describe two typical mistakes or "syndromes" which are causing many failures but seem to be difficult to root out.

#### 3.1. The "prestige syndrome" or the "Peter Principle" of prominent speakers.

A favorite trick to attract the public is to feature well known personalities as principal attraction or "decoration". Apart from the fact that a Nobel Prize is no a-priori qualification for omniscience, infallibility or even just competence in a field different from the one for which the prize was awarded, many of the prominent people are also victims of the manager syndrome or the "Peter Principle". If they only have to deliver a well specified "show" such as an opening speech or an invited lecture this may work perfectly well. But there are only few such prominent persons who have time to keep so well informed on all details and latest developments that they are able to survive a battle of public discussions with a well prepared team of opponents. Fortunately there exist excellent active scien-

tists and specialists who also are fine speakers with a flair for good presentation, didactics and effective discussions. You may be congratulated if you can get some of these for a meeting. But very often you will have to select between one of two types of prominent participants. One are the top managers of government agencies, research institutions etc. who by profession have to defend certain official positions. If they have enough routine with confrontations and get a thorough briefing by a good staff, things may go well. Otherwise they can sometimes be manoeuvred into a trap and may have to improvise answers to unexpected questions. Such answers often reach their "level of incompetence" and provide excellent weapons for opponents.

The other variety of prominent people could be compared to showstars (in his latest novel Koestler names them even "callgirls"). They seek publicity, like to be in the spotlight, want to see themselves in news media, and ride, for personal satisfaction or opportunism, on any popular "bandwagon". They are smart and often have a previous solid foundation of scientific or other achievements, they are good, witty speakers, and they are willing to deliver a talk on any favorite subject such as environmental pollution, world models, futurology or birth control, provided they get well paid and publicized. They have no scruples to change the bandwagon as long as they can ride on top. But they keep mostly to the negative side in some top level protest movement, for it is much easier, less time-consuming and more colorful to criticize, to ride sharp attacks, to appeal to unconscious feelings, fears and antagonism in the public than to study a problem in detail and to try to give a balanced but much less dramatic picture, or to offer constructive critique and useful proposals that are more than fancy, futuristic but entirely unrealistic dreams.

3.2. The "babylonian or ivory tower syndrome" is well known and only too frequent in public performances of all kinds. Many speakers or panelists do not care about the intellectual level or basic knowledges of their audience. They feel obliged to prove their competence by using a disguise of specialized or sophisticated language largely incomprehensible to the layman. They embark on long monologues that drift away from the problem, or they complicate it by details, boundary conditions and reservations, that nobody anymore can understand whether the answer is yes or no. All the public does understand is that they do not understand anything, and that the experts do not know everything and seem to disagree quite a lot. No wonder that the public has less and less confidence in experts. If somebody really knows a subject he should be able to talk about it in clear simple terms and make the essential meaning of it comprehensible even to a layman. But beware of the other extreme: you may turn an audience against you also if you misjudge their level to the low side and use a baby language for an intellectual public.

#### 4. How to improve our methods

Our forces and means are limited, much more than those of many "opponents" (this term will be used for the various partners or enemies in confrontations). We need better methods and systematic planning if we want to improve our situation. It can be compared to a small army fighting a much stronger one, and part of the solution to some of our problems lies in the application of old proven military practices in planning, decision-making and tactics. These methods have in recent years been rediscovered and "re-invented" as "modern" management techniques under fancy names, but to anybody who knows military staff methods they are old familiar practice. To summarize: we must analyze past experience, recognize the typical and main mistakes, learn from other, similar situations, draw the necessary lessons, build up a good information network and start a thorough and systematic planning of a well defined program.

It is a fundamental rule for military decision-making that before you can decide about the appropriate action, you have to analyze and evaluate the situation and its possible developments by considering the following factors :

- the task which you have to carry out or the goal that you have to reach. It must be well understood and clearly defined. Vague orders end often with failure.
- means, resources, manpower, allies that you can count on for your task;
- the "environment", "background" or "climate" in which the operations must be executed (time, place, duration, form, political, economical and psychological factors and influences etc.);
- Who is your enemy or opponent ? This includes real opponents as well as discussion partners or the audience. What is his position ? What are his motives ? What means and capabilities does he have ? What may be his plans and goals ? What support may he get from the audience ?
- What is your "firepower" ? What "weapons" are at your disposal or applicable, i.e. which arguments, documents, informations, proofs, experimental results, visual aids etc. are available for the support of your cause ? How effective are they against the probable targets ?

Once we have precisely defined the task or goal and impartially analyzed the situation, we must consider possible solutions or actions. These must be evaluated for efficiency, advantages and drawbacks, chances for success and possible reactions of the opponents. Only when we have gone through these mental exercises and can support them by some "experimental" evidence or "reconnaissance" are we ready to decide in principle how to act. Once the decision has been made, the tedious work of elaborating detailed action plans and preparing and testing all manpower, means and resources follows before we can start the action.

For our present defensive and involuntary confrontations part of that process will forcibly have to be shortened or improvised due to lack of time and freedom of action. But it would be inexcusable to omit it altogether even in "involuntary battles", and it would be a crime to start a voluntary action before all these mental and material preparations have been finished and tested. In the Appendices A and B a collection of hints and ideas for many possible situations is given.

We should also look for examples and models from other domains in everyday and public life with similar problems, where solutions have been found and tested and where we can learn a lot. Let me just name a few of them without getting into details: advertizing agencies, public relations in industry or government (Atomic Forum organizations, Technical Information Division etc.), professional societies such as ANS, IEEE or medical associations, accident prevention, traffic safety, environmental protection, industrial hygiene, civil defense and army, sports, churches, charity programs, political campaigns, educational programs and methods, mass media, etc.

## 5. A general action program for public information

### 5.1. Short-range emergency program :

No doubt there is an immediate need for an emergency program in order to hold the positions and survive in the flood of opponent charges and concurrent environmental protection fashions that, though late come, now try to monopolize the field and preach the only saving faith. The details of such "crash-programs" depend very much on local circumstances and resources, but some common problems can be seen.

Part of our efforts should aim at bringing the discussion back to solid ground to a fair, matter-of-fact exchange of rational and objective arguments and facts. This may in some extreme cases require a strong, well aimed action against some fanatic and unfair opponents in order to uncover, expose and isolate them. Even when we get attacked we must make a clear distinction between honest and knowledgeable opponents who merit our consideration, and unfair fanatics without sufficient qualifications whom we must openly declare not to accept as discussion partners on an equal basis.

A second goal must be to win back the initiative from opponents and opportunists. News media, public, politicians and authorities should get the basic and any new informations and facts first and without delay from us, from the specialists, not in a distorted or delayed way from news agencies, scandal reporters or biased opponent bulletins. We must inform quickly, openly and correctly, whether the information is favorable or not (incidents etc.), and we must establish direct information channels to all concerned. All this is of course easier if we have managed to remain neutral in the nuclear dispute and can keep above economical or ideological biases. A fine example for such correct information is the recent book by Lindell and Löfveberg on "Nuclear Power, Man and Safety" which unfortunately up to now only exists in its original Swedish version.

The information handling problems play a key role here as everywhere in modern life. The flood of correct or wrong informations, arguments, statements, reports in the public discussion is even larger than in the scientific and technical areas of radiation protection and safety, and it is often much more difficult to track down the original source and form of an information or to keep up-to-date with the latest developments and publications. This problem cannot be solved by individuals but needs a well organized cooperation on national and international levels. Below I shall offer some ideas on how our societies and IRPA might help.

### 5.2. Long-range program

Our long-range tasks have mostly educational aspects. We must familiarize the public, the news media people, the specialists of related fields, officials and politicians with the basic facts on radiations, their effects, protection, regulations, safety of nuclear technology and its applications, and relations to other risks in modern life. Radiation must become as familiar to them as space flight or stereophonic music.

This requires educational activities on all levels and for all ages from high school to professional or academic training. The best approach is to get first the teachers on all those levels interested and trained in the subjects, then incorporate the topics in the curricula of future teachers and instructors for public schools, higher education or professional training. The last step would be the integration into the curricula of the various educational programs. Besides regular school programs we must not neglect post-graduate programs, on-the-job training in industry, adult education programs of universities or television networks, evening courses etc. In some countries basic military or civil defense training may offer additional opportunities to inform large parts of the population. If it makes things easier, our subjects can be incorporated into larger ones such as hygiene or environmental protection as suitable carriers of more general appeal or interest.

### 6. Some suggestions for action by IRPA and its affiliated societies

Most of the work in the information field has to be done on the national level, it is therefore primarily a challenge for our societies. A few examples from the activities of the Fachverband für Strahlenschutz show some possibilities. One of our main goals is to keep the members in close contact with the society and each other and to give them as much information as possible. A news bulletin of about ten pages is mailed to them four times a year and contains all information on our society, its working groups, coming events, interesting news, publications etc. Each year's general assembly is combined with a symposium, the proceedings of which are given to all members free of charge. The exchange of informations and experiences and the cooperation are further supported by a number of informal working groups, started by the initiatives of interested members for the discussion of problems from a certain special area such as incorporation analysis, working place monitoring etc., or, to mention two groups of special interest for the topics of this paper, on education and on public relations problems. The results

of the sessions of these groups are reported in the bulletin. When a problem has been treated, a short report, review or recommendation is issued, published and distributed to all members and, as the bulletins and proceedings, to representatives of societies, government agencies etc. On the local level the few health physicists on place must bear the whole burden of information, and it is vital for them, the society and our profession to keep all members well informed and up-to-date. If we cannot manage the information and education problems and needs of our members we have no chance at all to handle the public information problems.

How could IRPA and its societies help us? When I say "us" I mean all individual health physicists wherever they are in any way active in public information or education. We urgently need an excellent, fast and reliable information network on an international basis. Many symptoms and "viruses" of the present radiation and safety "syndromes" originate from other countries, often the USA, and infect other countries very quickly, because some opponent organizations have a good and fast information network with wide distribution. If we cannot build up something at least equivalent, we will loose the race. Certainly, in the nuclear field the Atomic Industrial Forum and its equivalents in other countries, or the American Nuclear Society, have done a fine job, but they cover only part of our field and not always in the necessary details, apart from a certain bias towards the promotion of nuclear energy. As health physics also extends into medicine, environmental protection, industrial hygiene and other areas, we need our own system which covers all these fields. To subscribe to the information services of all the other societies and organizations is far above the financial means of a health physicist, and our only international link, the "Health Physics Journal", is far too slow and too expensive to fulfill this information task adequately. One reason why many societies almost never supply informations to the "news" section of the HPJ is the delay of several months before publication which often makes the information outdated. What we would need is an "IRPA Newsletter" similar to the fine examples of the newsletter of the Health Physics Society or the new "Radiological Protection Bulletin" issued by the British National Radiological Protection Board. This should be produced by cheap means and distributed, through the societies, to all members every 2-3 months. It should contain reviews of all new informations and developments, coming events, recommendations, new reports and publications etc. In order to produce such a newsletter, IRPA would need an Information Center with a small permanent staff and offset printing facilities. This IRPA Information Center should be in close continuous contact with all IRPA societies, their working groups and members and through those with national authorities, committees etc. Through the Executive Council of IRPA and directly close connections and information exchanges should be installed to such international bodies as ICRP, ICRU, ISO, IEC etc. and to organizations such as IAEA, NEA, WHO and others. This IRPA Information Center could become a sort of clearinghouse for information on all aspects of radiation protection and even develop into an international central information service which not only publishes newsletters, reviews, handbooks etc., but also could handle individual requests for special informations. My proposal does not aim at multiplying the flood of informations, but rather at forcing it back into one or a few reliable and fast channels, reaching all of us with a minimum of delay and supplying all necessary informations in a compact form but without gaps. Of course such a project would need active cooperation and support by the societies and probably higher financial contributions to IRPA. But it could create a very useful, internationally acknowledged function of IRPA and free it from the false image of being an organization which only sponsors some congresses. It would make IRPA as useful and renowned as ICRP or certain international organizations, but without the political drawbacks and restrictions of the latter and with a more practical note than the former.



## 7. Some implications for our profession

I believe it is an obligation for all health physicists to be or become active in the education and information of the public. This has also some consequences and feedbacks for our profession. Health physics must remain and become even more a respected, well known and well based profession of high standards with well trained and informed personnel on all levels. This can only be done if we support our societies and IRPA and actively take part in their programs, and if the societies learn to operate as efficiently as medical associations. We must not lose the initiative or the tasks to other, more active, professional societies or promotional organizations. Close and constructive cooperation with national and international organizations, authorities etc. is very important, among other reasons also for the adequate representation of practical viewpoints. Our position should be objective and as neutral as our respective jobs allow. Correct information and the truth must be disseminated. We must continue to discuss problems openly on an international basis as we have done up to now without being afraid of publicity, abuse by opponents or pressure from interested parties.

But a good deal of self-criticism is also necessary. We must constantly improve our professional standards, eliminate mistakes, bad practices and shortcomings. All health physics tasks in industry, medicine and research must be carried out by well trained and capable health physics personnel. We should never accept an unduly reduced or unsatisfactory health physics program with the argument that economical considerations are more important than adequate and reasonable safety standards and qualified personnel in sufficient numbers. If we do not fight such bad practices we shall soon lose face and credibility. Nuclear power, nuclear medicine and other applications of radiations should be promoted, but not on the costs of reduced protection.

\*\*\*

### Appendix A

#### Some ideas for actions

##### 1) "Rent-a-program"

Many professional societies, clubs and other organizations have a regular program of lectures, colloquia, information or refresher courses, excursions etc. The organizers of such programs are chronically short of topics and ideas. They welcome any reasonable proposal or offer. If our societies can offer ready "package programs" of speakers or excursions and have put those into operation a few times, a growing demand may be expected from many halls.

##### 2) "Do-it-yourself" or "autosuggestion"

People believe best what they have found out themselves. Students, laymen or specialists from other fields who are interested or even critical of protection or safety problems could be assembled in study or working groups and be given the task of studying a well defined problem. Our societies could sponsor such a study or at least support it actively by providing the necessary subtle guidance, assistance with literature and consultants and openly and critically discuss the findings with the groups. If the results are interesting, suitable publication should be arranged and coverage by newsmedia organized. Such an experiment has of course its risks and costs a lot of time, but it will help such a group to get a better insight into our problems, philosophies, working methods and the wealth of information available as well as the amount of work and experience necessary for judging problems.

### 3) "Group therapy"

We cannot educate or convince the public or its opinion-makers by large public meetings of the "Billy Graham" style. Producing mass hysteria of any kind never is a lasting therapy. The "single patient treatment" on the other hand costs too much time, money and manpower. But a suitable group therapy over a weekend or a working or vacation week may offer interesting possibilities for treating carefully selected groups of prominent people, opinion-makers, politicians, newsmen. Such people who get a lot of publicity cannot be converted, convinced or neutralized in a public event where they feel obliged to give the kind of "show" the public expects. The only chance to get across any barriers is to separate them from their audience and to offer them an attractive and pleasant opportunity for free, unrestricted discussion and useful information on an exciting topic in a family-type group of interesting people, without any official obligation, function or publicity. But do not mix newsmen and prominent people, for the former could not resist the temptation of reporting on the event, and the latter would feel obliged to continue their "show". A well prepared, even exclusive program frame is important, but sufficient time for informal personal contacts, discussions and brainstorming must be available. Do not preach or try to convert, be open, informative and matter-of-fact. Present your views and problems, but let also the guests give their view of the problems and suggestions for solutions. The most important goal is to win their confidence, to break down political or other barriers, to release tensions and to get rid of resentments. The effects will not be felt immediately, but such a program will bring positive results in the future to the profit of our long-range programs. This kind of treatment could also be called the "F-treatment" where F stands for fun, friendly, food etc, but also for "faed facts, fight fiction".

### 4) The "domino or bandwagon effect"

Newsmedia, politicians and many other prominent people like to jump on a bandwagon, i.e. to be on the forefront of any actual happening or event getting sufficient publicity. If we therefore can get some influential people or news media interested in our problems such that they give them a lot of publicity, there is a good chance that other mass media will cover this, too and try to follow such a trend in order to get their share of the profit. Careful selection of the primary target will produce an optimal amount of spin-off.

### 5) "Desensibilisation" or "vaccination"

The negative public reactions and fears are very much like allergic reactions or contagious diseases, and some treatment similar to desensibilisation or vaccination might help in these cases, too. We should try to feed repeated small doses of correct informations to the public in attractive forms. We might even imitate the "syndromes" by using "anti"-headlines as eye-catchers, but followed by the correct informations and facts. Look how advertising agencies launch a new product or idea and you will see what could be done.

\*

## Appendix B

### Some remarks, lessons, rules and tricks

#### Public performances :

- 1) Set a clearly defined goal for the performance.
- 2) Carefully select the chairman or moderator and the other active participants.
- 3) Choose well informed, witty, quick-minded and eloquent speakers. Avoid prominent but superficially informed or narciss-minded personalities except for well prepared harmless official functions such as opening ceremonies without public discussion. For the real battle respect the "Pater Principle" and select a well informed staff member rather than the top boss.

- 4) Prepare your defense and attacks. Locate the weak points of the opponent. Study and analyze his previous actions, publications and arguments and prepare your counterattacks. Keep up-to-date on the latest developments. Imagine what moves the opponents might try (each has his typical fashion). Look at your own arguments from the opponent's viewpoint, try to guess where he might find your own weak spots.
- 5) Analyze the tactics of left wing opposition groups, radical student movements and protest groups. There are many similarities in their methods to those of nuclear opponents.
- 6) A good training in dialectics and rhetoric is very useful.
- 7) Get your supporters into the audience. Let them ask those questions which the local audience should like to ask but does not dare to or does not get a chance to ask because organized opponents may try to monopolize the discussion. Do not forget the Trojan horse trick.
- 8) If you get selected for an involuntary type meeting, try to get the best possible and most complete informations on the following points :
  - Who are the organizers, what are their goals, background, connections ?
  - Who are the other active participants, what are their views on the topic, their backgrounds, interests and connections ?
  - What audience and what intellectual level can be expected ?
  - Local, political and psychological background and environment ?
  - Available time, technical resources etc. ?
  - Are there any plans or risks that the meeting might end with the "unanimous adoption" of a biased resolution ?
- 9) An informal contact (lunch) between the speakers and other active participants before the performance may release some tensions, clear up some misunderstandings, settle some problems or disputes beforehand under four eyes. But do not get caught or misled by nice manners and words, some people change and uncover unexpectedly when on stage and before an audience.
- 10) Say clearly "yes" or "no", call a thing either black or white, use pictures that are as simple as woodcuts. You never have enough time to get into details, so why trouble the audience with things they cannot understand ? Make your statements short, clear, impressive. They should hook like a good joke and be remembered. Be witty but do not exaggerate and do not imitate a clown or showmaster.
- 11) "Steal the show" by answering some likely arguments of your opponents before they get a chance to present them, let them blast open doors. Attack is the best defense. Avoid to be caught or attacked unexpectedly.
- 12) Keep calm and matter-of-fact. Select your weapons according to the type of battle and the weapons of the opponents. Make a difference between a public meeting with its catch-as-catch-can rules and the fair-play sunday-school atmosphere of a scientific meeting.
- 13) If you are well prepared you can often refute or disprove an opponent by his own words. Give exact citations and references.
- 14) Make it clear to the audience if and why you do not accept a certain opponent or "witness" as a real specialist or expert qualified to deal with the problem, despite all titles and other merits.
- 15) Reveal to the audience the motivation, background connections, interests and real goals of some opponents who often are presented under some harmless or impressive disguise.
- 16) Avoid controversial issues or arguments that are understood by specialists only. This would only nourish the impression that the specialists disagree or that many important things are not clear or known.
- 17) If you want to gain time for finding the best answer, either smoke a pipe and look like Rodin's "Thinker" for a while, or better : explain, analyze, qualify and simplify the question or problem for the audience. This makes it easier for you to find the right answer. Explain whether it is a fundamental, impor-

tant problem or just a secondary detail. If you do not know the answer or do not want to give it, state clearly why. Explain why the problem is too complicated for a short answer, or why somebody else will be better qualified to answer, or that the exact informations are not at hand but where they can be found, say whether an answer exists or not. Never leave the impression that you got caught in a trap or that an important lack of knowledge exists when this is not the case. A possible way to react may sometimes be to show first that the questioner did not understand the problem, but then the question must be answered anyway.

Mass media (and politicians):

- 18) Some general rules for using mass media :
  - Study the characteristics of each medium and of its individual representatives (e.g. various newspapers). Experiences from one region, country or representative do not necessarily apply to another one.
  - Analyze the previous position of the medium to the problem, its public or audience, the intellectual level, style etc.
  - Contact the responsible editor or producer. How much space or time is available. Which feature, program, page or section is best suited? What relation between text and pictures is desired?
  - for books : select the proper editor, series, size and price category.
  - Do a good media and market research such as done by advertising agencies.
  - Get either an optimal efficiency or skip it.
- 19) In dealing with news people (or politicians) it is very important to build up good personal contacts to some carefully selected key people. Do not try a "crash-program" of convincing them, but slowly develop a basis of mutual understanding and confidence. Ask them for help and advice in public relation problems. Help them with facts, informations, consulting, news, frequent press releases or conferences, some exclusive reports or interviews. A newsman does not like to be coached or tutored, but he will welcome support and help if he gets it easily and if it is useful to him for avoiding errors or blunders.
- 20) If news media publish incorrect or biased informations, only strong, multiple and quick individual, collective and official reactions and complaints on several levels, from the responsible editor to the top management, will cause a correction.
- 21) If you get interviewed, take care of your image, keep neutral, matter-of-fact and independent. Do not get seduced to play the expert in fields other than your own. Of course you may offer your personal opinion also on other problems, but as an educated layman. Distinguish clearly between your official standpoint due to your function and your personal views as a specialist or a layman.
- 22) Do not over-simplify. If an essential element is missing in an information, it may become incorrect, miss the target or even become a bait for attacks.
- 23) Separate clearly established facts from fiction, assumptions, extrapolations, prognostics etc.
- 24) Do not get upset if even a fair and objective reporter asks you a critical or uncomfortable question. It is part of their professional technique and a way to show their independence. Do not use the same yardstick of quality for mass media as for special or scientific publications or events.
- 25) make sure that your statements are published correctly. Request to see the proofs before publication or reserve a right to reply. Submit your statements in written form, make your own tape record or get a copy of the reporter's tape.
- 26) Do not get caught unexpectedly. Give no statements which you had not time to think about. If the problem really matters to the reporter, he will give you time for preparation and tell you what he intends to do with the information.

If a reporter is not willing to do so, something is suspicious anyway.

- 27) When you contribute an article to the press, keep your language simple, explain indispensable special terms. Give it an attractive form, write a popular style, supply good illustrations. Make short paragraphs and type with double space and wide margin.
- 28) Radio programs are not very suitable for complicated topics. Their main features are news and short informations, short comments, or panel discussions.

Television :

- 29) These remarks apply to European television without commercially influenced or sponsored programs or shows, which operate on principles similar to those of newspapers or magazines.
- 30) The main information carrier in TV is the picture, it must move, not stand still. Many producers and directors prefer visual esthetics, action and gags to all other considerations such as facts, balanced information and content, truth etc. The spoken word is subordinate to the visual development and must be short and clear.
- 31) A TV program is momentarily impressive by its dynamics, some highlights and gags, but it has no long-lasting effects.
- 32) A program needs an "eye-catcher" at the very beginning or in the title which attracts the audience and prevents them from switching channels. Best carriers are regular, highly rated and well attended programs and features. They must be carefully selected according to topic, moderator, background, audience, daytime, duration, style, actuality, concurrent programs on other channels.
- 33) The larger the audience the shorter is the time available for a special topic. The duration varies from a few minutes for actualities over a preferred average of 10 - 15 minutes to the rare cases where up to an hour or more can be devoted to a single topic, but usually only in special documentary or science programs or panels on topics of high interest. Sufficient time for treating a subject in detail would only exist in educational programs, but these require long and tedious preparations and are planned years ahead.
- 34) Popular quiz programs, panel or interview series might offer a chance to introduce a problem or topic to a large audience and get additional coverage by other news media such as newspapers or magazines. Very close cooperation with producers and moderators and careful selection and preparation would be required.
- 35) If you get interviewed by TV, try not to stick to a manuscript. Inquire beforehand in what context the interview will be shown.

\*\*\*\*\*

## PUBLIC CONFIDENCE IN NUCLEAR POWER

J.A. Bonnell  
G.C. Dale

Central Electricity Generating Board  
Courtenay House, 18 Warwick Lane,  
London EC4P 4EB, England.

### Abstract

Although, in the United Kingdom a healthy interest is maintained by the general public in the development of nuclear power as an energy source, concern has never been so widespread or as hostile as has been the case in the United States of America. This paper suggests reasons for this and discusses the origins of nuclear power in relation to the public and the government administrative machinery which has been built up around the subject. The method of obtaining consent to build and operate a nuclear power station is outlined and the intensive public relations exercise to which all parties contribute is put forward as a main reason why nuclear power is not only accepted by the majority but is actively encouraged.

### Background

In the United Kingdom there are only two electricity utilities which operate nuclear power stations. They are the South of Scotland Electricity Board and the Central Electricity Generating Board. Both are nationalised industries and the latter is responsible for the generation and bulk distribution of electricity in England and Wales. It owns and operates 174 Generating Stations with an output capacity of 56,000 megawatts composed of a mixture of coal fired, oil fired, hydro, gas turbine and nuclear power units. The nuclear component at present consists of 8 twin reactor stations of the gas cooled magnox type whilst a further 4 twin reactor stations of the advanced gas cooled type are currently under construction. Because of the way in which the electricity supply industry is organised the general public are in direct contact with the retail distribution organizations and do not come into contact with the CEGB except on relatively rare occasions such as new generating station projects or the siting of high voltage transmission lines. With such a large organisation under unified control as the CEGB it is able to support a headquarters staff in both the planning and nuclear health and safety fields. This has enabled a uniform policy to be agreed centrally and implemented effectively. A further advantage is that single authoritative channels of communication have been established between the Board and the several Governmental regulatory authorities which are concerned with the whole field of nuclear energy.

## History

The development of nuclear power in the United Kingdom started in a favourable climate in the early 1950s. Coal, the main energy source in the U.K. after the second world war was in short supply and increasing in price at a rate which was directly observable by the open coal fire burning public. Oil, then almost non-existent within the British Isles or its waters, was of foreign origin and subject to the uncertainties of foreign currency shortages and seizure of the wells mostly in the politically unsettled middle east. Gas was a rather expensive, and to many people a dangerous by-product of coal. The fanfare of publicity at the opening of the first nuclear power station at Calder Hall, dignified and given the seal of respectability by Her Majesty the Queen, did much to persuade the inhabitants of our cold and coal grimed cities that the millenium was at hand. The Windscale accident in retrospect, far from engendering hostility to the "atom", due mainly to the prompt and expert handling of the affair, did much to reassure possible antagonists that although accidents could happen, the Government and its traditional safety agencies were fully on top of the situation and no great catastrophe was likely in any way as devastating as the air raids so relatively recently endured. It is important to realize that the U.K. is so small and its communication and legislative structure so unified that attitudes and opinions are national rather than regional which the great area of the USA made almost inevitable in that country in the pre-universal television era.

With the conservative siting policy adopted for the first round of nuclear stations the plants were built in rural areas mostly on the coast and because of the influx of construction staffs, improved amenities and relatively large increases in taxes to the local authorities there was very little local opposition to the stations. Any opposition which existed was more concerned with the impact of the large structures on the visual amenities of the area and sometimes the effects of cooling water in the river or estuary.

## Planning

Before a power station can be constructed irrespective of the prime heat source, planning permission is required under an Act of Parliament dealing with the supply of electricity in general. Notices giving the intent to build the station are posted in prominent places adjacent to the site and in public buildings such as the Post Office and Town Hall. Any individual, municipal authority or company business may object by writing, stating the reasons for so doing, to the Government Department concerned, which in this case is the Department of Trade and Industry, within 28 days. In practice in the interests of the public this period may be extended quite considerably.

Prior to this stage the Planning Department of the CEGB has discussed the project informally with the local government officials and their support for the proposal is solicited. If there are many objections then a public inquiry may be held at the discretion of the Secretary of State for Trade and Industry but if the local planning authority objects then the law requires

positively that such an inquiry be held. The decision as to whether to allow the station to be built rests with the Secretary of State and the inquiry merely serves to assist him. The CEGB mounts a public relations exhibition in the locality and senior Board officials including nuclear safety experts are in attendance to answer any questions in an informal atmosphere. If objections are mainly in the form of questions, public hearings are held and by discussing the various aspects of the station many of the objections are subsequently withdrawn. Quite distinct in law but coincident in time with the planning application, when a nuclear station is involved, a request for a nuclear site licence is made also to the Department of Trade and Industry and by previous informal discussion with officers of the Nuclear Installations Inspectorate, the site characteristics are derived so that argument of a lost cause is avoided. Although objections to the issue of a licence for a nuclear power station are not legislated for except by the freedom of the individual to write directly to the Chief Inspector of Nuclear Installations, questions concerning nuclear safety are asked and answered at the planning inquiry, by both CEGB and Government witnesses.

#### Waste Discharge

The control of discharge of radioactive waste from any source is vested in two Ministries, the Department of the Environment and the Ministry of Agriculture Fisheries and Food. Neither of these organisations has any administrative connection with the Nuclear Installations Inspectorate and each has a long tradition dating back to the 19th Century, of responsibility for controlling the disposal of various industrial wastes of all kinds. The Radioactive Substances Act 1960 in effect extended the powers of these old established ministries to include radioactive wastes and the skilled administration which over many years had won the respect of the public in looking after its interest was readily accepted as being completely impartial by both the nuclear industry and the public at large. An important part of the requirements of the Act is that before an authorisation to dispose of radioactive waste is granted the Ministries must inform and consult representatives of the local authorities in detail about the proposals. Objections can be resolved and no public inquiry is called for at any stage. Consultation at a technical level takes place between the scientists of both the CEGB and the Government Department before a formal application to discharge is lodged and thus any divergence of opinion can be discussed calmly out of the limelight of lobbyists and extremist environmentalists.

#### Local Liaison

Having dealt with the discussions and consultations between the licensee and the statutory bodies before and during construction there is in addition a continuing relationship with local authorities and other interested parties. One of the more interesting of these is the setting up of Local Liaison Committees whose terms of reference are

- (a) To provide information and reassurance on the manner in which radioactive materials are used at the power stations.



- (b) To explain the significance of radiological measurements which are made outside the station boundaries.
- (c) To discuss schemes for the protection of the public in the event of an accident, the schemes to be prepared by the Board in consultation with the appropriate officers of the County and local authorities.

The committee is set up by the Generating Board and representatives are invited from County and local authorities - elected members and officers together with medical officers of health - local bodies who have statutory functions such as water undertakings and river authorities, farming interests both with the National Farmers Union and Country Landowners Associations. In addition any specific organisations within 4 - 5 miles such as Trinity House (due to the lighthouse on Dungeness Head) and the Lydd Airport Authority, also at Dungeness, are invited to attend. In addition senior representatives of the authorising Ministries responsible for the control and discharge of radioactive wastes are members of the committee. The chairman is the Station Superintendent and there are representatives of the station management and Headquarters Nuclear Health and Safety Department. The press are not in attendance at the meeting but an agreed press statement is issued at the end of each meeting.

Meetings take place at the power stations once or twice each year and the results of the routine district surveys and the total curies discharged to the environment as liquid and gaseous waste are reported by the representative of the Department of the Environment; a report of station operation is given by the Station Superintendent and details of emergency plan rehearsals are given by the Station Health Physicist and discussed in detail.

The district survey which is carried out by the Station consists of measurements of airborne radioactivity and gamma dose rate from deposited radioactivity. These readings are obtained at different distances from the reactor in order to give comparative rather than absolute results. In addition any land and marine species of animals which form part of the food chain to man are assayed and the results compared with derived working levels agreed between the regulatory authority and the licensee. Particularly, in rural areas milk is collected and analysed, both for its own sake and also because the cow is a very convenient collector and averager of several radioactive isotopes which may be contaminating pasture land.

#### Emergency Plans

The Emergency Plan is a statutory requirement. It is very comprehensive and specifies the actions required by both CEGB personnel and outside organisations. Frequent discussions and consultations take place with local authority officers during its preparation but particularly with the police who would have the job of instituting action including distribution of stable iodine tablets to local inhabitants and in the ultimate carrying out of any necessary evacuation. Copies of the plans are given to all members of the Local Liaison Committees and they report back to their parent organisation. In all cases the recipients have

behaved in a responsible manner accepting the effort which is put into ensuring safety both of site personnel and local inhabitants rather than using the necessity for such plans as implying an impending hazard.

Regular training sessions of site personnel in emergency actions are carried out including first aid and rescue. Once each year there is a complete exercise of the plan in which a specific incident is simulated and the plan is brought into action, those taking part in the exercise having no prior knowledge of the supposed accident. The police are involved but not the general public. The exercise is witnessed by inspectors of the licensing authority and by the Board's Nuclear Health and Safety Department. Post-exercise discussions and criticism takes place in which all participants, and inspecting authorities, take part and as mentioned above the results of these exercises are discussed at the Local Liaison Committee meetings.

We believe these committees have been a most useful public relations experiment, people have been treated like intelligent and rational human beings and they have behaved like it, at no time has any attempt been made to cover up any incident no matter how minor and on all occasions the committee members have reacted in a helpful manner. Some of these elected members who have been members of these committees for some years have become remarkably knowledgeable.

#### Public Interest

The Board encourage visitors to all its sites both during construction and operation. Special low radiation routes through the station are planned including a visitors gallery on pile cap. Guides consisting of the wives of staff employed on the station are provided with uniforms and given instruction about the power station and they attend as and when required. A typical nuclear power station may have as many as 20,000 visitors each year including parties of children from local schools.

Two projects which are both interesting and important relate to fish farming and trout fishing.

The possibility of using power station discharges for rearing marine fish and shellfish was first discussed with the White Fish Authority about 10 years ago. The objective was to maintain the growth of these animals over the winter months, thereby bringing them to marketable size much sooner than occurs in the sea.

Initial studies were undertaken at the South of Scotland Electricity Board's Hunterston Nuclear Power Station and a conventional CEGB power station. These preliminary investigations using flatfish showed that increased growth occurred. It was decided then, that nuclear stations with their high load factors would be the most suitable sites at which to undertake future studies. Also, many of these stations are sited on open coasts where the water is unpolluted and the salinity is less variable than in estuaries.

At Hinkley Point Nuclear Power Station a private individual

in collaboration with the CEGB, is undertaking pilot scale studies on rearing crustaceans (prawns) and molluscs (oysters and clams) together. He has progressed considerably towards developing the necessary technology for a more extensive project.

The Ministry of Agriculture Fisheries & Food is planning to establish a laboratory on the Wylfa Nuclear Power Station site to investigate all aspects of rearing shellfish. They have undertaken extensive laboratory studies to develop the technology of spawning and rearing larvae. At the same time, rainbow trout have been grown in the discharge on a small pilot scale basis.

Fish farming will be a large scale development of the future, and many countries are undertaking extensive research programmes into the rearing of a variety of species. It is very probable, that in the temperate latitudes warmed discharges will make a valuable contribution to the success of such ventures. Already in the Board, provision is made at suitable new stations to build-in the necessary access to C.W. systems, so that if need be, water can be drawn for use in a fish farm.

The Trawsfynydd Nuclear Power Station, 500 MWe, has been operating successfully since 1965, it uses an inland lake as its source of cooling water. The lake was formed in 1924 to serve a small hydro-electric station to provide a local supply of electricity. An angling club, the Prysor Angling Association, was formed by local inhabitants in the early 1930's to use the natural fishing of the lake comprising brown trout coming from local streams. The lake was emptied and enlarged in 1959 prior to the commencement of the construction of the nuclear power station. At the public inquiry the Board undertook to preserve and assist the fishing rights of the angling club which has in fact continued to control the fishing of the lake. Its activities are managed by a Lake Management Committee and the Power Station Superintendent is a member of the Committee by invitation. The club realised that the lake could be developed far beyond the naturally occurring fish and since 1967 it has been trying to increase the fish population artificially. Unfortunately the brown trout, although giving good sport and good eating, is difficult to rear artificially due, amongst other things, to the predatory activities of perch. It was decided to introduce rainbow trout into the lake and the current programme consists of adding 16,000 fish per annum. Some brown trout continue to be raised on an experimental basis.

The success of the experiment is illustrated by the fact that recently the trials to choose the Welsh team for International fishing competitions were held on the lake. In addition the Board use the lake water to raise trout for stocking other lakes in the area.

The liquid radioactive effluent from the station is discharged into the lake which is also used as a source of cooling water for the turbine condensers.

The presence of small but detectable quantities of radioactive materials in the trout has not deterred the fishing and eating of the catch.

### Local Involvement

Power stations employ small numbers of people, large power stations even of 2,000 MW capacity rarely employ more than 500 people. The Board's policy is to employ locally recruited labour and therefore the power station employees rapidly become part of the local community, unlike large Atomic Energy Authority establishments employing several thousand people imported into a locality and who remain in a separate group. The result is that a power station very quickly becomes "our power station" and "our nuclear power station" is even better.

Finally the activities of some of the radiological protection societies notably the Society for Radiological Protection should be given an honourable mention. This Society provides a lecturing service, free of charge, to a variety of organisations from Rotary Clubs to Mothers' Unions and this has proved a very popular and widely appreciated service to the community as a whole.

### Conclusions

It is believed that the urgent need for nuclear power in the United Kingdom predisposed the public to accept the large structures comprising the power stations as necessary. The accompanying small risk of radiation exposure was also readily accepted because of the confidence which had been built up in the Government Agencies involved by virtue of their historical role in the so-called conventional safety matters such as chemical and biological control.

Whilst the future cannot be predicted with confidence it is hoped that ten years successful and safe operation now achieved will reinforce the tolerance and goodwill which has always existed between the CEGB and its neighbours.

## INFORMING THE PUBLIC ABOUT NUCLEAR ENERGY

H. Kiefer, W. Koelzer, G. Stäblein  
Health Physics Division  
Karlsruhe Nuclear Research Center  
Karlsruhe, Federal Republic of Germany

### Abstract

When the Karlsruhe Nuclear Research Center was founded in 1956, it soon became apparent that the population of the neighboring communities had to be informed about nuclear energy in an unbiased, objective way. Those discussions centered chiefly around arguments of radiation protection. The experience gained from our public relations activities in those years was used and continued by the Nuclear Engineering School of the Karlsruhe Nuclear Research Center, e.g., in special courses run for the information and education of teachers and journalists who passed this information on as opinion leaders. In a similar way it was possible to discuss the objections against siting of the first nuclear power stations in Germany with the population in the environment of those sites.

It was not until the controversy imported from the United States, which can be described simply by the names of Gofman, Tamplin and Sternglass, that organized groups of the population turned up who were against the use of nuclear power. This at the same time made the arguments more emotional, expanding them into problems of energy policy and sociological questions under the headings of "environmental protection". This different situation must be taken into account in public relations activities.

Our experience from numerous and varied discussions with action groups against nuclear power has clearly shown that big rallies and use of the mass media can result only in general information and education of the public. More complex subjects with discussions pro and con, which arise when it comes to the establishment of a nuclear plant, need early and specific approaches to homogeneous groups. These contacts should always be supported by arguments of fact and should cover only a limited subject.

## Introduction

The resumption of nuclear research in the Federal Republic of Germany after the conclusion of the Paris Treaties of 1955 suffered from the severe burden of atomic energy: the bombs of Hiroshima and Nagasaki. This primarily created a sceptic attitude relative to the establishment of research facilities for the peaceful utilization of nuclear energy.

### "Paleozoic" - 1955 - 1960

For the same reason the establishment of the Karlsruhe Nuclear Research Center in 1956 highlighted the necessity of factual information and education about nuclear energy of the population in the communities in the vicinity. The possibility of visiting the plants under construction, experimental lectures about health physics and nuclear technology in schools and institutions for adult education created the first contacts. Study tours of foreign nuclear research facilities, for instance Saclay near Paris in France and Mol near Brussels Belgium, were organized for special groups. This habit introduced in the early years of the Karlsruhe Nuclear Research Center of making the facilities of the Center accessible to all the interested parties at any time was retained in the years to come. At present approximately 15 000 visitors, in groups and individually visit the Karlsruhe Nuclear Research Center; they can get all the information they want, and the staff of the Center are available for discussions with them.

When the Karlsruhe Nuclear Research Center was founded, most of the questions asked by the population referred to radiation protection and radiation exposure of the environment. Most of these questions indicated a genuine concern. In a few cases, however, questions with respect to radiation protection and safety were just a pretence covering up for economic interests. These opponents were afraid that the establishment of a Research Center could cause the workers employed in their small local industries to change to jobs in the Research Center which would offer better pay.

At the earlier meetings informing about nuclear energy homogeneous groups, such as teachers, members of municipal councils, members of agricultural associations, etc. were preferably approached.

### "Mesozoic" - 1960 - 1968

In the early sixties the establishment and the expansion of a Nuclear Engineering School at the Karlsruhe Nuclear Research Center made it possible to pass on information about nuclear research and the peaceful uses of nuclear energy to the public through courses and information meetings. Experience gathered in the early years was thus made use of and expanded.

Besides purely technical courses in radiochemistry or reactor technology, radiobiology and health physics, special courses were organized for specialized teachers in secondary schools from all over the Federal Republic of Germany. In this way it was possible to use teachers as "opinion multipliers" and make use of their educational possibilities and capabilities in order to pass on factual information to the younger generation to be trained in an understandable way.

In those years more and more reports were found in the press which unintentionally gave wrong information. In most cases this indicated an insufficient amount of technical knowledge with many journalists. Consequently, journalists were invited to attend brief courses at the Nuclear Engineering School where experts talked about specific selfcontained subjects, such as "biological and medical problems in the utilization of nuclear power", "reactors of the future", "reprocessing of fuels", or "nuclear safeguard methods".

Both groups, teachers and journalists, greatly helped in the publicity of nuclear knowledge through their capacity as "opinion leaders". In this way problems of radiation protection and safety were discussed, thus preparing a critical public.

These same years saw the construction of the demonstration nuclear power stations of Obrigheim and Gundremmingen in Germany, which gave rise to a thorough discussion with the population in the areas of these plants about problems of site selection. Proper Commissioning then proceeded without any major interruptions.

### "Neozoic" - 1969 - ....

Since 1969 greatly exaggerated reports have also appeared in

the press of the Federal Republic of Germany questioning the arguments of nuclear safety of nuclear power stations and thus creating unrest even among the experts. This controversy, which was imported from the USA and can be outlined by mentioning the names of Gofman, Tamplin and Sternglass, resulted in the organized association of a few groups of the population opposing the application of nuclear power. Although this nuclear controversy and its extension from the USA to Europe, especially to the Federal Republic of Germany, had been recognized by a few experts, the scope and the possible effects had not been correctly assessed and the rate at which this phenomenon spread had been underestimated by industry.

Two factors, most of all, influenced the generation and the extent of the controversy: A generally improving environmental consciousness among broad groups of the population coupled with a certain hostility towards technology or a reduction of faith in technology.

It was necessary to take account of the representation of the problems in a popular book written by a number of experts in which anybody could be able to find factual information on the subject. For this purpose, Deutsche Verlagsanstalt of Stuttgart in early 1970 published a book entitled "Kernenergie - Nutzen und Risiko" [1]. However, it was evident from the outset that a nonfiction book would not be sufficient. Sensational reporting had to be attacked by other means. For this purpose, almost simultaneously a "collection of arguments and counterarguments" [2] was published by the Swiss Association for Atomic Energy and a volume entitled "Kernfragen" [3] by the German Atomic Forum.

Switzerland was early to recognize the direction in which the conflict threatened to move, as a consequence of the reaction of certain groups of the public to the reactor incident at Lucens (January 1969).

At an information and discussion meeting organized in Bern in the fall of 1970 by the Swiss Association for Atomic Energy the situation was indicated. Many German observers experienced their first encounter at this meeting with groups discussing only on an emotional basis. Indeed, dealing with reactor safety and radiation protection at a public forum in this way was a successful venture



with the Swiss population which seemed to be used to democratic discussions. Afterwards, the technical questions discussed were published in a generally understandable form and made available to the interested general public.

#### Development of Arguments

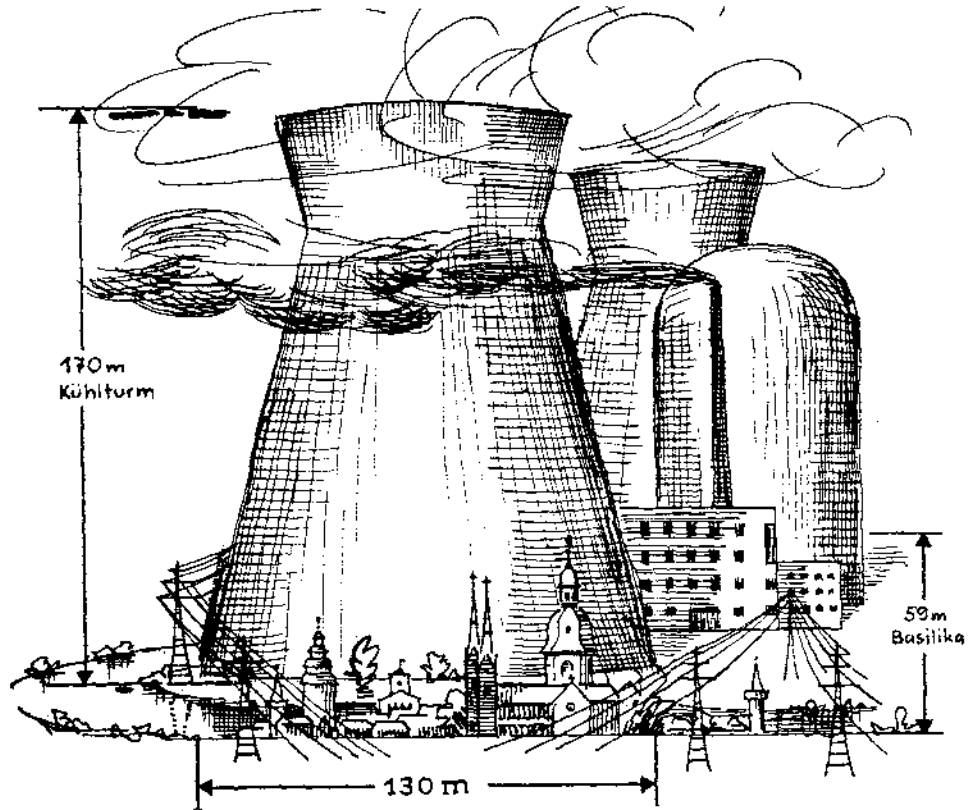
The development of arguments in discussion meetings during those years can be classified as follows:

- During an initial phase there was a generally factual, objective discussion with the interested public which mostly had a limited background knowledge.

- As a second phase organized groups expanded the arguments to other areas (emergency cooling, frequency of cancer, etc.). The discussions became more difficult. Talks with the "atomic opponents" require experts in the respective fields, such as reactor technology or medicine. This phase is the era of sensational stories, exemplified by the name of Mary H. Weik. However, scientific and technical articles are able to convert most of the sceptics from their former negative opinions about the peaceful uses of atomic energy.

- The third phase is determined essentially by the general efforts towards environmental protection. This made the arguments more emotional, extending them under the heading of "environmental protection". Their often hysteric expression leads away from problems of reactor technology and radiation protection or safety to global problems of the future such as "thermal pollution," landscaping and aesthetics, problems of energy policy and sociology. Especially the latter points are supported by political groups operating with the keywords of "changes of the system" against the profit maximization of utilities." This confronts potential reactor operators with a very complex set of questions in this third phase. One example of the emotional, distorted description presented by an action group against nuclear power stations is shown in Fig. 1. Such action-associations do not want any factually correct information, such as Fig. 2, a photomounting of the cooling tower for the nuclear power station at Gösigen, Switzerland.

# Die Stadt „zur schönen Betonaussicht“



Gemeinnützige Aktion FORTSCHRITT FÜR ALLE D 8501 Feucht, Postf. 1230

Fig. 1: Cooling towers, sketch of an action against nuclear power stations



Fig. 2: Photomounting of the cooling tower for Gösgen Nuclear Power Station/ Switzerland (Courtesy Motor Columbus, Baden/Switzerland)

The success of a specific approach of the public can be well studied by the example of the nuclear power station of Neckarwestheim Germany, now under construction.

A large number of hearings were arranged within a very short period at which all the subjects could be finalized in a discussion. The villages in the community, general practitioners, journalists, etc. were invited for discussions.

Even observers not directly linked with the proposed plant, such as clerical organizations, used the opportunity and asked for a discussion between leading members of the project and opponents.

The positive outcome of this informative action in the case of the Neckarwestheim Nuclear Power Station is partly due to a native characteristic of the population of the area: they are realistic and sober people. The problems were really finalized in a discussion, and die-hard opponents were not convinced of the contrary, but the credibility of their arguments was greatly shaken in the eyes of the majority.

The situation is quite different in another siting discussion at Breisach on Kaiserstuhl, a well known winegrowing area near the Southern Black Forest.

Contrary to Neckarwestheim, which mostly covered the symptoms listed under phases 1 and 2 above, the Breisach discussion clearly highlighted the arguments of phase 3. Less specific problems of nuclear power plants were discussed rather than general problems of environmental protection: Cooling processes with wet or dry cooling towers, meteorological effects on winegrowing at a distance of a few kilometers, protection of the landscape, and in particular, the necessity of this nuclear power plant from the point of view of energy policy.

As far as the method used by the nuclear opponents is concerned, it can be said:

Local groupings like to cluster around locally well known personalities, such as the doctor, representatives of the community, chairmen of some associations, etc. who have previously shown their interest in public affairs.

The press of the organized opponent groups is well versed in the art of lending credibility to their arguments by quoting from well known experts. Objections raised in the way of stories even sometimes catch well versed experts by surprise.

Observations have shown that our opponents like to quote foreign technical literature. In Germany American literature is quoted, whilst in the United States, as far as we know, it also applies vice versa. Linguistic incompetency and the inability to follow the quotation often kill any answers that might be given, which weakness is played upon quite consciously by the opponent. In this way any quotation taken out of its context, even if it is a quotation from a well known expert - preferably Nobel prize winners are quoted here - disturbs the listener. He is ashamed of his lack of information and no longer participates in the discussion.

### Experiences

This changed situation must be borne in mind in public relations work. Our experience from numerous and varied discussions with individuals and committee actions against nuclear power has shown quite clearly that large-scale meetings and the use of the mass media will be able to produce only a general information and education of the public. Events of this kind are not the right way of explaining even to an interested public more complex situations, such as the problem of the risk probability, in sufficiently accurate mathematical terms, to make the population risk conscious or to explain problems of cost benefit relations. Alternative thinking when it comes to problem solutions often verges in the well known German quotation of St. Florian: "St. Florian, pass by my house, hit others."

Good results were experienced with homogeneous groups in which one specific subject was discussed at a time. Such groups consisted for instance of physicians, teachers, students, municipal councillors and the leaders of local government groups.

In the light of personal experience gained in meetings of various kinds the authors would like to make the following recommendations:

- No excessive technical specialization.

- Simple, uncomplicated language without any sayings and without any technical terminology.
- Problems should be simplified to a permissible extent in order to meet the understanding of the respective target group.
- Meetings should consist of a brief introduction to the problem followed by a discussion.
- The subject to be discussed should be clearly defined before the meeting by mutual agreement among the groups.
- Organizing several small-scale discussions with greatly varying audiences is preferable to one large-scale meeting.

For each subject that is likely to be touched upon one well-trained expert should be available who has sufficient technical and formal knowledge of the problems.

#### Present Activities

In the light of the overall situation, the management of "Gesellschaft für Kernforschung" advised by the Scientific and Technical Council decided, to establish a department within the health physics division responsible for "Nuclear Power and the Public." This new department is to engage in the discussion between the public and environmental committees and nuclear power. It is to help return the controversy from the emotional, aggressive atmosphere which seems to be preferred, or even sought, by many environmental action committees back into a sober factual atmosphere which will be the only basis for fruitful work in the field of peaceful utilization of nuclear power.

The activities to be pursued by the department will be the observation and critical evaluation of public hearings and the scanning and assessment of all those publications which deal with the subjects of environmental protection in general, and nuclear power and technology in particular.

Other important duties of the department are the informing of all interested parties on the current state of discussion between nuclear technology and environmental committees, which time and again maintain that they represent the public at large; crystallize the controversies and their arguments and finally, make avail-

able to the public factual information about nuclear power and technology through publications and by actively participating in public discussions.

#### References

- [1] K.H. Lindackers, K. Aurand, O. Hug, H. Kiefer, H. Krämer, J. Seetzen, R.R. Trott  
Kernenergie - Nutzen und Risiko  
Deutsche Verlagsanstalt, Stuttgart, 1970
- [2] Kernenergie - Argumente und Grundlagen  
Schweizerische Vereinigung für Atomenergie, Bern, 1971
- [3] Kernfragen  
Deutsches Atomforum, Bonn, 1971

# METABOLISM OF RADIONUCLIDES

## A STUDY OF INHALED SODIUM-22

D. Bush, University Radiation Protection Officer,  
University of Birmingham, Birmingham B15 2TT, England

### Abstract

This is a study of small intakes by inhalation of  $^{22}\text{Na}$  which occurred during the machining of an irradiated target. The arrangements for handling and machining the targets are described. Measured levels of surface contamination and airborne activity, together with particle size data, are given. Whole body counting results, and measurements of the distribution of activity in the body and its variation with time are presented. The information obtained is considered in relation to predictions based on the models of the I.C.R.P. Task Group on Lung Dynamics, and the data given in I.C.R.P. Publication 10.

### Introduction

$^{22}\text{Na}$  is produced on the Nuffield Cyclotron at the University of Birmingham by bombarding magnesium targets with deuterons. At the end of the irradiation, during which about 50 mCi of  $^{22}\text{Na}$  is normally produced, the target is removed from the cyclotron and stored for several weeks to allow short-lived activity to decay. The  $^{22}\text{Na}$  is present in a thin layer on the surface of the target, and this active layer has to be removed so that the  $^{22}\text{Na}$  can be recovered and processed.

This paper reports studies made to estimate the radiation dose resulting from intakes of fine dust produced during the machining of an irradiated target. The intakes were accidental and the bulk of the activity taken into the body is considered to have been inhaled due to the failure to wear a breathing mask.

### Target Machining

Using long handling tongs, the irradiated target is transferred from the storage facility to a shielded enclosure within which is a scraping machine. The walls of the enclosure are made of interlocking lead bricks and a lead-glass window is included to allow the machining process to be observed. Access to the enclosure is via a two part perspex lid, one part of which can be moved. At the time of these intakes, the lead enclosure itself was not ventilated but the room in which the equipment is housed has extract ventilation. The general layout of the room and equipment is shown in Fig.1.

The active layer is removed from the surface of the target by a scraping tool which traverses its face and removes a thin strip of metal. The scraping process is carried out dry without the use of a cutting fluid. The scrapings fall down an inclined trough into a can which is capped and then manually removed using tongs. For the whole operation the operator spends a total of about an hour in the room.

### Surface Contamination Levels

When this scraping facility was originally designed it was not thought that contamination outside the lead enclosure would be significant. However when the facility was brought into use, surveys revealed that loose activity was escaping from the enclosure. Typical levels of loose surface contamination at the end of a scraping run, at the positions indicated in Fig. 1., are given below i.e., much of the room was found to be contaminated to quite significant levels.

Position (See Fig.1)	Surface Activity Level ( $\mu\text{Ci}/\text{cm}^2$ )
S1	$6 \times 10^{-4}$
S2	$1 \times 10^{-3}$
S3	$5 \times 10^{-4}$
S4	$5 \times 10^{-4}$

### Air Contamination Levels

Air samples have also been run during the target loading, machining and can removal stages of the operation. Samples have been taken using air samplers positioned as shown in Fig.1., these positions being close to the positions occupied by the operator. Typical levels of airborne activity are shown below and these should be compared with the 40 hour week M.P.C. air value of  $2 \times 10^{-7} \mu\text{Ci}/\text{cm}^3$  and  $9 \times 10^{-9} \mu\text{Ci}/\text{cm}^3$  for 'soluble' and 'insoluble' materials as given by I.C.R.P.<sup>1</sup>.

Position (See Fig.1)	Air Activity Level ( $\mu\text{Ci}/\text{cm}^3$ )
A1	$1 \times 10^{-8}$
A2	$2 \times 10^{-7}$

### Size Selective Sampling of Airborne Activity

In order to obtain information required for specifying the requirements of filters to remove this airborne activity, size selective sampling has been carried out using a cascade centripeter of the type described by Hounam<sup>2</sup> and calibrated by O'Connor<sup>3</sup>. Samples were taken at the positions shown in Fig. 1 during target loading, machining, and can removal and results typical of those obtained are shown below. This distribution is very close to a log-normal distribution with an activity median aerodynamic diameter of  $2.7 \mu$  and a geometric standard deviation of 1.9.



Aerodynamic diameter (microns)	Percentage of particles less than stated diameter
12.5	99.3
4.0	69.4
1.5	19.6

#### Whole Body Counting after Accidental Intake

On one occasion the target scraping machine operator failed to wear a breathing mask. When this was known it was realised that an inhalation had probably occurred, so it was decided to carry out whole body counting in order to investigate the distribution of activity and its variation over time and to estimate the dose.

Strictly speaking the intake was not a single well-defined intake but several small intakes over a period of some hours. The first whole body count was made 2 days after the estimated mid-point of the intake and a further 7 counts were made, the last being 67 days after intake. Counting was in the Na<sup>24</sup> photopeak, and the results obtained have been corrected for normal background and <sup>40</sup>K contribution by using data from an uncontaminated person of similar build to the person concerned.

At the first count, a total count of 22,680 was recorded, corresponding to 0.4 $\mu$ Ci of <sup>22</sup>Na activity in the body at that time. The results of the whole body counts were plotted to estimate the count and activity on the day of the intake (t=0), and the results were then normalised to the result at t=0. These are plotted in Fig. 2.

#### Distribution of Activity in the Body

The whole body counter used for this study has four large sodium detectors. Two are above the subject and positioned at the chest and head and the two below the subject are at the head and knees. For all the counts that were made, the distribution of counts among the four detectors was generally constant and not significantly different to the distribution found following the injection of <sup>24</sup>Na into a human subject.

As well as counts made using the whole body counter, checks were also made using a collimated sodium iodide detector which could be accurately positioned over various parts of the body. Counts were made 2, 11, and 18 days after intake with the detector positioned over each lung and the lower part of the abdomen on each occasion. These measurements showed no significant change in the distribution of activity at these three positions over the time interval mentioned above.

#### Predicted Behaviour of Inhaled <sup>22</sup>Na and Comparison with Recorded Data

From the size selective air sampling data, the amounts of activity deposited at various sites can be predicted using the Deposition Model of the Task Group on Lung Dynamics<sup>4</sup>. This model gives the following predicted depositions.

Region	Percentage of Inhaled Activity Deposited
Nasopharynx	60 - 65
Tracheo-bronchial	8
Pulmonary	15 - 20

The I.C.R.P. Task Group on Lung Dynamics also proposes a clearance model and suggests clearance times and routes from various regions for several categories of inorganic compounds. In the case of this study, the radioactive material was sodium, but most of the original magnesium was also present and it was of interest to know how the active sodium would behave in this situation. In contact with moisture and body fluids the hydroxides of these metals would form. Sodium hydroxide is listed by the I.C.R.P. Task Group as a Class D material exhibiting rapid clearance from the lung, whereas magnesium hydroxide is a Class W material exhibiting intermediate clearance. For the radioactive sodium hydroxide, the Task Group's Clearance Model predicts clearance as below.

Region	Clearance percentage, route, and biological half-life
Nasopharynx	50% to systemic blood, 4 mins 50% to G.I. tract, 4 mins
Tracheo-bronchial	50% to systemic blood, 10 mins 50% to G.I. tract, 10 mins
Pulmonary	80% to systemic blood, 30 mins 20% to lymph, 30 mins, and then all to blood, 30 mins

Therefore if the sodium quickly separates from the magnesium, within a few hours of the inhalation all the activity will have been transferred to the G.I. tract and systemic blood. That transferred to the G.I. tract will also quickly transfer to the blood<sup>5</sup> so the behaviour after that time would be expected to be identical to that of orally administered  $^{22}\text{Na}$ . Confirmation that this is effectively so from two days after the intake is given in Fig. 2 where the normalised whole body counts are compared with the data given in Publication No. 10 of I.C.R.P.<sup>6</sup>, in which the clearance of  $^{22}\text{Na}$  is described by a three component exponential derived from whole body counting studies of orally administered  $^{22}\text{NaCl}$ . It is seen that there is quite good agreement between the measured whole body activity and that predicted by the I.C.R.P. clearance formula, particularly bearing in mind the reported variability<sup>7</sup> in biological elimination rate that can occur as the stable sodium intake is varied. Unfortunately no data is available to confirm the predicted very rapid clearance from the lung, but the data from 2 days after the exposure confirm that no translocation of activity from the lung occurred after that time. This is in accord with the findings of similar studies made from 9 to 285 days after the inhalation of another Class D material ( $^{137}\text{Caesium sulphate}$ ) by Miller<sup>8</sup>.

### Dose Estimate

From the I.C.R.P. Task Group on Lung Dynamics deposition model, of the total initial deposit of 0.5  $\mu\text{Ci}$ , the deposits in the nasal, tracheo-bronchial and pulmonary regions are expected to be 0.34, 0.046, and 0.114  $\mu\text{Ci}$  respectively. For the nasal and tracheo-bronchial regions it has been assumed in each case that the activity has been uniformly deposited over an area of 100  $\text{cm}^2$ , cleared according to the biological half-times of 4 and 10 minutes, and the resulting doses have been calculated to be 3.3 and 1.1 mrem respectively. Using the data given by I.C.R.P.<sup>1</sup> the average dose to the lung, assuming a biological half-time of 30 minutes, has been calculated to be 0.14 rem, and from the data given in I.C.R.P. Publication 10, the whole body dose has been estimated to be 10 mrem.

### Conclusion

From the above evidence it is concluded that the 0.5  $\mu\text{Ci}$   $^{22}\text{Na}$  inhaled was rapidly cleared from the lung and thereafter exhibited the same behaviour as does orally administered  $^{22}\text{Na}$ . The most significant dose was the whole body dose of 10 mrem, the estimated additional doses to the nasal, tracheo-bronchial, and pulmonary regions being only 3.3, 1.1 and 0.14 mrem respectively.

### Acknowledgements

I am pleased to acknowledge the help of Dr. H. James, Experimental Pathology Department, who carried out the whole body counting, and of Mr. R.G. Harris of the Physics Department. The co-operation of the person involved in this study is also greatly appreciated.

### References

- 1. Recommendations of the I.C.R.P., Report of Committee 2. I.C.R.P. Publication 2, Pergamon Press, Oxford (1959).
- 2. Hounam R.F. The Cascade Centripeter, AERE-M 1328 (1964)
- 3. O'Connor D.T. Calibration of a Cascade Centripeter Dust Sampler, AHSB (RP) R108 (1971).
- 4. Report of the I.C.R.P. Task Group on Lung Dynamics, Health Physics 12, 173-207 (1966).
- 5. Stara J.F., Nelson N.S., Della Rosa R.J., and Bustad L.K., Health Physics 20, 113-137 (1971).
- 6. Recommendations of the I.C.R.P., Report of Committee 4. I.C.R.P. Publication 10, Pergamon Press, Oxford (1968).
- 7. Smilay M.G., Dahl L.K., Spraragen S.C., and Silver L., Journal of Laboratory and Clinical Medicine, 58, 60 (1961).
- 8. Miller C.E., Health Physics 10, 1065-1070 (1964).

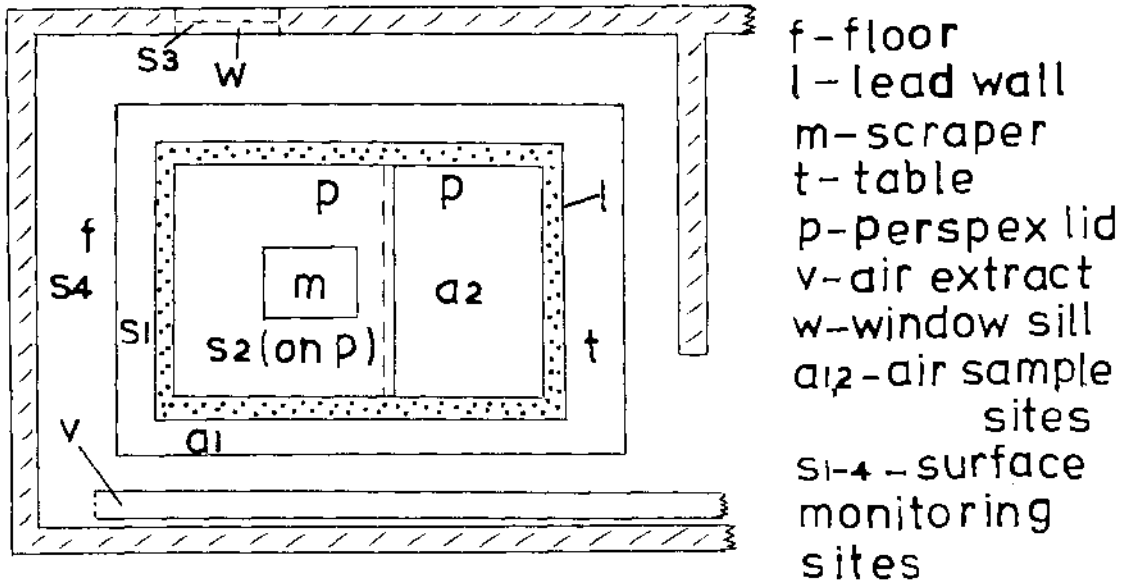


Fig.1. Layout of scraping facility.

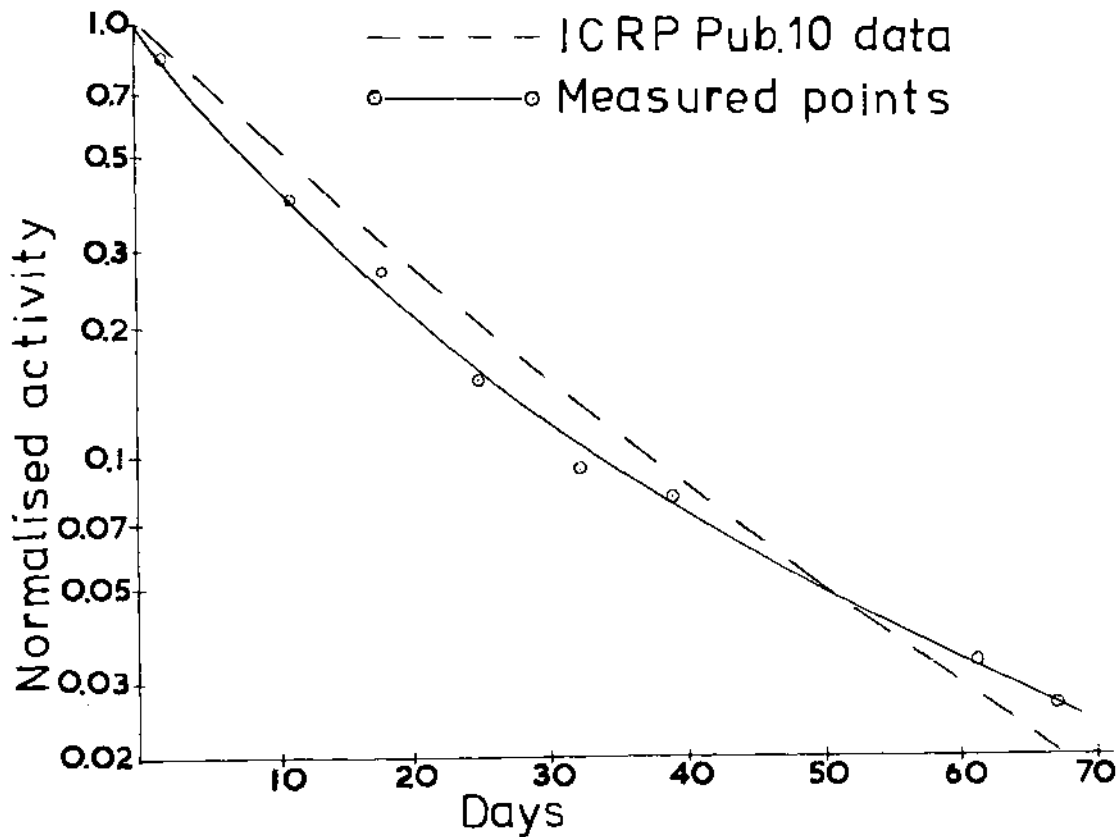


Fig. 2. Variation of whole body activity.

## МЕТАБОЛИЗМ СВИНЦА-210 И ПОЛОНИЯ-210 В ОРГАНИЗМЕ

А.И. Нижников, Е.И. Долгирев, М.С. Ибатуллин,

П.В. Рамзаев, М.Н. Троицкая

Институт радиационной гигиены, Ленинград, СССР

### ABSTRACT.

Metabolism parameters of Pb-210 and its product (in the body) Po-210, necessary for dosimetry and standardization are obtained in man during short-termed observations. The authors of this report have carried out their investigations on a volunteer group. These investigations deal with the specification of absorption and clearance rate of these isotopes administered per os as indicators and sufficient for many years' observations using in vivo measurements and excretion analyses. Genetically related mixture of Pb-210 and Po-210 was administered orally to the four people (the authors) whose excretions had been previously analyzed for Po-210 for a year after a single incorporation. This experiment, as the authors think, will contribute to the understanding of Po-210 metabolism. The first results of the investigations are presented in this paper and we intend to go on with these experiments in the forthcoming years.

### Реферат

Параметры метаболизма свинца-210 и образующегося из него (в организме) полония-210, необходимые для дозиметрии и нормирования, в настоящее время получены у человека в основном в кратковременных наблюдениях.

Авторы данного сообщения выполнили исследование на группе добровольцев по уточнению коэффициентов всасывания и скорости выведения этих изотопов, принимавшихся через рот в индикаторных количествах, достаточных для многолетних наблюдений по приближенным измерениям в теле и по экскретам. Четыре человека (авторы исследования), у которых предварительно в течение года исследовалась экскреция полония-210 после однократного его поступления, приняли через рот генетически связанную смесь свинца-210, висмута-210 и полония-210. Такая схема опыта по замыслу авторов позволяет выяснить обмен полония-210, который формируется в организме непосредственно из свинца-210. В докладе приводятся первые результаты исследования, которое будет продолжено в последующие годы.

### Введение

Если метаболизм полония-210, первично поступающего в организм человека, изучен достаточно полно для целей дозиметрии и нормирования, то этого нельзя утверждать по отношению к свинцу-210 и, особенно, к образующемуся из него в организме полония-210. В пуб-

ликациях  $^{10}$  и  $^{10A}$  (МКРЗ)<sup>1</sup> данные по метаболизму свинца- $^{210}$  не приводятся. В литературе отсутствуют также и сведения о судьбе полония- $^{210}$ , образующегося в организме из свинца- $^{210}$ , без которых расчёт поглощенных доз не может быть выполнен, поскольку основная доза создается не самим свинцом- $^{210}$ , а его дочерним полонием. Различия в обмене полония, первично поступающего в организм ("внешний" полоний) и полония, образующегося в местах фиксации свинца через висмут- $^{210}$  ("внутренний полоний"), вряд ли могут быть отвергнуты без данных эксперимента.

Принятая система параметров метаболизма свинца- $^{210}$  (публикация-2, МКРЗ)<sup>2</sup> включает: коэффициент всасывания 0,08, биологические периоды полувыведения для всего тела (в сутках) - 1460, скелета - 3650, печени - 1947, почек - 531, отношение полония- $^{210}$  к свинцу- $^{210}$  во всем теле - 0,09, почках - 0,18, печени - 0,18, скелете - 0,10 и др.

Приведенные значения в последние годы были подвергнуты проверке рядом исследователей (Харш и Суомела<sup>3</sup>, Хольцман<sup>4</sup>, Кохен, Рен и Айзенбад<sup>5</sup>, Бланхард<sup>6</sup>, Хилл<sup>7</sup>). По нашим предыдущим исследованиям высказывалось сомнение, в частности, по коэффициенту всасывания свинца- $^{210}$ , который в отдельных случаях достигал 50%. Появились также доказательства, что отношение полония- $^{210}$  к свинцу- $^{210}$  в скелете равно 0,7-0,8, а не 0,1 как это принято в публикации 2 МКРЗ.

Все изложенные соображения дают достаточно оснований для постановки заранее запланированных исследований по изучению метаболизма свинца- $^{210}$  у человека в условиях надежного контроля поступления изотопа и его выведения в многолетних наблюдениях.

#### Методы и материалы исследования

Четыре практически здоровых человека (три мужчины и одна женщина) в возрасте от 45 до 50 лет приняли индикаторное количество хлористого полония- $^{210}$  однократно через рот в виде раствора при  $pH=3$  в количестве 0,5 микрокюри на человека. Удельная активность раствора определялась на четырех-пийном счетчике по альфа-излучению. В течение недели ежедневно, а затем 1 раз в неделю в течение месяца и далее каждый месяц в течение 8 месяцев собирались полные суточные выделения мочи и кала. В пробах выделений определялся полоний- $^{210}$  после его осаждения на фольгу из никеля. Суточное выведение изотопа выражалось в % от введенной дозы. Этот предварительный опыт был поставлен для того, чтобы получить данные по обмену первично поступающего "внешнего" полония- $^{210}$  у каждого испытуемого и в последующем опыте учесть их при определении "внутреннего" полония- $^{210}$ . Далее те же лица, когда у них остатки изотопа стали пренебрежимо малы, приняли однократно в растворе соляной кислоты ( $pH=3$ ) генетически связанную смесь Рв- $^{210}$ , Вд - $^{210}$  и Ро- $^{210}$ , полученную при многолетнем хранении стеклянных ампул с радоном-222. В смеси оказалось 1,0 микрокюри Ро- $^{210}$ , 1,9 микрокюри Вд - $^{210}$  и 3,3 микрокюри Рв- $^{210}$ . Определение принятой активности в растворе осуществлялось:

полония- $^{210}$  - по альфа-излучению на четырехпийном счетчике,  
висмута- $^{210}$  - по бета-излучению на том же счетчике,  
свинца- $^{210}$  - по равновесному висмуту- $^{210}$  (бета-излучение), по  
гамма-излучению свинца- $^{210}$  и по альфа-излучению равновесного  
полония- $^{210}$ .

Стандарты 100% введенной дозы готовились также путем введения раствора изотопов в чистые суточные пробы мочи и кала. Ошибка измерения дозы не превышала 1%.

После приема изотопов осуществлялся сбор суточных выделений по ранее описанной схеме. Одновременно испытуемые измерялись на

счетчике всего тела (гамма-спектрометре с датчиком из кристалла йодистого натрия толщиной 1 мм и диаметром 15 см). Детектор размещался против лобной кости. Регистрировалось излучение свинца-210 с энергией 46 кэв.

В выделениях проводились определения:

- полония-210 как по альфа-излучению высушенных проб, так и после осаждения его на фольгу из никеля,
- свинца-210 в высушенных пробах на гамма-спектрометре, по бета-излучению равновесного висмута-210 и по повторному осаждению полония-210 на никелевую фольгу после предварительного накопления.

### Результаты исследования

Полный анализ всех отобранных проб еще не закончен. Он требует значительного времени. По мере отдаления от времени приема дозы и уменьшения уровней в экскретатах методы суммарной альфа и бета-активности, как и гамма-спектрометрия проб оказываются неприемлемыми из-за возрастающих ошибок измерений (более 10%). Уже через 2 недели мы были вынуждены определять свинец-210 по полонию-210, повторно осаждаемому на диски никеля после накопления в течение 1-2 месяцев.

Приведенные в таблицах 1-4 материалы и вытекающие из них выводы ко времени открытия конгресса могут быть значительно пополнены. Здесь из-за ограниченности места мы хотим обратить внимание читателя лишь на явно доказуемые положения, не требующие обширных обосновывающих выкладок:

1. Среднее всасывание полония-210 при приеме его в чистом виде или в смеси со свинцом-210 превышает 18% (таблица 1) и 22% (таблица 2); среднее всасывание свинца-210 превышает 18% (таблица 3).

Таблица 1

Выведение "внешнего" полония-210 в % введенной дозы  
(в предварительном опыте, без приема свинца-210)

Экскреты	Среднее выведение по 4 испытуемым в дни после приема									
	1	2	3	4	5	6	35	41	145	160
Кал	28,92	38,40	13,58	3,87	1,44	0,82	0,03	0,03	0,025	0,010
Моча	0,29		0,07	0,04	0,03			0,03	0,007	0,0014

2. Выведение "внешнего" полония-210 в интервале времени от 6 до 160 суток после приема через рот происходит по  $T_{эф} = 36$  суток, что хорошо согласуется с  $T_{эф} = 40$  суток, принятым в публикации Ю МКРЗ. С калом выводится полония-210 в 10-20 раз больше, чем с мочой; хотя в отдельные сутки эти различия не всегда проявлялись.

3. В начальном периоде после поступления смеси свинца-210 и полония-210 вычленив в экскретатах "внутреннего" полония-210 не представляется возможным. Судя по расчетам экскреции дочернего "внутреннего" изотопа и полагая его обмен одинаковым с "внешним" полонием, в первые 5 дней "внешний" полоний в сотню раз превышал уровень выделения "внутреннего" изотопа. И лишь через 50-150 дней можно ожидать превалирования "внутреннего" полония над "внешним" в 2-6 и более раз. Фактическую динамику выведения и ее параметров предстоит получить в дальнейших наблюдениях.

4. Выведение из организма свинца-210 происходит все с более

Таблица 2

Выведение "внешнего" и "внутреннего" полония-210 у людей в % от введенной "внешней" дозы. Изотоп принимался в смеси со свинцом-210

шифр испытуемого		Сутки после введения								
		1	2	3	4	5	6	7	14	
РПВ	кал	47,26	20,10	8,49	1,59	0,21	0,46			0,021
	моча		0,05			0,02	0,03			
ТМН	кал	44,50	26,11	14,57	4,78	1,07	1,26	0,38		0,127
	моча		0,12	0,31	0,02	0,04	0,05	0,02		0,054
НАИ	кал	36,27	46,88	7,70	0,53	0,26	0,26	0,11		0,166
	моча	0,02	0,02	0,04	0,03	0,01	0,04	0,08		0,010
ИМС	кал	28,84	11,86	17,00	0,00	4,27	0,75	2,34		0,616
	моча		0,02	0,21	0,01	0,01	0,05	0,03		0,011
Среднее	кал	39,22	26,29	11,94	2,30	0,51	0,68	0,94		0,232
	моча	0,02	0,06	0,19	0,02	0,02	0,04	0,04		0,025

Таблица 3

Выведение свинца-210 у людей в % от введенной дозы

Шифр испыту- емого		Сутки после введения									
		1	2	3	4	5	6	7	14	21	28
РПВ	кал	55,69	15,57	7,83	1,03	0,23	0,22		0,07	0,02	0,01
	моча	0,47	0,21	0,13	0,09	0,07	0,08		0,07		
ТМН	кал	40,35	27,62	11,64	4,58	0,51	0,51	0,09	0,07	0,05	0,08
	моча	0,32	0,03	0,025	0,05	0,06	0,02	0,02	0,08		
НАИ	кал	34,94	44,51	8,68	0,32	0,12	0,06	0,05	0,06	0,05	0,06
	моча	0,18	0,08	0,07	0,03	0,02	0,03	0,03	0,03		
ИМС	кал	38,56	19,72	23,74	0,00	7,55	0,29	0,33	0,09	0,04	0,015
	моча	0,46	0,10	0,09	0,03	0,15	0,07	0,06	0,01		
Сред- нее	кал	42,39	26,35	12,97	1,98	0,29	0,27	0,16	0,07	0,04	0,04
	моча	0,36	0,11	0,08	0,06	0,05	0,05	0,04	0,05		

Таблица 4

Скорость счета от свинца-210 (30-56 кэв) в лобной кости ( в % от первого измерения )

Шифр испытуемого	Сутки после введения							
	2	8	15	22	29	36	99	128
РПВ	100		125	90	87	80	68	
НАИ	100		94	90	80			
ИМС	100	100	155	120	110	135	65	45
ТМН	100	209	200		209	213		148
Среднее по пе- рвым трем испы- туемым	100	100	127	101	96	98	67	45



замедленной скоростью и достаточно заметный спад начинается примерно через 15 дней после приема активности. С 15 по 29 сутки, если судить по измерениям лобной кости, скорость выведения свинца-210 из организма соответствует 34,6 дням, а за последующие 99 суток - 90,5 дням. Скорость выведения свинца-210 по экскретам оказалась примерно такой же как и уменьшение счетности в лобной кости.

### Заключение

Полученные данные позволяют предположить, что некоторые коэффициенты метаболизма свинца-210 и полония-210, принятые в существующих рекомендациях по нормированию, не обеспечивают соответствующих гарантий. Всасывание этих изотопов (18-20%) у человека выше в 2-3 раза, чем принято. При поступлении их в составе пищевых продуктов, как было ранее нами показано<sup>8</sup>, достигает 50%.

В длительных многолетних опытах предстоит получить и другие константы метаболизма свинца-210 и полония-210, которые до настоящего времени не могут считаться представительными.

### Литература

1. Radiation Protection. ICRP Publication 10, Oxford, 1968.
2. Радиационная защита. Вторая публикация МКРЗ. Госатомиздат, 1961.
3. Hursh J.B. and J.Suomela. Absorption of Pb-210 from the gastrointestinal tract of man. Acta Radiol., v.7, 1968, pp108-20.
4. Holtzman R.B. Pb-210 and Po-210 metabolism in radium-dial painters. Proceedings of the Meeting on Biology and Ecology of Polonium and Radiolead. 1970, Belmont, UK.
5. Cohen N., Cohen L.K., Wrenn Mc.E. and M. Eisenbud. The retention and distribution of Pb-210 in the baboon and the rat. Ref.4.
6. Blanchard R.L. Use of 210-Pb and 210-Po blood levels. Ref.4.
7. Hill C.R. Routes of uptake of Po-210 into human tissues. In. Radioecological Concentration Processes, Oxford, 1968, pp. 297-302.
8. Ramzaev P.V., Nizhnikov A.I., Litver B.Y., Troitskaya E.N., Ibatulin M.S., et al. Absorption of 210-Pb from gastrointestinal tract of rat and man. 1970, ref.4.

USE OF A SPECIALIZED ANALOG COMPUTER FOR THE MODEL OF  
THE METABOLISM OF SOME RADIONUCLIDES

I.A.Likhtarev, G.V.Arkhangel'skaya, A.I.Dobroskok,  
I.A.Zvonova, N.F.Korelina, G.P.Krasnoshyeykova,  
V.S.Repin, A.P.Ushakova

The Institute of Radiation Hygiene

The RSFSR Ministry of Public Health

Leningrad, USSR

Introduction

Methods for mathematical modelling of the radionuclide metabolism in the human body and experimental animals and the method of compartment models, in particular, have been successfully employed in the solution of many theoretical<sup>10</sup> and applied<sup>5</sup> problems for a long time.

When the behavior of a radionuclide-containing system is described by a linear model, the solution is, generally, easy to obtain. The computation difficulties involved usually arise either from the amount of computing to be performed or from the incompleteness of the set of the model parameters.

However, there are a number of situations where the linear kinetic models of radionuclide transport cannot be applied. These include cases where coefficients in differential equation systems are functions of time or contain products of functions. Such systems cannot be integrated analytically, as a rule, even in quadrature solutions. Meanwhile, the solution of such problems is of considerable practical and theoretical value, since this class of models includes the interaction of radionuclides with chelate complexing agents, e.g. DTPA, the protective action of stable iodine and, finally, the models simulating the changes of metabolic "constants" with aging.

A continuous solution for non-linear models may be offered and the structure of these solutions can be studied for arbitrary input functions through the application of analog computers (AC) equipped with special devices for non-linear systems of the abovementioned types.

Although the use of analog computers for the purposes of the compartment analysis has been already discussed<sup>2,4</sup>, the application of AC in the studies of non-linear models of the DTPA action, iodine metabolism and age-related changes of some parameters is a new aspect to the problem.

This paper deals with these models and some methods of solution and discusses the effectiveness of this computer analysis technique. Space does not permit a detailed discussion of each problem and an exhaustive analysis of the solutions obtained. Thus, the aim of this paper is to examine the techniques involv-

ed in the solution of these problems and to suggest suitable structural schemes to be analysed on analog computers. Therefore, specific solutions for the models under consideration and the numerical evaluation of their parameters will be outside the scope of this work.

### 1. Non-Linear Model of the Decorporative Effect of DTPA

It was assumed in constructing the model that:

(a) A single dose of hepatoosteotropic radionuclide, e.g. plutonium, is injected and distributed among the organs and tissues of the organism which may be represented as a system of four compartments (Fig.1). These are a compartment of blood including the pool of soft tissues X, two compartments representing the parts of the skeleton  $Y_1$  and the liver  $Y_2$  where exchange occurs and, finally, a general compartment Z of the radionuclids which slowly exchanges in the skeleton  $Z_1$  and the liver  $Z_2$  ( $Z_1+Z_2=Z$ ).

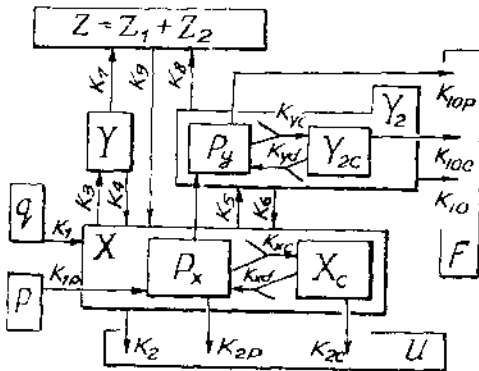


Fig.1. Nonlinear metabolic DTPA-Pu model

(b) DTPA (P) administered to the bloodstream ( $P_x$ ) may be further transferred to the exchange compartment of the liver ( $P_y$ ) or go to urine (U) and faeces (F).

(c) The formation and disintegration of the DTPA - radionuclide ( $X_c$  and  $Y_c$ ) complex occurs in the blood compartment and the exchange portion of the liver, the excretion rates of DTPA and the complex being the same and exceeding by far that of the radionuclide itself.

This mechanism underlies the decorporative effect of DTPA.

(c) The rate of complex formation is proportional to the product of concentration values of the radionuclide and chelate, and for the total volume of distribution - to the product of their amounts X  $P_x$  and Y  $P_y$ . The proportionality factor represents the effectiveness and selective properties of the chelate in the organism media.

The above model is described by the following system of differential equations:

$$\frac{dX}{dt} = k_1 q - (k_2 + k_3 + k_5)X - k_{xc} P_x X + k_{xd} X_c + k_4 Y_1 + k_6 Y_2 + k_9 Z;$$

$$\frac{dY_2}{dt} = k_5 X - (k_6 + k_8 + k_{10})Y_2 - k_{yc} P_y Y_2 + k_{yd} Y_{2c};$$

$$\frac{dY_{2c}}{dt} = k_{yc} P_y Y_2 - k_{yd} Y_{2c} - k_{10c} Y_{2c};$$

$$\frac{dU}{dt} = k_2 X + k_{2c} X_c; \quad (\text{Eq. 1})$$

$$\frac{dX_c}{dt} = k_{xc} P_x X - k_{xd} X_c - k_{2c} X_c;$$

$$\frac{dF}{dt} = k_{10} Y_2 + k_{10c} Y_{2c};$$

$$\frac{dY_1}{dt} = k_3 X - (k_4 + k_7)Y_1;$$

$$\frac{dP_x}{dt} = k_{1p} P - (k_{2c} + k_{5p})P_x + k_{xd} X_c;$$

$$\frac{dZ}{dt} = k_7 Y_1 + k_8 Y_2 - k_9 Z;$$

$$\frac{dP_y}{dt} = k_{5p} P_x - k_{10c} P_y + k_{yd} Y_c$$

on the initial condition that:

$$q(0)=1; P(\tau)=1; x(0)=y_1(0)=y_2(0)=Z(0)=P_x(\tau)=P_y(\tau)=0 \quad (\text{Eq.2})$$

The above system (Eq.1) was studied on an analog computer, some operational amplifiers of which were used for the modelling of DTPA transport and some other - for the nuclide transport (Fig.2).

As seen from the system (Eq.1), the complexing process is included in the terms  $KXcPx$  X and  $KycPyY_2$  of the model. This pro-

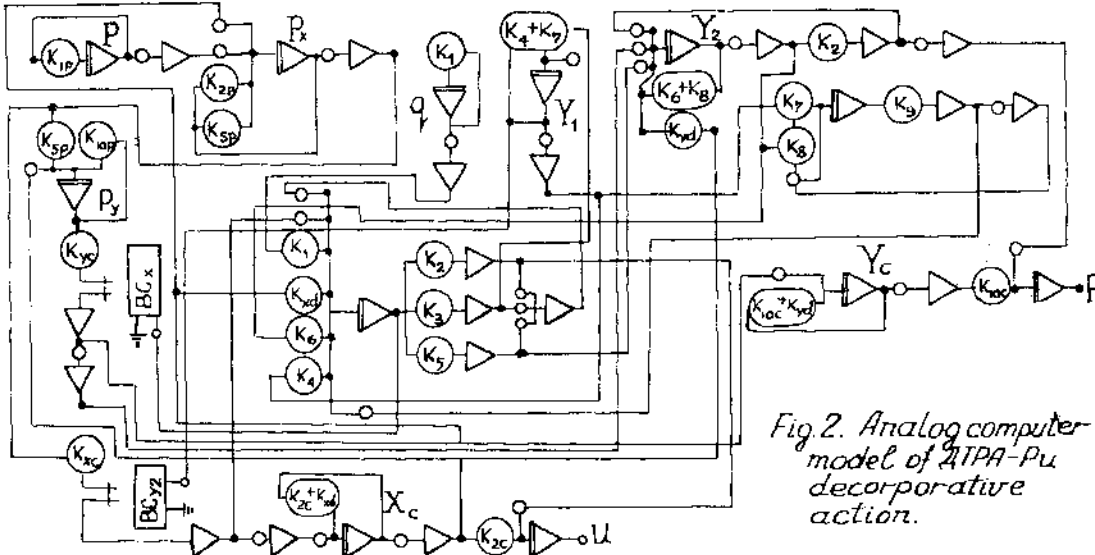


Fig.2. Analog computer model of DTPA-Pu decorporative action.

cess was performed by special units of the computer which effected the same reaction through the scheme of  $KXcPx \delta_0(x)$  and  $KycPy \delta_0(Y_2)$ .  $\delta_0(x)$  and  $\delta_0(Y_2)$  are Kronecker's symbols.

$$\delta_0(x) = \begin{cases} 1, & x > 0 \\ 0, & x = 0 \end{cases}, \quad \delta_0(y) = \begin{cases} 1, & y > 0 \\ 0, & y = 0 \end{cases}$$

$\delta_0$  imply the termination of complexing, when the free radio-nuclide content of the compartments X and  $Y_2$  becomes equal to zero.

Application of multiplier units results in a considerable error and a decision bias due to the output residual voltage.

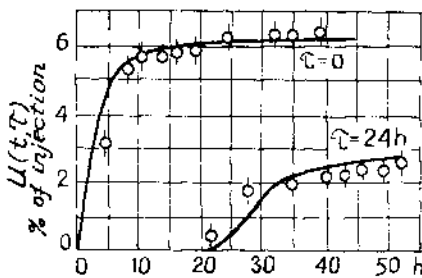


Fig.3. The solution  $U(t)$  of system (1).

To compare the predictions with the real behavior of the system, the results of our own study of the initial stage of the metabolism and excretion of plutonium citrate in the rat's organism, following a single administration of 1 ml 5% solution of DTPA, were used (Fig.3).

As the graphs show, the computed prediction for the function  $U(t)$  is consistent with the experimental findings. Also, similar results were obtained for the functions  $F(t)$  and  $X$ . The predictions and experimental curves for citrates  $^{241}\text{Am}$  and  $^{144}\text{Ce}$  were also found to be in good agreement, the magnitude of constants  $K_i$  being, naturally, quite different.

To summarize, it should be noted that the application of AC offers an apparent advantage of producing multiple sets of continuous solutions for any compartment, for arbitrary functions of the  $q$  and  $P$  intakes. The model in question reproduced on the analog computer may be used in the study of different modifica-

tions of DTPA administration with a view to devising an optimal protection.

## 2. Non-Linear Model of Iodine Metabolism

Below follows a description of the structural scheme that may be used both in the evaluation of tactics of protection of the thyroid gland with potassium iodide and in the case of radiation destruction of the compartment "thyroid". Consider the two-compartment model of iodine metabolism (7) shown in Fig.4.

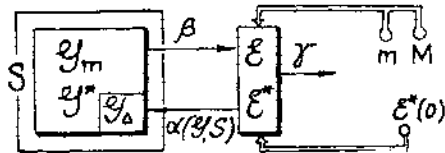


Fig. 4. Two-compartment model of iodine metabolism.

The disturbance in exchange processes is induced by a change in the rate of the iodine uptake by the thyroid, i.e. by a change in the communication constant of  $\alpha$ . Bonnel and Adams' offered a linear approximation of the  $\alpha$  dependence on the level of the thyroid globulin depot repletion:

$$\alpha = R(1 - y_m/S), \quad (\text{Eq.3})$$

where  $R$  corresponds to the maximal iodine absorption from plasma ("full iodine insufficiency").

$G_m$  - level of iodine pool repletion (mg iodine) due to its dietary intake (mg in 24 hrs).

$S$  - full iodine pool - the size of the globulin depot expressed in terms of mg iodine the maximum amount of which can be taken up by the thyroid.

The uptake of large amounts of stable iodine results in a rise in the  $G_m$  level which is limited by  $S$ .

An exposure of the thyroid to doses of tens of kilorads may cause the destruction of the globulin depot  $S$ .

By substituting (Eq.3) into the set of equations which describe the model, we obtain:

$$\begin{aligned} dy/dt &= R\epsilon - R y_m \epsilon / S - \beta y, \\ d\epsilon/dt &= -(R + \gamma)\epsilon + R y_m \epsilon / S + \beta y + m + M(\tau) \end{aligned} \quad (\text{Eq.4})$$

$m$  - dietary intake of iodine

$M(\tau)$  - quantity of stable iodine injected for the thyroid blocking at a moment  $\tau$ .

A modification of the same system for radioactive iodine will be:

$$\begin{aligned} dy^*/dt &= R y_m \epsilon^* / S - \beta y^*, \\ d\epsilon^*/dt &= -(R + \gamma)\epsilon^* + R y_m \epsilon^* / S + \beta y^*, \end{aligned} \quad (\text{Eq.5})$$

where  $S = S(D)$ , while, in its turn,  $D = K_D \int y^*(t) dt$ .

The approximate form of this function for humans will be as shown in Fig.5.

The differential equations (4) and (5), as shown in a general form, have no analytical solutions.

To construct a model of this system on AC, a relevant structural scheme should cover:

- (a) the state of iodine metabolism described by the equilibrium values of the pools and flows of stable iodine;
- (b) changes occurring in the system, when large quantities of stable iodine are administered; the blockage of the thyroid;
- (c) an adequate response to the administration of the radioactive tracer;
- (d) the destruction of the globulin depot  $S$  caused by the radiation injury of the thyroid gland.

Fig.6. shows a structural scheme which meets all the above re-

quirements.

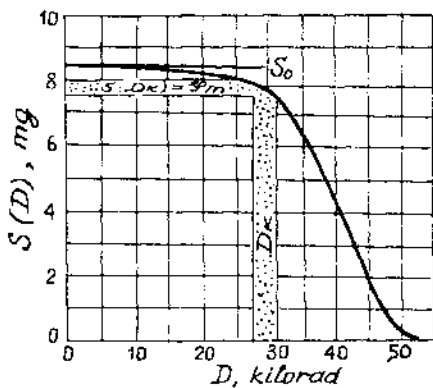


Fig. 5. The size of globuline depot as a function of irradiation dose.

To cause a disturbance in the analog model, the computer is supplied with doser elements which make it possible to generate voltage pulses varying in frequency and amplitude.

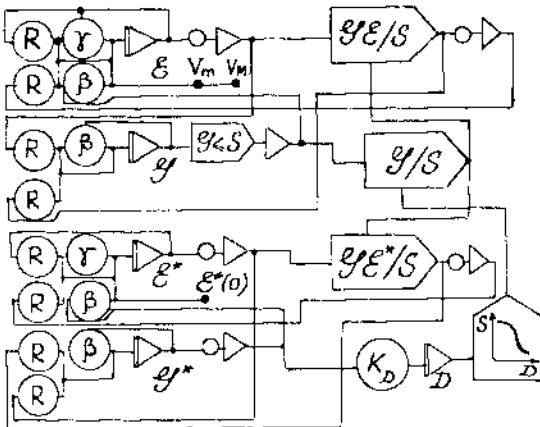


Fig. 6. The structural scheme of non-linear model of iodine metabolism.  $\square$  non-linear transformations unit

### 3. Age-Related Model of Strontium and Calcium Metabolism

The dietary intake of global strontium-90 by humans commences as early as the birth, and even earlier, in prenatal life. The existing strontium-calcium metabolism models, however, are based on the parameters of adult standard man. It stands to reason that these parameters, including those of the compartments, should change with aging  $\tau$ . These changes are primarily due to the build up of the skeletal calcium pool in the course of its development with time.

The changing requirement in the calcium taken with diet  $q(\tau)$  is related to the skeletal pool increment  $dx/d\tau$  and the size of this pool  $X$  by the following non-linear differential equation:

$$dx/d\tau = q(\tau) - \kappa(\tau)x. \quad (\text{Eq. 6})$$

This equation is a special case of a more general four-compartment model<sup>6</sup> and a solution may be obtained by numerical methods.

Using the data on the age-associated requirements in calcium

The S-level changes are induced by diode elements, as the dose function is fed to their inputs. The G magnitude with respect to the level of S is limited by means of a diode limiter.

The application of dividers and multipliers imposes certain restrictions at the choice of the scale. For instance, the equilibrium values of  $G_m$  and S are 7 and 8 mg, respectively. Stable iodine doses may be as large as 500 mg. If a 100 V scale of the analog computer is used, voltages corresponding to  $G_m$  and S values will be within 10 V. Since the procedures of division and multiplication of low values of voltage involve a considerable error, the stability and reproducibility of solutions may be adversely affected.

An optimal method of protection may be developed by means of simulating different patterns of radioactive and stable iodine intakes with the aid of these doser elements.

Fig. 7 gives an illustration of the computer-predicted curve for radioiodine excretion from the thyroid in conditions of large dose burdens. The sharp bend corresponds to such magnitude of  $S(D_k)$  that  $S(D_k) = G_m$ , while the destroyed portion of the globulin depot  $G_a = S(0) - G_m$ . At this moment  $\alpha(G, S) = 0$ . A general discussion of this problem can be found in our paper<sup>7</sup>.

found in<sup>11</sup> and the Mitchell curve<sup>9</sup> which gives the rate of calcium

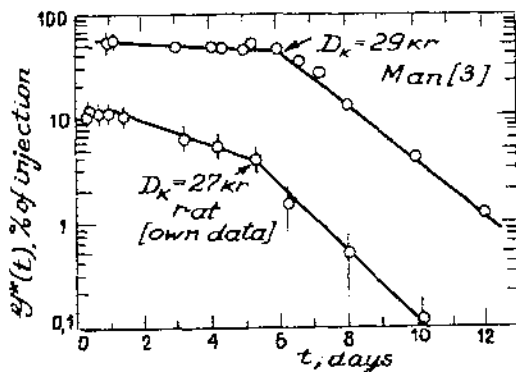


Fig.7. Computer prediction of iodine excretion from thyroid at great dose barbens  
o - experimental data.

increment in children's skeleton, the function  $K(T)$  is easily derived from Eq.6. By introducing the compartment of plasma the calcium concentration in which does not change with age, it is possible to construct a more general two-compartment model (Fig. 8a) which will correspond to the system (Eq.7).

$$\frac{dX_{pt}}{dt} = [K_u(\tau) + K_1(\tau)]X_{pt} + K(\tau)X + q(\tau), \quad (\text{Eq.7})$$

$$\frac{dX}{dt} = K_1(\tau)X_{pt} - K(\tau)X.$$

When modelling the system in analog computer  $q(\tau)$  function and non-linear parameters  $K(\tau)$  and  $K_1(\tau)$  were simulated with

the aid of special non-linear units. AC solutions are in good agreement with calculation results.

### Conclusion

Three types of non-linear problems of the compartment analysis are considered. The problems are studied and solved by means of an analog computer. To this end, a suitable structural scheme was developed for each problem. The obtained predictions are in good agreement with some experimental findings, which give every reason to consider the suggested models to be adequate.

Thus, the analog computer modelling provides an effective means for the quantitative analysis of the sufficiently complicated non-linear problems of radionuclide transport in the living organism.

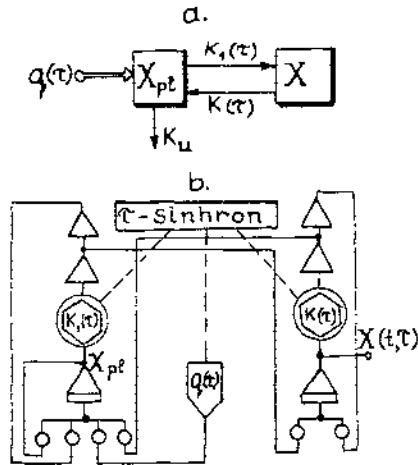


Fig.8. a. Age-related model of Sr and Ca metabolism  
b. The structural scheme of AC-model.

### References

1. Adams C.A., Bonnell J.A., Health Phys., 7, 127, (1962)
2. Fish B.R., Health Phys. 1, 276 (1958)
3. Goolden A.W., Davey J.B., Br.J. Radiol., 36, 340 (1963)
4. Gregg E.C., "Multi-compartment analysis of tracer experiments" Ann., of the New York Academy of Sciences, 108, 128 (1963)
5. ICRP Publication 00, ICRP /69/ C4 - 2/4 (1969)
6. Лихтарев И.А. и др., Мед.радиология, I, 70 (1972).  
Ильин Л.А. и др. "Радиоактивный йод в проблеме радиационной безопасности", М., Атомиздат, (1972).
7. Marshall et al., (ICRP Publication 20), Health Phys., 2,125, (1973)
8. Mitchell H.H. et al., Biol. Chem., 158, 625 (1945).
9. Sheppard C.W., and Housholder A.S., J.Appl. Phys., 22, 510 (1951)
11. Standard Physiological Values for Standard Man: Intake, Metabolism and Excretion, ICRP, April (1968).

INTERPRETATION OF EXPERIMENTAL DATA ON POLONIUM-210  
METABOLISM FOR COMPUTING ADMISSIBLE LEVELS

Yu.D. Parfenov

Institute of Biophysics of the USSR Ministry of Health  
Moscow Union of Soviet Socialist Republics

Abstract

Experimental data on  $^{210}\text{Po}$  metabolism in animals show that there are some uncertainties for the value of absorption into the blood from the gastro-intestinal tract and from the respiratory system; for the choice of the critical organ and for assessment of the nonuniformity of the internal irradiation. However there is no real basis for radical changes in the existing admissible levels of  $^{210}\text{Po}$  intake into the human body.

$^{210}\text{Po}$  is one of the most highly toxic radionuclides /1/. The existing ICRP recommendations are based on the admissible  $^{210}\text{Po}$  content in the body of the professionals equal to 0.03  $\mu\text{Ci}$ , and the spleen is taken as a critical organ with  $^{210}\text{Po}$  content equal to 0.002  $\mu\text{Ci}$ /2/. This paper sets out to analyse experimental data on  $^{210}\text{Po}$  metabolism in animals for calculating admissible levels of this radionuclide intake in man.

Absorption from the gastro-intestinal tract

ICRP recommendations are based on the value of  $^{210}\text{Po}$  absorption from gastro-intestinal tract into the blood equal to 0.06. This value was obtained by administering inorganic compounds of  $^{210}\text{Po}$  to animals /3, 4, 5/. Our data for dogs and rats confirms this value. However Morrow et al. /6/ ha



shown already that absorption into the blood depends on physico-chemical properties of the  $^{210}\text{Po}$  compound administered. According to their data for cats the rate of absorption from intestinal tract for soluble polonium citrate was 10 times higher than for colloidal polonium. It was Hill /7/, Litver /8/, Kauranen and Miettinen /9/, who first drew attention to the fact that the assessment of the natural  $^{210}\text{Po}$  intake by people from the Arctic regions who eat reindeer meat gives a value of  $^{210}\text{Po}$  absorption from the intestinal tract, which is much higher than was assumed from experiments. Our indirect assessment of the natural  $^{210}\text{Po}$  absorbed into the blood, which enters the human body from the environment mainly through food, is 0.35 /10/. This high absorption into the blood may be explained by the fact that  $^{210}\text{Po}$  which enters the body with meat or other food stuffs is in form of organic compounds, where it is bound with highly soluble aminoacids. Another reasonable explanation for this fact is that the  $^{210}\text{Po}$  natural intake into the body is  $10^5$ - $10^6$  times less the amounts of  $^{210}\text{Po}$  administered to experimental animals. This difference may influence the physico-chemical state of  $^{210}\text{Po}$  microquantities with pH in the intestinal tract, and thus the level of absorption into the blood. Johnson and Watters' latest data /11/ show that  $^{210}\text{Po}$  entering rats in the form of organic compounds with milk from exposed cows is absorbed from the gastro-intestinal tract into the blood in much higher levels than with administration of inorganic compounds. This may serve to confirm the first proposition. Thus the level of absorption of  $^{210}\text{Po}$  in the form of organic compounds in food stuffs is higher than that in the form of inorganic compounds. It would thus seem that there is no reason to increase the value of the  $^{210}\text{Po}$  absorption coefficient in order to calculate the admissible intake for professionals, as they are dealing with inorganic  $^{210}\text{Po}$  compounds. However, when calculating the  $^{210}\text{Po}$  intake via food chain it is necessary to take into account the fact that organically bound radionuclide is more easily absorbed into the blood.

#### Absorption from the respiratory system

The level and rate of radionuclide absorption from the lungs into the blood are mainly determined by the degree of solution of the inhaled compound.

Berke and DiPasqua /12/ consider that  $^{210}\text{Po}$  absorption into the rats' body after multiple inhalation amounts to 20-26% of the radionuclide inhaled. The  $^{210}\text{Po}$  absorption coefficient for rabbits' lungs which was obtained through comparison of  $^{210}\text{Po}$  retention after intravenous and intratracheal administration, the retention being measured by histoautoradiography and counting tracks, is 27-48% of the total amount of radionuclide retained in the lungs /13/. For rats, 29.4% of the  $^{210}\text{Po}$  administered intratracheally is absorbed into the blood. Smith et al. /14/ obtained a value of 20.3-32.3% for the  $^{210}\text{Po}$  absorbed by dogs inhaling polonium chloride. Little and McGandy /15/ studied the  $^{210}\text{Po}$  absorbed into the blood through smoking. According to their assessment at least 38% of the total amount deposited in the lungs is absorbed

into the blood. Thus the probable quantity of the inhaled compound, assessed on the basis of above-mentioned data, allows us to say that about 19-34% of the  $^{210}\text{Po}$  inhaled is absorbed into the blood.

The dynamic lung model for retention and clearance of radionuclides inhaled into the respiratory system /16/, which was developed by ICRP, is an important step forward compared with their former recommendations. According to this model polonium and its compounds belong to the "W" class (moderately soluble compounds). This model's parameters, characterizing the  $^{210}\text{Po}$  behavior in the respiratory system, correspond to the actual process of  $^{210}\text{Po}$  clearance from the lungs. Our data for rats and other experimental data /14/ fully confirm that the longest biological half life from lung is 50 days.

According to the ICRP model the quantity of radionuclide absorbed into the blood from respiratory system is determined by the following expression:

$$\frac{D_3 f_a \lambda_a}{\lambda_a + \lambda_r} + \frac{D_4 f_c \lambda_c}{\lambda_c + \lambda_r} + \frac{D_5 f_e \lambda_e}{\lambda_e + \lambda_r} + \frac{D_5 f_h \lambda_h f_i \lambda_i}{(\lambda_h + \lambda_r)(\lambda_i + \lambda_r)}$$

where  $D_3, D_4, D_5$  - the corresponding coefficients for deposition in three regions of the respiratory system (nasopharynx, tracheobronchial region and pulmonary region) which depend on the aerodynamic diameter of the particles;  $f$  - corresponding fractions of radionuclide in each region cleared with the according constants of biological elimination -  $\lambda$  ( $f$  and  $\lambda$  do not depend on the particles' size);  $\lambda_r$  - the constant of radioactive decay. Substituting the corresponding figures for  $D, f$  and  $\lambda$  for the "W" class and  $\lambda_r = 0.005$  for  $^{210}\text{Po}$  and solving this expression, we obtain an absorption coefficient of 9.4-14.5% for the particles in size range of 0.01-10  $\mu\text{m}$ . As can be seen from the above, this calculated value is substantially less than the absorption value for  $^{210}\text{Po}$  obtained from actual experiments. This divergency can be explained by the fact that  $^{210}\text{Po}$  in the body can not be considered a homogeneous, moderately soluble compound, because in pH in the body polonium can be found in two fractions simultaneously - aggregated (insoluble) and ionic (soluble) - which have their own rates of clearance from the system.

Analyzing the routes of  $^{210}\text{Po}$  metabolism in the respiratory system according to the ICRP model, it can be seen that 80% of the material deposited in the pulmonary region is transported by the cilia through the tracheobronchial tree to the gastro-intestinal tract and only 15% with  $T_{1/2}(\text{biol.})=50$  days is absorbed through alveolar membranes into the blood. It would seem that the value of  $f_e$  - fraction transported from the pulmonary region into the blood, for  $^{210}\text{Po}$  amounts to 0.45 instead of 0.15. There follows a parallel reduction in the values for the  $f_i$  and  $f_s$  - fractions transported to the gastro-intestinal tract, to 0.3 and 0.2 respectively. Once the parameters have changed in this way the absorption coefficient will be equal to 15.8-32%, which corresponds more closely the actual experimental data.

## Distribution in the body

$^{210}\text{Po}$  in the blood is found in two forms - in an ionic, highly dispersed state and in an aggregated state simultaneously.

The highly dispersed ionic form of  $^{210}\text{Po}$  probably bound with organic acids, is easily soluble and transportable;  $^{210}\text{Po}$  in this form is distributed in the body correlating with sulphur distribution, being its analogue and possible substitute in organic compounds.  $^{210}\text{Po}$  in this form is easily excreted through the kidneys with urine.

The other fraction is the aggregated form of  $^{210}\text{Po}$ . Its basis is the colloidal or pseudocolloidal forms of  $^{210}\text{Po}$  with the body pH. This form of  $^{210}\text{Po}$  can be nonspecifically bound with protein. The aggregated  $^{210}\text{Po}$  is not diffusible because it is in the form of large aggregates which cannot enter the membranes and the walls of blood capillaries. They are phagocitized by macrophages and the cells of the reticulo-endothelial system. That is why this form of  $^{210}\text{Po}$  is mainly deposited in the liver, spleen, lymphatic nodes and, partly, in the adrenal glands. This form of  $^{210}\text{Po}$  is excreted through the intestines with the bile. These states of  $^{210}\text{Po}$  do not appear to be stable.  $^{210}\text{Po}$  can transfer from one state to the other. Thus, for example, aggregated  $^{210}\text{Po}$  can be destroyed transferring to the highly dispersed form under the influence of complex-formation with organic ligands.

A part of  $^{210}\text{Po}$  (apparently in the dispersed form) is adsorbed to the surface of the erythrocytes, and when the latter are destroyed it is deposited in the spleen and liver. The ratio of these two forms of  $^{210}\text{Po}$  depends on many factors: the pH environment, the presence of other chemical compounds (phosphates, citrates, thiols et al) and, finally, on the concentration of polonium atoms.

## Critical organ

Though at present the spleen is considered as a critical organ for  $^{210}\text{Po}$ , there exist at least three more organs with the same radiosensitivity level, where  $^{210}\text{Po}$  retention is 2-3 times higher than in the spleen - that is the kidneys, liver and lymphatic nodes. At the same time  $^{210}\text{Po}$  concentration in the gonads, regarded as belonging to the first group of radiosensitivity, is only 2-3 times less than in the spleen. We observed that the relative  $^{210}\text{Po}$  retention in the spleen decreases in the following succession: mouse-rat-rabbit-dog-man. This speaks for the fact that it is hardly reasonable to choose the spleen as a critical organ.

The earliest changes under the influence of minimal  $^{210}\text{Po}$  quantities exceeding admissible levels 10-50 times, may be found in the function of the liver enzymatic system, in the skin and endothelial capillaries, in the blood system and in the state of enzymatic systems and bile secretion function of the liver. Those changes are as follows: transient bilirubinaemia, increased aldolase content in the blood serum, changes in the volume of renal plasma flow; displacement of adsorption exponents of  $^{131}\text{I}$  bengal-rose by the liver cells /17/. With higher

levels of radiation after  $^{210}\text{Po}$  administration to experimental animals the liver and kidney also show more signs of serious damage compared with other organs. However, the relatively uniform  $^{210}\text{Po}$  distribution with prevailing retention in the reticulo-endothelial system leads to the irradiation of practically all the organs and tissues. In connection with this, damage to the neuro-endocrinal systems is characteristic for  $^{210}\text{Po}$ ; this damage in its turn indirectly furthers the development of radiation damage in various organs stemming from the direct impact of  $\alpha$ -irradiation /18/. The essentially adaptive reactions of the sympatho-adrenal system and the pituitary-adrenal cortex system may exceed the physiologically expedient level and aggravate the progression of radiation sickness. One should bear in mind that the direct impact of  $^{210}\text{Po}$  on the hypothalamopituitary region and on the reproductive system is more important than its effects on the reticuloendothelium, where the radioisotope preferentially accumulates. The concept of "the critical organ" or the preferentially irradiated organ is inadequate for analysis of the pathological process following exposure to  $^{210}\text{Po}$ . It is probably fair to say that the critical organ concept is valid only when we consider the reaction of the body as a whole that develops as a result of the direct impact of the isotope on tissues and its indirect effects. An this considerably complicates the choice of the critical organ for  $^{210}\text{Po}$ . However calculation of admissible irradiation levels taking various systems as the "critical organ" (kidneys, liver, spleen, reproductive system, the whole body) reveals no essential divergencies for assessing admissible intake.

#### Non-uniformity of irradiation

In spite of the generally uniform  $^{210}\text{Po}$  distribution in the body, the difference between the highest concentration (in kidneys) and the lowest (in the skeleton) constitutes two orders. In the homogeneous tissues the  $^{210}\text{Po}$  distribution is fairly diffusive and relatively uniform. However in some organs (kidney, spleen) the distribution of  $^{210}\text{Po}$  is non-uniform. Thus, for example, the  $^{210}\text{Po}$  concentration in the renal convoluted tubules is approximately 30 times higher than in the medulla, as was obtained by the histoautoradiographic method used for rabbits /13/. Even with the regular intake of radioactivity by man at uranium mines the  $^{210}\text{Po}$  concentration in the renal cortex is 4.2 times higher than in the medulla /19/. In rabbit spleen after a single administration of  $^{210}\text{Po}$  the ratio of red pulp/phollicule concentrations is about 5/13/. On the whole, the maximum concentrations of the radionuclide in the kidney and spleen are usually 2-3 times higher than the mean concentration for organ, which is used to calculate admissible levels. In connection with this fact one should take into account the non-uniformity of distribution while assessing the maximum levels of irradiation on the basis of the mean concentration for the organs (or the total content in the body divided by the organ's weight).

### Elimination half-life from the organs

The decrease of  $^{210}\text{Po}$  in the organs after a single administration is well described by one exponential function during a long-term observation. Only in the initial phase, which is limited to several days after administration, it is possible to determine for certain organs one more rapidly eliminated fraction with a short half-life (lymphatic nodes, liver, kidney). The rate of decrease for  $^{210}\text{Po}$  amounts, as a result of biological elimination and radioactive decay expressed through  $T_{\text{eff}}$ , can be taken as similar for all organs and tissues. The  $T_{\text{eff}}$  differences in the experimental data can be explained by the insufficient number of measurements, by individual deviations and by the range of measurements. This speaks for a similar mechanism of  $^{210}\text{Po}$  metabolism in tissues. The  $T_{\text{eff}}$  value ranges from 29 to 42 days /18/ as was confirmed by experimental data on dogs, rats and mice after a single  $^{210}\text{Po}$  administration. The value of 37 days may be generally accepted. This value corresponds well to the data for man which range from 22 to 47 days /21-24/. The rate of  $^{210}\text{Po}$  excretion from rabbits is considerably higher.  $T_{\text{eff}}$  for rabbits is about 6-19 days /25/.

### Conclusion

Thus after analyzing the experimental data on  $^{210}\text{Po}$  metabolism obtained from animals, it is possible to conclude that there are some uncertainties regarding the coefficient of absorption into the blood from the gastro-intestinal tract (organically bound compounds of  $^{210}\text{Po}$ ) and from the respiratory system; regarding the choice of the "critical organ" and the assessment of non uniform internal irradiation in organs. However at present, these uncertainties do not provide adequate ground for substantially modifying existing admissible levels of  $^{210}\text{Po}$  intake for man.

### References

1. Morgan, K.Z. et al. Health Phys. 10 151 (1964).
2. ICRP. Report of Committee II on Permissible Dose for Internal Radiation. 1959. Health Phys. 3 (1966).
3. Antony, D.C. et al., Int. Conf. peaceful Uses atom. Energy (Proc. Conf. Geneva, 1955) 13, UN. New York (1956), 215.
4. Stannard, I.N. Radiation Research. Suppl. 5, 49 (1964).
5. Fink, R.M. (ed.). Biological Studies with Polonium, Radium and Plutonium. McGraw-Hill, New York, Toronto, London (1950).
6. Morrow, P.E. et al. Radiation Research. Suppl. 5, 60 (1964).
7. Hill, C.R. Radioecological concentration Processes. Oxford. London (1967) 297.
8. Litver, B.J. The  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  migration through lichen-reindeer-man chain. Thesis. Leningrad (1972).
9. Kauranen, P., Miettinen, I.K. Health Phys. 16 3 287 (1969).
10. Ladinskaya, L.A. et al. Arch. Environ. Health (in press).
11. Johnson, I.E., Watters, R.L.  $^{210}\text{PoO}_2$  metabolism in Ruminants. Final Report and Summary. COO-2044-5 (1972).
12. Berke, H.L., DiPasqua, A.C. Radiation Research 16 591 (1962).
13. Parfenov, Yu.D., Solovjev, A.I. Strahlentherapie 143 3 362 (1972).

14. Smith, F.A. et al. Am. Ind. Hyg. Ass. J. 22 201 (1961).
15. Little, I.N., McGandy, R.B. Arch. Environ. Health 17 693 (1968).
16. ICRP. Task Group on Lung Dynamics. Health Phys. 12 173 (1966).
17. Guskova, A.K., Baisagolov, G.D. Radiation sickness of man. Moscow. Medicina (1971).
18. Moroz, B.B., Parfenov, Yu.D. Polonium-210 effect on organism. Moscow. Atomizdat (1971).
19. Blanchard, R.L., Moore, I.B. Health Phys. 21 499 (1971).
20. Jackson, S., Dolphin, G.W. Health Phys. 12 4 481 (1966).
21. Guskova, A.K. et al. Med. radiologija 9 51 (1964).
22. Kalmykov, L.Z. et al. Med. radiologija 12 26 (1969).
23. Taylor, N.A. Health Phys. 19 1 147 (1970).
24. Foreman, H. et al. Am. J. Roentgenol. 79 1071 (1958).
25. Parfenov, Yu.D., Polubojarinova, Z.I. Int. J. Rad. Biol. (in press).

"REGULARITIES IN METABOLISM OF RADIOACTIVE ISOTOPES  
UPON INCIDENCE ON THE SKIN"

Ilyin L.A.

Institute of Biophysics Ministry of Health USSR  
Moscow, USSR

Abstract

This report based on the experimental data and calculated-dosimetric estimates for the first time presents a discussion on the metabolic regularities of radionuclides of the elements of various groups of the Mendeleev periodic system when their solutions are applied to the skin. Common features in the distribution of these agents in the skin irrespective of their chemical origin have been established. Essential differences in the accumulation levels of radioactive substances in the skin and in the level of percutaneous resorption have been found. It is shown that transfollicular route is the main way of radionuclide penetration into the body through the skin.

In order to develop the problem of setting standards for skin contamination with radioactive substances and to find effective decontamination means, one must have proper data on the regularities in metabolism of radioactive substances when they come in touch with the skin integument.

Three consecutive stages in the metabolism of radioactive substances are conventionally assumed as follows:

on the surface of the skin, within the skin and within the body.

1. The analysis of the kinetics of radioactive contamination of the skin and of the dynamics of its decontamination may allow us to assume that radionuclides on the surface of the skin are distributed superficially at least as three layers /1,2 /. The upper friable and easily removed layer is formed as a result of mechanical deposition of radionuclides (carriers). The second layer is formed due to physico-chemical processes that determine the interaction of a radioactive

substance with active groups (radicals) of the skin superficial structures. Formation of the third layer /<sup>2</sup>/ is stipulated by ionic exchange between the macrocomponent (the surface of the skin) and the microcomponent (the radioactive ion).

Results of hystoautoradiographic studies allow us to come to the conclusion that the main route for the penetration of the majority of radioactive substances into the skin is the transfollicular route and to a lesser extent it is the trans-epidermal route /<sup>3</sup>/.

2. Metabolism of radioactive substances within the skin layer after their solutions are deposited on the surface of the skin has been studied mainly by Soviet scientists /<sup>3-5</sup>/ as well as by Japanese specialists /<sup>6-9</sup>/. Methods of solving this complex task have been found and use the technique of radiometry of consecutive horizontal skin sections 20-40  $\mu\text{m}$  thick /<sup>3</sup>/. This allowed us to have data on the levels of accumulation, the mode of the intercutaneous distribution and elimination from the skin of pigs of uranium fission fragments ( $\text{Cs}^{137}$ ,  $\text{Sr}^{89}$ ,  $\text{Ba}^{140}$ ,  $\text{Y}^{91}$ ,  $\text{Ce}^{141,144}$ ,  $\text{Nd}^{147}$ ,  $\text{Pr}^{143}$ ,  $\text{Te}^{132}$ ,  $\text{Mo}^{99}$ ,  $\text{I}^{131}$ ) /<sup>3</sup>/ as well as of  $\text{Po}^{210}$  /<sup>5</sup>/ and transuranium elements ( $\text{Pu}^{239}$ ,  $\text{Am}^{241}$ ) /<sup>4</sup>/.

Analysis of the experimental data has established that the character of the distribution within the skin of uranium fission products is the same despite their chemical origin. The characteristic feature is a sharp reduction in the concentration of radionuclides within the skin according to the depth of the layers down to 200  $\mu\text{m}$ , a more moderate reduction in the layer from 200 to 600  $\mu\text{m}$  and almost a uniform distribution in still deeper layers.

Analogous regularities have been also noted in principle for  $\text{Po}^{210}$ ,  $\text{Pu}^{239}$  and  $\text{Am}^{241}$  /<sup>4,5</sup>/.

The curve showing the distribution within the skin of fragmentary radionuclides down to the depth of 600  $\mu\text{m}$  is well described by the following exponential equation:

$$C_x = C_0 (a_1 e^{-K_1 x} + a_2 e^{-K_2 x} + a_3 e^{-K_3 x}), \text{ where}$$

$C_x$  is the concentration of a radionuclide at the depth of  $x$ ,  $\mu\text{Ci}/(\text{cm}^2 \cdot \mu\text{m})$ ,  $C_0$  is the concentration extrapolated to the depth  $x=0$ ,  $x$  is the depth in  $\mu\text{m}$ ;  $a_1$ ,  $a_2$ ,  $a_3$  are the contribution of each exponent to the general function of distribution;  $K_1$ ,  $K_2$ ,  $K_3$  are the constants characterizing the gradient of decreased concentration for the corresponding exponents,  $\mu\text{m}$  (see Table 1).

Experimental data and calculated absorbed doses within the skin for  $\beta$ -sources of various energies prove the adequacy of the surface distribution model for estimating absorbed doses of  $\beta$ -radiation in the skin, beginning with the energy of 0.2 MeV. In the case of more soft  $\beta$ -radiation sources, account should be made of the contribution to the dose due to the effect of radioactive substances that have penetrated into the skin /<sup>3</sup>/.

Osanov and his colleagues /<sup>4</sup>/ have estimated experimentally the dose distribution function for a thin plane  $\alpha$ -source.



While the general character of the intercutaneous distribution of radioactive substances within the skin is quite of one and the same type and does not depend considerably on the time of the radionuclide exposition on the surface of that organ, the levels of their accumulation within the skin differ considerably. According to this index, all the radionuclides that have been studied may be arranged in the following order:  $I^{131}$ ,  $Po^{210}$  >  $Ce^{144}$ ,  $Pr^{143}$ ,  $Nd^{147}$  >  $Mo^{99}$ ,  $Te^{132}$  >  $Sr^{89}$ ,  $Ba^{140}$  >  $Cs^{137}$  >  $Pu^{239}$ ,  $Am^{241}$ .

According to the data obtained by us, the maximum accumulation of the radionuclides within the skin is achieved in a relatively short interval after the radioactive solution is applied onto the skin (within 15 minutes to one hour). The amount of radioactive substances in the skin (the surface layer 30  $\mu$ m deep being excluded) is within the range from 4-5 per cent ( $I^{131}$ ,  $Po^{210}$ ) to 0.1 per cent ( $Pu^{239}$ ) of the amount of the radionuclide applied to the surface of the skin.

In order to study the intimate exchange mechanisms of radioactive substances in the skin it is very important to have data available pertaining to the peculiarity of their interaction with various biochemical components of that organ.

Table 2 presents the data of the distribution of the radionuclides of molybdenum, tellurium, strontium, barium, yttrium, cerium, praseodymium and neodymium between various skin fractions (soluble in lipids, soluble in water and residual).

These studies carried out in cooperation with Shvydko, N.S., /3/ have also considered the distribution of radionuclides in the above mentioned fractions after their isolation from epidermis, derma and subcutaneous connective tissue. All the experiments yielded identical results that testified to the fact that despite their chemical origin, all the radionuclides present in the skin were mainly linked with insoluble proteins and hardly solving inorganic compounds (phosphates, sulphates, etc).

A relatively small amount of radionuclides interact with lipids and proteins soluble in water. All this allows us to suppose one type of mechanism that determines parameters of time for exchange and elimination of radionuclides from the skin.

This hypothesis is to some extent corroborated by the results of preliminary studies /5,10/ aimed at estimating effective half-lives ( $T_{eff}$ ) of a number of radioactive substances in the skin (see Table 3).

As one can see from Table 3, despite the origin of a radionuclide in this or that group of elements of the Mendeleev periodic system, they have a common feature which is a rapidly exchangeable fraction (with  $T_{eff}$  equal to several hours) that accounts for 65-95 per cent of the amount of radionuclides deposited in the skin. The contribution of the relatively slowly exchangeable fraction (with  $T_{eff}$  equal to several days) is of the order of 5-35 per cent.

3. The levels of percutaneous resorption of radioactive substances into the body depend on the type of the chemical compound and the aggregate state of a radionuclide. As can be seen from systematized data obtained during our studies and from the published data, the range of values of this index lies within several hundredth fractions of a per cent up to several per

cent of the amount of the radionuclide applied to the surface of the skin. This allows us to classify radioactive substances according to their low, medium and high level of percutaneous resorbtion. With due consideration to some exceptions depending on the type of the chemical compound we classify the following elements: uranium, thorium, transuranium and transplutonium elements, radionuclides of the sulphur subgroup (tellurium, polonium) as radionuclides with the low level of percutaneous resorbtion (to the tenths fractions of a per cent); radionuclides of the elements of the II<sup>nd</sup> group, uranium fission products as radionuclides with the medium level of percutaneous resorbtion (from several fractions of a per cent to one per cent) and radionuclides of the alkaline elements, the chrome subgroup, the VII<sup>th</sup> and VIII<sup>th</sup> groups of the periodic system as those with high level of percutaneous resorbtion (more than one per cent). At the same time the analysis of all the available data on the exchange of radioactive substances being applied onto the surface of the skin did not reveal any correlation between the levels of their accumulation in the skin and within the body.

4. Comparison of the peculiarities in the distribution within various organs and tissues of radionuclides of the elements of various groups of the Mendeleev periodic system depending on the route of their entering the body, allows one to conclude that the character of their distribution when they are applied on the skin is the same as in the case when they enter the gastro-intestinal tract.

Under the conditions of additional total body X-irradiation, the exchange of radioactive substances applied onto the skin varies insignificantly. Therefore, this factor should not be listed among those that may aggravate radiation effects on the living organism.

Table 1

The values of the parameters of the function characterizing the change in the concentration of  $\beta$ -radionuclides in the skin according to various depths

Parameter	$a_1$	$a_2$	$a_3$	$K_1$	$K_2$	$K_3$
Value	0.83	0.15	0.02	$6.93 \times 10^{-2}$	$1.73 \times 10^{-2}$	$2.56 \times 10^{-3}$

Table 2

The distribution of radionuclides in various skin fractions, %

Radionuclide	Fraction		
	soluble in lipids	soluble in water	residual
Mo <sup>99</sup>	14.5±2.5	29.6±6.4	55.9±8.0
Tl <sup>132</sup>	11.8±2.6	6.5±1.6	81.7±11.4
Sr <sup>89</sup>	10.3±2.7	14.4±4.3	75.3±17.2
Ba <sup>140</sup>	11.1±2.9	7.7±2.4	81.2±11.3
Y <sup>91</sup>	11.4±2.2	4.4±1.2	84.2±1.3
C <sup>141,144</sup>	12.5±0.5	3.8±0.2	83.7±0.2
Pr <sup>143</sup> +Nd <sup>147</sup>	5.8±0.7	1.9±0.1	92.3±0.7

Table 3

The kinetics of elimination from the skin of some radioactive substances

Radio-nuclide	compound	T <sub>eff</sub> <sub>1</sub> , hours	per cent	T <sub>eff</sub> <sub>2</sub> , days	per cent	Bibliography
H <sup>3</sup>	H <sub>2</sub> O	1	95	12	5	(10)
Cs <sup>137</sup>	CsCl	8	65	2	35	(10)
Pu <sup>239</sup>	Pu(NO <sub>3</sub> ) <sub>4</sub>	13	80	5	20	(10)
Po <sup>210</sup>	Po(NO <sub>3</sub> ) <sub>4</sub>	8	70	15	30	(5)

## References

1. Genaud P. Protection from radioactive elements. Translated from French. *Izd. inostr. literatury*, 1954, p. III.
2. Ilyin L.A. *Hyg. truda i prof. zabolevaniya* 3, 28, (1960).
3. Ilyin L.A., Norets T.A., Shvydko N.S., Ivanov E.V. Radioactive substances and skin. *Atomizdat*, 1972.
4. Osanov D.P. et al. *Health Phys.* 20, 559 (1971).
5. Ilyin L.A., Khodyreva M.A. et al. Sec. European Congress on Radiation Protection, 3-5 May 1972, Budapest. *Abstr.* 1972, p. 30.
6. Wadachi I. et al. *J. Atomic Energy Soc. Japan* 5, 938 (1963).
7. *Ibid.* 5, 994 (1963).
8. *Ibid.* 7, 942 (1965).
9. Tashiro S et al. *Radioisotopes*, 15, 224 (1966).
10. Osanov D.P. et al. *Radiobiologiya* 1, 154 (1971).

"DETERMINATION OF THE CONTENT AND KINETICS OF THE  
BEHAVIOUR OF SOME RADIONUCLIDES IN THE BODY OF  
MAN BY THE IN VIVO TECHNIQUE"

A.D. Turkin, V.P. Stolyarov, Yu.Ja. Sokolov

The Institute of Biophysics Ministry of Health USSR  
Moscow, USSR

Abstract

The regularities in accumulation, distribution and elimination of krypton-85, xenon-133 and iodine-131 from the body of man have been studied on volunteers. It is shown that intake of the inert radioactive gasses and gaseous iodine into the human body is primarily through the respiratory organs. The intact skin is of little importance in this process.

Были исследованы закономерности накопления, распределения и выведения из организма человека криптона-85, ксенона-133 и йода-131. [1]

Эксперименты проводились с привлечением добровольцев-мужчин в возрасте от 27 до 50 лет. Время контакта с криптоном-85 и ксеноном-133 составляло от 0,5 до 66 часов. Концентрация этих радиоактивных изотопов в герметичной камере объемом 3,1 м<sup>3</sup> составляла  $5 \cdot 10^{-8} + 10^{-8}$  кюри/л в зависимости от цели эксперимента. Экспериментальные исследования показали, что накопление и выведение криптона-85 и ксенона-133 из отдельных органов и тканей человека достаточно точно подчиняется экспоненциальному закону.

Следовательно, для расчета дозы при внутреннем облучении радиоактивными изотопами инертных газов можно использовать уравнение (1)

$$D = 0,59 \cdot E \cdot q \cdot Q \cdot R \cdot \frac{T^P}{T^P + T_{\text{ЭК}}^{\sigma}} \left\{ t_{\text{ЭК}} - \right.$$

$$\left. - \left[ 1 - \exp - \frac{0,693(T^P + T_{\text{ЭК}}^{\sigma})}{T^P + T_{\text{ЭК}}^{\sigma}} \cdot t_{\text{ЭК}} \right] \left[ \frac{T^P \cdot T_{\text{ЭК}}^{\sigma}}{0,693(T^P + T_{\text{ЭК}}^{\sigma})} - \frac{T^P \cdot T^{\sigma}}{0,693(T^P + T^{\sigma})} \right] \right.$$

$$\left. \cdot \left( 1 - \exp - \frac{0,693(T^P + T_{\text{ЭК}}^{\sigma})}{T^P + T^{\sigma}} \cdot t \right) \right\} \dots \dots \dots (1)$$

где:  $D$  - доза облучения, рады;  
 $E$  - энергия излучения, Мэв;  
 $q$  - фактор, учитывающий поглощение излучения;  
 $Q$  - концентрация нуклида в воздухе, кюри/л;  
 $R$  - коэффициент распределения нуклида в органе или ткани, л/кг;  
 $t_{\text{ЭК}}$  - время экспозиции в атмосфере нуклида;  
 $t$  - время после экспозиции;  
 $T^P$  - физический период полураспада нуклида;  
 $T_{\text{ЭК}}^{\sigma}$  и  $T^{\sigma}$  - биологические периоды полувыведения соответственно при экспозиции и после экспозиции.

Параметры, характеризующие накопление, распределение и выведение криптона и ксенона из организма человека, которые входят в уравнение (1), приведены в таблице 1.

Обращает на себя внимание тот факт, что скорость накопления инертных газов в организме человека превышает скорость их выведения. Это явление особенно четко прослеживается на криптоне-85, накопление которого в жировой ткани людей заканчивается практически через 4-5 часов, а выведение через 8-9 часов. Более высокие темпы накопления инертных газов в организме по сравнению с темпами выведения, по-видимому, можно объяснить неодинаковым перепадом концентраций инертных газов в крови и в насыщаемых тканях. В процессе поступления инертного газа в организм этот перепад концентраций всегда больше, чем при выведении. Объясняется это тем, что в начале контакта концентрация газа в насыщаемой ткани близка к нулю, тогда как в крови она достигает равновесного значения через несколько минут, вследствие интенсивного газового обмена в легких. В процессе же выведения, после того, как в насыщаемых тканях накопился газ, перепад между концентрациями будет незначительным из-за непрерывного перехода газа из ткани в кровь.

В таблице 2 приведены результаты расчетов по формуле (1) тканевых доз в теле человека при внутреннем облучении радиоактивными изотопами инертных газов. Было принято, что  $q = 1$ .

Таблица 1

Параметры, характеризующие накопление, распределение и выведение изотопа криптона и ксенона из организма человека

Органы и ткани	Изотопы криптона			Изотопы ксенона		
	$R_{д/кг}$	$T_{эк}^{\sigma}$	$T^{\sigma}$	$R_{д/кг}$	$T_{эк}^{\sigma}$	$T^{\sigma}$
Жировая ткань	0,46	1,4 часа	2,7 часа	1,4	5 часов	6,3 часа
Мышцы и другие ткани	0,047	8 мин	8 мин	0,13	0,4 часа	0,7 часа
Кровь	0,046	30 сек	30 сек	0,17	30 сек	30 сек
Легкие	2	30 сек	30 сек	2	30 сек	30 сек

Таблица 2

Тканевые дозы в теле человека, создаваемые радиоактивными изотопами криптона и ксенона при внутреннем облучении за 36 часовую рабочую неделю

	$D$ , рад/неделя при $a = 10^{-7}$ кюри/л			
	Легкие	Кровь	Мышцы	Жировая ткань
<sup>89</sup> Kr	0,27	0,006	0,007	0,6
<sup>85m</sup> Kr	3,6	0,09	0,09	0,7
<sup>85</sup> Kr	3,7	0,084	0,08	0,9
<sup>87</sup> Kr	20	0,45	0,4	2,2
<sup>89</sup> Kr	18	0,42	0,14	0,2
<sup>90</sup> Kr	19	0,45	0,03	0,03
<sup>91</sup> Kr	4,5	0,1	0,009	0,009
<sup>131m</sup> Xe	2,0	0,17	0,13	1,4
<sup>133m</sup> Xe	2,5	0,21	0,13	0,5
<sup>133</sup> Xe	2,1	0,18	0,14	1,5
<sup>135m</sup> Xe	1,0	0,085	0,02	0,03
<sup>135</sup> Xe	4,6	0,4	0,26	2,0
<sup>137</sup> Xe	18	1,5	0,12	0,16
<sup>138</sup> Xe	12	1,0	0,27	0,45

В опытах по изучению поступления ксенона-133 через кожные покровы участвовали трое добровольцев.

Испытуемый, тело которого не было защищено одеждой, помещался в экспозиционную камеру. После герметизации в камеру вводили ксенона-133. Органы дыхания испытуемого были изолированы от воздуха экспозиционной камеры с помощью плема противогаса, в

подмасочное пространство которого подавали чистый воздух для дыхания. Время экспозиции составляло 3 часа.

Установлено, что поступление ксенона-133 через кожные покровы составляет не более 0,4% по сравнению с поступлением через органы дыхания. В опытах, в которых делалась попытка определить выделение ксенона-133 из организма испытуемых через кожные покровы, количественных данных получить не удалось в силу незначительности эффекта. Проведенные опыты показывают, что поступление и выведение инертных радиоактивных газов из организма человека происходит в основном через органы дыхания. Неповрежденные кожные покровы в этом процессе играют исчезающе малую роль.

Аналогичные исследования [2] были проведены с газообразным радиоактивным йодом-131. Коэффициент  $F$  характеризует суммарную скорость поступления газообразного йода через органы дыхания ( $F_1$ ) и через кожные покровы ( $F_2$ ), т.е.

$$F = F_1 + F_2 \dots \dots \dots (2)$$

В свою очередь  $F_1 = \gamma \cdot Q \cdot V \dots \dots \dots (3)$ ;  $F_2 = \alpha \cdot Q \cdot S \dots \dots \dots (4)$

где:  $Q$  - концентрация йода в воздухе, кюри/л;  
 $V$  - скорость легочной вентиляции, л/час;  
 $S$  - площадь кожного покрова, м<sup>2</sup>;  
 $\gamma$  - доля активности, остающаяся в органах дыхания и переходящая в кровь;  
 $\alpha$  - коэффициент, характеризующий поступление йода через кожные покровы,  $\frac{\text{л}}{\text{м}^2 \cdot \text{час}}$

Определение параметров, входящих в уравнения (3) и (4), осуществлялось в опытах с привлечением добровольцев. Всего было проведено 23 опыта. Из них в 18 исследовалась резорбция йода неповрежденными кожными покровами и в 5 опытах изучалось поступление йода через органы дыхания.

Концентрация радиоактивного йода в экспозиционной камере поддерживалась с помощью генератора. Аэрозольная фаза и пары элементарного йода улавливались фильтрами. В воздушное пространство камеры попадали только газообразные соединения йода. Систематический контроль концентрации йода-131 в камере давал возможность при необходимости корректировать в ходе опыта ее значение путем изменения скорости подачи из генератора. Колебание концентрации йода-131 в воздухе экспозиционной камеры не превышало  $\pm 20\%$  от среднего значения.

Результаты исследований представлены в таблице 3. Как видно из таблицы 3, коэффициент задержки газообразного йода в органах дыхания у разных испытуемых отличается на небольшую величину, тогда как резорбция газообразного йода кожными покровами колеблется в 5 раз ( $\alpha$  меняется от 1,5 до 7,3). Разброс скорости поступления газообразного йода через кожные покровы нельзя объяснить неодинаковыми исходными значениями концентрации йода-131 в воздухе, так как ее колебание в 100 раз не привело к заметному изменению величины коэффициента  $\alpha$  у испытуемого К.А.

Все остальные параметры в процессе эксперимента (температура, влажность, режим питания и др.) были также одинаковыми. По-видимому, колебания скорости поступления газообразного йода через кожные покровы можно объяснить в основном индивидуальными фи-



Таблица 3

Основные параметры, характеризующие поступление газообразного йода в организм человека через органы дыхания и неповрежденные кожные покровы

Испы- туе- мый	Эффективный пе- риод полувыве- дения йода-131 из крови, $T_1$	Эффективный пе- риод полувыве- дения йода-131 из щитовидной железы $T$	Коэффициент за- держки йода в органах ды- хания $\lambda$	Доля активности йода-131, пос- тупившая из крови в щитовидную же- лезу $a_1$	Коэффициент пос- тупления газооб- разного йода через неповрежденные кожные покровы, $\alpha$ , л/м <sup>2</sup> , час
С.А.	6,2 часа	6,2 дня	0,75	9,1%	5,1 4,8 <sup>■</sup>
П.С.	-	-	-	-	2,5 <sup>x</sup> 4,5 <sup>■</sup>
К.А.	5,3 часа	6,4 дня	0,65	5,0%	2,1 2,1 2,6
К.В.	-	-	-	-	2,1 <sup>x</sup> 2,7 <sup>x</sup> 2,9 <sup>■</sup>
А.В.	4,7 часа	6,0 дня	0,58	8,0%	2,7 1,5 2,0 <sup>■</sup>
Ю.М.	4,3 часа	6,0 дня	0,8	3,7%	7,3
С.Ю.	-	-	-	-	5,1 <sup>x</sup>
Ф.В.	6,0 часа	7,3	0,67	10,6%	-

$$\bar{T}_1 = 5,3 \pm 0,8 \text{ часа}; \quad \bar{T} = 6,4 \pm 0,5 \text{ дня}; \quad \bar{\lambda} = 0,7 \pm 0,08; \quad \bar{a}_1 = 7,2 \pm 2,9\%; \quad \bar{\alpha} = 3,8 \pm 2,1$$

ПРИМЕЧАНИЕ: x) - рассчитано при  $\bar{a}_1 = 7,2\%$ ; ■ - определено с физической нагрузкой.

физиологическими особенностями испытуемых. В опытах с нагрузкой средней тяжести значения коэффициентов  $\alpha$  практически не отличаются от значений коэффициентов при спокойном состоянии испытуемых. Для некоторых испытуемых была оценена относительная величина скорости поступления газообразного йода через органы дыхания и через неповрежденные кожные покровы

$F_1/F_2$	127	47	40	80
Испытуемый	К.А.	С.А.	Ю.М.	А.В.

Из приведенных данных видно, что резорбция газообразного йода кожными покровами составляет  $1 \pm 2\%$  по сравнению с резорбцией органами дыхания.

Можно отметить, что резорбтивная способность кожных покровов человека по отношению к газообразному йоду выше чем к ксенону, проникновение которого через кожные покровы составляет не более 0,4%. Качественно такое различие в проницаемости кожи по отношению к газообразному йоду и ксенону можно объяснить различием механизма проникновения этих изотопов. Несмотря на то, что в настоящее время не существует единого мнения относительно механизма кожной проницаемости, но, вместе с тем, по мнению авторов [3] поступление радиоактивных веществ характеризуется следующими тремя процессами.

1. Активный физиологический процесс всасывания.
2. Процесс диффузии через кожный барьер.
3. Способность кожи, как губки, впитывать в себя и удерживать вещества, находящиеся на ее поверхности.

Можно однозначно утверждать, что инертный ксенон проникает через кожный барьер вследствие диффузии, тогда как газообразный йод вероятнее всего поступает через кожу за счет трех выше перечисленных процессов.

В связи с тем, что исследования основных параметров, характеризующих поступление газообразного йода в организм человека, проводились в лабораторных условиях на соединениях йода неизвестного физико-химического состава, представляется очень важным оценить пригодность полученных данных к реальным условиям.

В настоящее время определение физико-химического состава парообразных соединений йода представляет самостоятельную проблему, решение которой сопряжено с большими трудностями. Однако с точки зрения радиационной опасности даже при известном физико-химическом составе газообразного йода в воздухе определяющим параметром является величина радиоактивности, накопленная в организме. Эксперименты по определению коэффициента задержки газообразного йода в реальных условиях на водо-водяном реакторе и на радиохимическом заводе представлены в таблице 4 [2].

Несколько повышенное значение коэффициента задержки в реальных условиях можно объяснить присутствием в воздухе элементарного йода и йода в виде аэрозолей, которые, по-видимому, в органах дыхания задерживаются с более высокой эффективностью, чем парообразные соединения. Коэффициент задержки в органах дыхания человека газообразного йода лежит в пределах 0,7 - 0,9, что подтверждает принятое в рекомендациях МКРЗ значение этого коэффициента, равное 0,75.

Таблица 4

Значения коэффициентов задержки газообразного радиоактивного йода в органах дыхания человека

Место эксперимента	Формы йода в воздухе, %			Коэффициент задержки
	Аэрозоли	Элементарный йод	Летучие соединения	
Лаборат.исслед.	-	-	100	0,7
Водо-водяной реактор	3-21	9-16	70-86	0,84
Радиохимический завод	3	5-10	90-95	0,9

## ЛИТЕРАТУРА

1. Туркия А.Д.  
Дозиметрия радиоактивных газов. Атомиздат, 1973 год.
2. Городинский С.М. и др.  
Поступление парообразных соединений йода в организм человека через органы дыхания.  
Статья в сб. "Распределение, кинетика обмена и биологическое действие радиоактивных изотопов йода".  
Из-во "Медицина", г. Москва, 1970 г. стр. 45-50.
3. Осанов Д.П., Лихтарев И.А., Радзиевский Г.Б.  
Дозиметрия излучений инкорпорированных радиоактивных веществ.  
Атомиздат, 1970 год.

RETENTION AND DISTRIBUTION OF INORGANIC MERCURY (  $^{197}\text{Hg}$ ,  $^{203}\text{Hg}$  )  
IN THE HUMAN BODY AFTER SINGLE INHALATION

Y. IZUMI, M. FUJITA, A. YABE and T. HATTORI

Japan Atomic Energy Research Institute,  
Tokai-mura, Ibaraki-ken, Japan

ABSTRACT- Inorganic mercury ( $^{197}\text{Hg}$ ,  $^{203}\text{Hg}$ ) was inhaled by two workers during decontamination procedures in a hot cell. The retention and distribution of the mercury in the body were studied with a whole-body counter. The biological half-lives in the whole body were about 33 days, which was shorter than those reported for organic mercuries. The biological half-lives in the upper abdomen were about 50 days.

Scanning along the body axis and counting at several different points above the upper abdomen revealed that the mercury deposited mainly in the kidneys and in the liver. The activities of the mercury-203 in the kidneys and in the liver were determined using the counting efficiencies obtained from a REMAD phantom. The total activities in the kidneys 10 days after inhalation were almost equal to those in the liver. The high concentration in the kidneys, about 5 times as high as that in the liver, suggested that the kidneys were the critical organ for the inhalation of inorganic mercury.

INTRODUCTION

A solution of radioactive mercury-197 (2 ml, 100 mCi,  $\text{Hg}(\text{NO}_3)_2$ ) was spilled in a hot cell and the decontamination was carried out by four workers with full face masks and protection clothes. At first, it was thought that no inhalation of the radioactive mercury-197 had occurred in the human subjects. However, to make sure of it, the subjects were monitored with a whole-body counter on the 10th day after the decontamination procedures. It was found that two out of the four subjects were contaminated with radioactive  $^{197}\text{Hg}$  and  $^{203}\text{Hg}$ . Therefore, to assess the radiation doses of the radioactive mercuries to the critical organ, the distribution in the body and the effective half-lives of the radioactive mercuries were studied with the whole-body counter.

METHOD

Subject, contaminant and whole-body counter

The age, height and weight of the contaminated workers are given in Table 1. Their body builds were not so different from that of a typical Japanese. The whole-body counter used in this study consisted of a NaI(Tl) crystal of 8 inch  $\phi$  x 4 inch and a 400 channel pulse height analyser. The monitoring room was shielded with 200 mm Fe + 3 mm Pb.<sup>1</sup> The measurements of the radioactivities within the body were carried out by three different geometries, i.e., 1) by standard chair geometry, 2) by placing the detector above the upper abdomen of the human subject who lay on a bed in a supine position (see Fig. 1) or in a prone position and fixing the distance from the detector surface to the bed as 22.5 cm and 3) by scanning along the body axis. In the case of scanning, the detector was provided with a collimator having a 5 cm slit, and the distance from the detector to the bed was kept as 42.5 cm.

Determination of counting efficiencies for  $^{203}\text{Hg}$  in the kidneys and liver

Table 1. Age, height and weight of the contaminated workers

Subject	Age	Height (cm)	Weight (kg)
A	29	160	53
B	32	169	50

The counting efficiencies for  $^{203}\text{Hg}$  were obtained using a REMAB phantom (Alderson Research Lab. U.S.A.). The volume of the right kidney of the phantom was 90 ml and that of the left was 110 ml and the total volume was 200 ml. The volume of the liver of the phantom was 1300 ml. The average weight of the kidneys of adults of Japanese is 270 g and that of the liver, 1440 g.<sup>2</sup> Therefore, there were some discrepancies in the organ sizes between the phantom and a typical Japanese.

The vessels for the kidneys and liver of the phantom were filled with a standard solution of  $^{203}\text{Hg}$ . The solution of  $^{203}\text{Hg}$  was prepared as follows. Mercury-203 was dissolved in a solution containing  $\text{HgCl}_2$  (2 mg) +  $\text{KClO}_3$  (7 mg) per ml of 3N HCl. The activity of  $^{203}\text{Hg}$  in the solution was 15.84  $\mu\text{Ci/ml}$ . One ml of this solution was divided into two kidney vessels in proportion to their volumes (i.e., right, 0.45 ml, left, 0.55 ml). The radioactive solution was diluted until 200 ml for both the vessels with a diluting solution containing  $\text{NaCl}$  (10 mg) +  $\text{HgCl}_2$  (2 mg) per ml of 0.5N HCl. The vessel for the liver also was filled with the same radioactive solution and diluting solution as those for the kidneys.

On the condition that the NaI(Tl) detector was placed above the central part of the upper abdomen and the distance from the detector to the bed was fixed as 22.5 cm, the counting efficiencies for the  $^{203}\text{Hg}$  in the kidneys and liver were determined in the supine and prone positions. They were as follows.

$$\begin{aligned} \eta(k)_s &= 0.013 \text{ (cpm/dpm) for } ^{203}\text{Hg} \text{ in the kidneys in the supine position,} \\ \eta(k)_p &= 0.065 \text{ (cpm/dpm) for } ^{203}\text{Hg} \text{ in the kidneys in the prone position,} \\ \eta(l)_s &= 0.054 \text{ (cpm/dpm) for } ^{203}\text{Hg} \text{ in the liver in the supine position,} \\ \eta(l)_p &= 0.012 \text{ (cpm/dpm) for } ^{203}\text{Hg} \text{ in the liver in the prone position.} \end{aligned}$$

## RESULTS

### Determination of effective half-life of mercury in the whole body

Since it was difficult to determine the counting efficiencies for the radioactive mercuries in the whole body, the radioactivities in  $\mu\text{Ci}$  in the whole body were not determined. Only net counting rates in the energy ranges of  $77 \pm 43$  keV (for  $^{197}\text{Hg}$ ) and  $279 \pm 59$  keV (for  $^{203}\text{Hg}$ ) were measured by standard chair method from the 10th to 36th day after inhalation. To obtain the net counting rates, the contribution of the  $^{40}\text{K}$  and  $^{137}\text{Cs}$  in the body to the energy ranges of  $^{197}\text{Hg}$  and  $^{203}\text{Hg}$  was subtracted. Also, the contribution of  $^{203}\text{Hg}$  to the counting rate in the energy range of  $^{197}\text{Hg}$  was subtracted. The contribution was estimated to be 0.81 (for subject A) and 0.83 (for subject B) times the counting rates in the photopeak of  $^{203}\text{Hg}$ . These values were obtained after 29th day postinhalation when the  $^{197}\text{Hg}$  had decayed sufficiently.

The net counting rates of the  $^{197}\text{Hg}$  and  $^{203}\text{Hg}$  in the whole body as determined by standard chair geometry are plotted against time in Fig.2. The effective half-lives were obtained by the least square method. The effective and biological half-lives of  $^{203}\text{Hg}$  are listed in Table 2.

### Distribution of radioactive mercury in the body

The distribution of radioactive mercuries in the body was estimated by scanning along the central body axis with the detector provided with the collimator. The energy range used in this scanning was from 34 to 338 keV to include the energies of  $^{197}\text{Hg}$  and  $^{203}\text{Hg}$ . The result of scanning on the 10th day after inhalation for subject A is illustrated by a solid line in Fig.3. It was found that the radioactive mercuries deposited mainly in the upper abdomen, but somewhat in the chest, in the lower abdomen and in the other parts of the body. On

Table 2. Effective and biological half-lives of  $^{203}\text{Hg}$  in the body

Subject	Effective half-life in		Biological half-life in	
	whole body (day)	upper abdomen (day)	whole body (day)	upper abdomen (day)
A	19	23	32	45
B	20	24	35	49

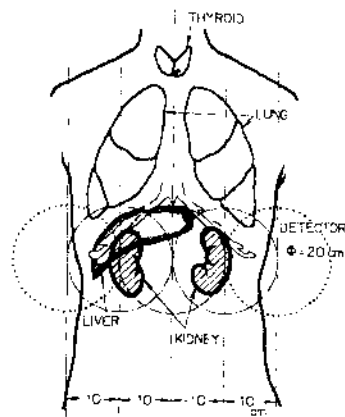


Fig.1. Relative position of detector and the kidneys and liver.

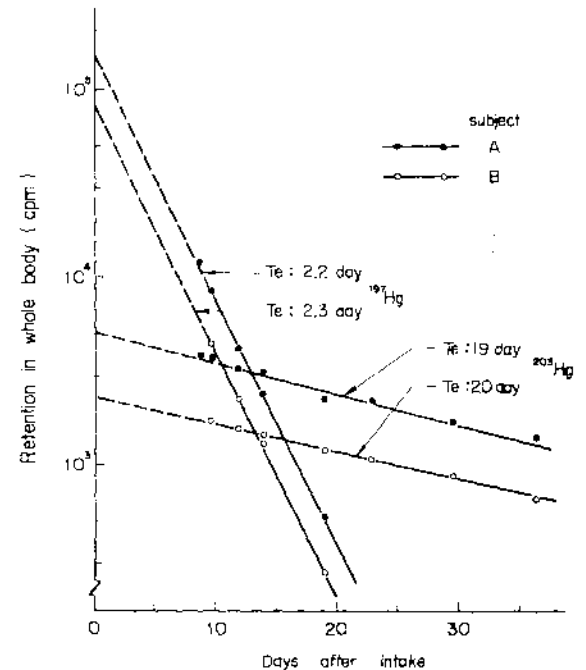


Fig.2. Retention of  $^{197}\text{Hg}$  and  $^{203}\text{Hg}$  in the whole body after single intake. (Te : effective half-life)

the assumption that the counting efficiencies were the same for any points along the body axis, it was calculated that about 60 per cent of the mercuries in the total body deposited in the upper abdomen. To know the organ in which the radioactive mercuries in the upper abdomen mainly deposited, the position of the detector (without collimator) was moved above the upper abdomen from the right to the left perpendicularly to the body axis at 10 cm intervals in the supine position (see Fig.1). The counting rates at each position decreased in the following order; center > 10 cm right > 10 cm left > 20 cm right > 20 cm left. This result supported that the mercuries deposited mainly in the kidneys and liver.

Determination of effective half-life of mercury in the upper abdomen

The human subjects lay on the bed in a supine position and the distance between the NaI(Tl) detector and the bed was fixed as 22.5 cm as usual. The detector was placed above the center of the upper abdomen (see Fig.1). On these conditions, the mercuries deposited in the upper abdomen would be detected without much error, because the mercuries in the other parts of the body were relatively small in quantity as shown in the above paragraph for distribution.

Here, again, the net counting rates of the  $^{197}\text{Hg}$  and  $^{203}\text{Hg}$  were obtained by the same procedures that taken for the mercuries in the whole body. And, the

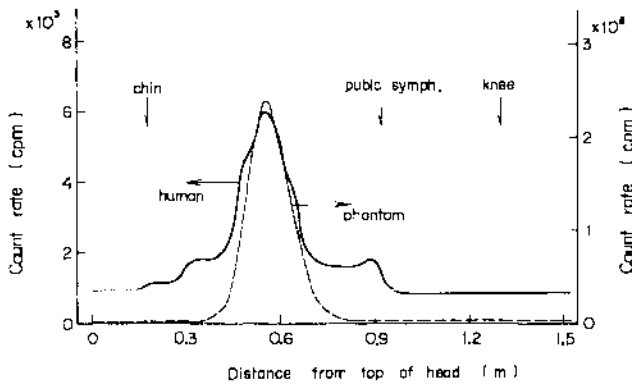


Fig. 3. Profile curves from human subject and phantom.

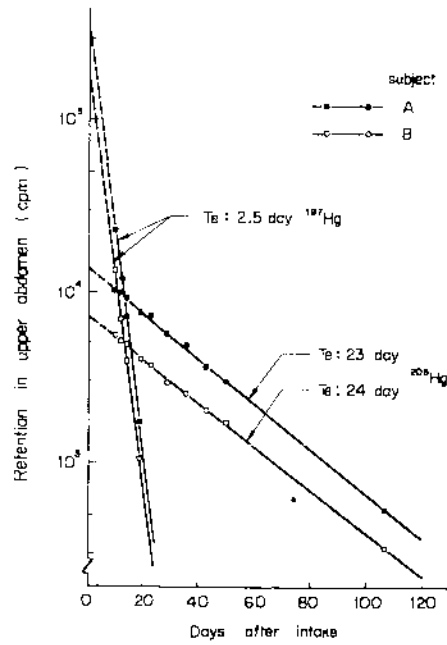


Fig. 4. Retention of  $^{197}\text{Hg}$  and  $^{203}\text{Hg}$  in the upper abdomen.

contribution of the  $^{203}\text{Hg}$  to the counting rates in the energy range of  $^{197}\text{Hg}$  were 0.72 (for subject A) and 0.68 (for subject B) times the counting rates in the  $^{203}\text{Hg}$  photopeak.

The change with time of the net counting rates of the  $^{197}\text{Hg}$  and  $^{203}\text{Hg}$  in the upper abdomen is shown in Fig. 4. The effective and biological half-lives of  $^{203}\text{Hg}$  are listed in Table 2, showing that the half-lives in the upper abdomen were longer than those in the whole body.

#### Determination of the radioactivities of $^{203}\text{Hg}$ in the kidneys and liver

It was possible to determine the radioactivities of  $^{203}\text{Hg}$  in the kidneys and in the liver from the measurements of the human subject lying on the bed in the prone and supine positions.

If no mercury is involved in the body except for the kidneys and liver, the counting rates in the prone position,  $P(\text{cpm})$ , and those in the supine position,  $S(\text{cpm})$ , are given by the following formulas,

$$P = C E \{ \eta(k)_p K + \eta(l)_p L \}$$

$$S = C E \{ \eta(k)_s K + \eta(l)_s L \}$$

where  $C : 2.22 \cdot 10^6$  (dpm/ $\mu\text{Ci}$ ),

$E : 0.83$ , emission rate of  $\gamma$ -rays of 279 keV per disintegration of  $^{203}\text{Hg}$ ,

$K$  : organ burden in  $\mu\text{Ci}$  of  $^{203}\text{Hg}$  in the kidneys, and

$L$  : organ burden in  $\mu\text{Ci}$  of  $^{203}\text{Hg}$  in the liver.

As described already, on the condition that the distance from the detector to the bed was fixed as 22.5 cm,

$\eta(k)_p$  and  $\eta(k)_s$  : 0.065 and 0.013 (cpm/dpm), respectively, and

$\eta(l)_p$  and  $\eta(l)_s$  : 0.012 and 0.054 (cpm/dpm), respectively.

From the above formulas,

$$L (\mu\text{Ci}) = \frac{1}{CE} (19.6 S - 4.08 P)$$

$$K (\mu\text{Ci}) = \frac{S}{0.0135 CE} - 3.96 L .$$

Therefore, it was possible to evaluate the activities in the kidneys and in the liver from the counting rates in the prone and supine positions. The activities in these organs on the 10th day and 50th day after inhalation were calculated

from the counting rates on those days. The results are given in Table 3. The table shows that on the 10th day the ratio of the activities in the kidneys to those in the liver was about 1 : 1 in both the subjects. If the subjects had the kidneys of 270 g and the liver of 1440 g like a typical Japanese, the concentration of  $^{203}\text{Hg}$  in the kidneys must be about 5 times as high as that in the liver on that date, suggesting that the kidneys were the critical organ for the inhalation of inorganic mercuries.

Table 3. Radioactivity of  $^{203}\text{Hg}$  in the kidneys and liver on the 10th and 50th day after inhalation

Subject	Organ	Activity ( $\mu\text{Ci}$ ) on day		Ratio of activities (kidney/liver) on day	
		10th	50th	10th	50th
A	kidney	0.079	0.028	0.97	1.17
	liver	0.082	0.024		
B	kidney	0.045	0.018	1.00	1.34
	liver	0.045	0.013		

### DISCUSSION

#### Biological half-life

Table 4 summarizes the effective and biological half-lives of  $^{203}\text{Hg}$  reported by several workers for the whole body. <sup>3-7</sup> These data suggest that the biological half-lives of inorganic mercuries are shorter than those of organic ones, and the retention function of mercury should be expressed by three components.

Table 4. Comparison of effective and biological half-lives in the whole body. (Figures in parentheses show biological half-lives)

Chemical form of Hg	Route of entry	Effective and biological half-life			Author
		fast component (day)	intermediate component (day)	slow component (day)	
inorganic	inhalation	-	-	20, 19 (35) (32)	this study
inorganic	oral	-	-	22 (42+3)	Rahola et al.
$^{203}\text{Hg}$ -methyl mercury	oral	-	-	29 (76±3)	Rahola et al.
$^{203}\text{Hg}$ -neoohydrin	oral	-	-	30 (84)	Johnson et al.
$^{203}\text{Hg}$ -neoohydrin	oral	0.22 (0.22)	7 (8.2)	-	Greenlaw et al.
monomethyl $^{203}\text{Hg}$ nitrate	oral	-	-	28, 27 (71) (66)	Falk et al.
			8.2 (10)		ICRP <sup>7</sup>

The present study showed that the mercuries in the upper abdomen have longer half-lives than those in the whole body. This is consistent with the finding by Falk et al. that the monomethyl- $^{203}\text{Hg}$  nitrate deposited in the liver region decreased more slowly than those in the other regions of the body.

#### Distribution

Falk et al. have studied the distribution of the monomethyl- $^{203}\text{Hg}$  nitrate



in the body by scanning along the body axis. Their profile curves of the net counts are very similar to ours (Fig.3) as a whole. Their conclusion was that the mercury mainly accumulated in the liver region and somewhat in the cerebellum region. Our observation could not reveal special accumulation in the cerebellum, but this may be attributed to the poor resolution of the detector used in this study and/or to the difference of the chemical form of the mercury absorbed. The present study suggests that the kidneys are the most important organ where mercury deposits in the highest concentration. Falk et al. did not refer to the deposition in the kidneys.

#### Assessment of dose commitment due to the radioactive mercuries in the kidneys

Unfortunately, in this study the retention function of the  $^{203}\text{Hg}$  in the kidneys could not be accurately estimated, for the measurements of the  $^{203}\text{Hg}$  started on the 10th day after inhalation, and the fast and intermediate components of the retention were missed. However, it was probable that the dose commitment to the kidneys due to the fast and intermediate components would not be greater than that due to the slow component, as was estimated by Johnson et al. for the dose commitment to the whole body.

The dose commitment to the kidneys delivered by the slow component of the retention of  $^{203}\text{Hg}$  were estimated as 104 and 57 mrem for subject A and B, respectively, and those to the liver, 21 and 12 mrem, respectively, on the assumption that the slow component of the retention of  $^{203}\text{Hg}$  in the kidneys and in the liver had the same effective half-lives as those observed for the upper abdomen.

Since the radioactivity of  $^{197}\text{Hg}$  in the body was not determined, the calculation of the dose commitment due to the  $^{197}\text{Hg}$  was impossible, but the dose commitment to the kidneys and liver were perhaps the same order of magnitude as those due to the  $^{203}\text{Hg}$ , inferring from the effective energies, the counting rates and their effective half-lives in the upper abdomen (see Fig.4). Anyway, the sum of the dose commitment due to the  $^{197}\text{Hg}$  and  $^{203}\text{Hg}$  were far less than 8 rem, the ICRP permissible dose for 3 months.<sup>8</sup>

#### Error in the determination of radioactivities in the kidneys and liver

Some error may be introduced in the estimation of the radioactivities of  $^{203}\text{Hg}$  in the kidneys and liver, because the estimation was made on the incorrect assumption that no radioactive mercury was involved in the organs and tissues other than the kidneys and liver. However, as the deposition in the organs and tissues other than the kidneys and liver was not large as shown in Fig.3, the error would not be serious.

#### REFERENCES

1. Directory of whole-body radioactivity monitors, JA 1.1. IAEA, Vienna (1970).
2. S.Aimi, A.Yasoshima, M.Sugai, B.Sato, T.Sakai and Y.Nakajima, *Acta Pathologica*, 2, 173-200 (1952).
3. T.Rahala, R.Aaran and L.K.Miettinen, Assessment of radioactive contamination in man, pp.553-562, IAEA, Vienna (1972).
4. J.E.Johnson and J.A.Johnson, *Health Physics* 14, 265-266 (1968).
5. R.H.Greenlaw and C.M.Guaife, *Radiology* 78, 970-973 (1962).
6. R.Falk, J.O.Snihs, L.Ekman, U.Greitz and B.Aberg, *Acta Radiologica* 9, 55-72 (1970).
7. ICRP publication 2, Report of ICRP committee II on permissible dose for internal radiation, Pergamon Press, Oxford (1959).
8. ICRP publication 9, Recommendations of the International Commission on Radiological Protection, Pergamon Press, Oxford (1966).

A CASE STUDY OF HUMAN CONTAMINATION DUE TO  
INHALED THULIUM-170 OXIDE

A. Yabe, H. Yamagishi, H. Fukuda,  
Y. Takahashi, S. Mizushita  
and T. Hattori  
Japan Atomic Energy Research Institute,  
Tokai, Ibaraki, Japan

Abstract

One worker incidentally inhaled submicron particles of thulium-170 oxide. Data obtained by in-vivo counting and bioassay over about 450 days after inhalation fitted to a sum of three exponential terms having effective half-lives of about 3, 23 and 90 days, respectively, though the half-life of 3 days was not distinctly determined. Attempts were made to express the daily fecal and urinary excretion by power function and to relate the excretion with the decrease of the chest burden. In addition, the distribution and transfer of thulium in the body were discussed.

1. Introduction

An incident occurred in the course of the sealing of thulium-170 sources for non destructive test in a hot laboratory at Oarai Laboratory, Japan Atomic Energy Research Institute (JAERI). The process of the sealing which involved the mounting of a neutron activated thulium oxide pellet (about 50 Ci of  $^{170}\text{Tm}$ ) in a titanium metal capsule (6 mm  $\phi$  x 11 mm) and subsequent arc welding (3,000°C, in argon gas), was carried out in a hot cell by one worker. Following the welding process, the worker stepped in the cell and stayed there for about 5 minutes so as to carry out the sources from the cell. Leaving the cell he made a contamination check and found his hands and clothing contaminated in some measure. Then, he felt a doubt whether he had inhaled air born particles of the thulium oxide. Any air monitor was not running throughout the time of the work because it was early in the morning.

Unfortunately, four days later the worker was sent to Tokai Laboratory, JAERI, where a whole-body counter was installed. By the first measurement it was found that an appreciable amount of  $^{170}\text{Tm}$  was deposited in his chest. Though the exposure was not itself very serious, to obtain as much information as possible the determination of  $^{170}\text{Tm}$  by in-vivo counting and bioassay was followed until the levels of the activity were very low.

2. Methods

2. 1. In-vivo counting

Measurement was made to determine the deposition of  $^{170}\text{Tm}$  in the chest of the contaminated subject with a whole-body counter.<sup>1</sup> The other series of measurements with this counter were carried out to know the distribution of  $^{170}\text{Tm}$  in the body. The whole-body counter consisted of a 8 in.  $\phi$  x 4 in. NaI(Tl) detector in a cubical steel room, a multichannel analyzer and associated electronics.

In chest counting, the 8 in.  $\phi$  x 4 in. NaI(Tl) detector was placed above the chest at the distance of 22 cm from the bed on which the subject was lying on the back. In this case, the xyphoid sternum was adjusted to an edge of the crystal. Occasionally the position of the subject on bed was replaced with a prone position, to obtain the distribution of  $^{170}\text{Tm}$  in the chest.

In profile counting, the crystal provided with a 5 cm thick lead collimator having a 5 cm slit, was moved manually along the body axis to four fixed positions, that is, above the head, chest, lower abdomen and the thigh, keeping the distance from the crystal to the bed at 42 cm.

All measurements were carried out, using the energy band from 25 to 90 keV which contains the X- and gamma-ray lines of  $^{170}\text{Tm}$  (52, 84 keV).<sup>2,3,4</sup>

In order to obtain the counting efficiency, calibrating measurement with a RANDO phantom (Alderson Research Lab., USA) was made using 149  $^{170}\text{Tm}$  sources in small polyethylene capsules, the total activity of which was 0.40  $\mu\text{Ci}$ . These capsules were inserted in hole grids within the lungs of the phantom. The arrangement of capsules simulated the uniform distribution of the contaminant in lungs. Therefore, it might differ from the truth and some systematic error might exist.

The lung size of the subject was evaluated by means of radiograph techniques, and the effective tissue thickness of the subject's chest wall was estimated according to the method which was described in detail else where.<sup>5,6</sup> As the result, both data of the subject were somewhat larger than those of the phantom, and it was concluded that the counting efficiencies observed on the phantom required 10 per cent corrections.

## 2. 2. Bioassay

After the detection of  $^{170}\text{Tm}$  with the in-vivo counter, the subject was asked to collect the samples of urine and feces. Consequently, the sampling program was not started until 4 days after the inhalation. Both programs of the urine and fecal sampling were initiated on the 24-h sampling basis but afterward changed on the 2 or 4 consecutive day sampling basis. These procedures of sampling were continued until 134 days. After a long interval, additional samples of urine and feces were obtained for three days from 445 to 447 days.

The thulium-170 contents of the samples were determined by beta counting, for which a gas flow proportional counter was used. The counter had a detection limit of about 3 pCi. For the counting the simplest procedure was adopted as follows. In urine samples the radioactive thulium was coprecipitated with basic calcium phosphate after adding a thulium carrier. The resulting precipitates were dried, powdered and prepared for counting. Feces was ashed in a furnace before counting.

## 3. Results

### 3. 1. Retention in the chest and distribution in the whole-body

Measurement for  $^{170}\text{Tm}$  in the chest of the subject with a whole-body counter was carried out over the period from 4 to 447 days after the inhalation. The results of twenty-four measurements are plotted in Fig. 1(a). A least squares best fit analysis by a computer was tried on the plots to fit the data to a sum of exponentials. A good fit to a sum of two exponentials was obtained as shown in Fig. 1(a) with effective half-lives of  $23 \pm 4$  days and  $90 \pm 11$  days.

Location of  $^{170}\text{Tm}$  in the whole-body was roughly measured with a whole-body counter by means of the manual scan technique described above. The counting was carried out 9, 73 and 126 days after inhalation. Since it was difficult to determine the activity in the four different parts of the body, i.e., the head, chest, lower abdomen and the thigh, the deposition of  $^{170}\text{Tm}$  in those parts of the body was expressed in terms of counting rate. The counting done on the 3 different days showed that the deposition in the chest predominated over the rest of the body, but the tendency of the decrease of the counting rates at the four different parts of the body suggested the redistribution in the body at the

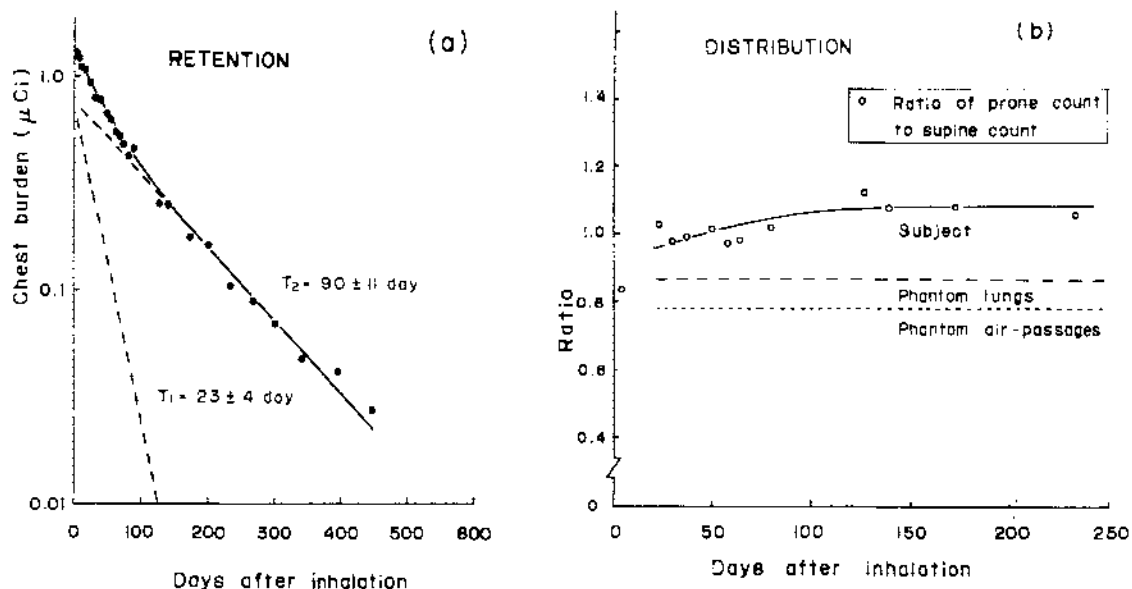


Fig.1. Retention (a) and Distribution (b) of thulium-170 in chest.

late stage of contamination (126 days).

### 3. 2. Distribution in the chest

To get an idea of detailed distribution of  $^{170}\text{Tm}$  in the chest, the subject was measured both in a prone and in a supine positions by a fixed crystal-bed geometry. Twelve measurements were made over the period from 4 to 232 days after inhalation. The ratios of prone counts to supine counts were calculated. In Fig. 1(b) the ratios are plotted against time. A fitted-curve is shown by a solid line.

In a geometry similar to that taken for the human subject the phantom was measured in a prone and supine positions two times each; one, with  $^{170}\text{Tm}$  sources in the lungs, and the other, with the sources in the lower air-passages below the throat. In the latter, the thulium-170 sources were simulated to the deposition in the trachea and the bronchia. The ratios of the counting in both positions were 0.87 and 0.78 for the sources in the lungs and in the lower air-passages, respectively, as shown in Fig. 1(b) by a broken line and by a dotted line. The ratios obtained from the phantom were lower than any ratios of the subject, which were between 1.0 and 1.1, except for 0.84 at the first measurement. The ratio for  $^{170}\text{Tm}$  in the lungs (0.87) was higher than that for  $^{170}\text{Tm}$  in the lower air-passages (0.78).

From these results, it is presumed that the minimum ratio of the subject at the first measurement (0.84) might indicate the initial deposition of  $^{170}\text{Tm}$  in the tracheo-bronchial parts of the air-passages. The rather high ratio of the subject over a period of observation as compared with that of the phantom seemed to be explained by 1) the difference of geometry due to the size of the body and due to inhomogeneous deposition in the chest and by 2) the difference of the absorption and scattering of the photons through the media, i.e. the lungs, adjacent organs, surrounding soft tissue, rib cage, etc.<sup>7</sup>

### 3. 3. Urinary, fecal and total excretion

Forty-seven 24-h urine samples were subjected to analysis until 134 days after inhalation. The results obtained through beta counting are shown in Fig. 2(a) where the urinary excretion rates in  $\mu\text{Ci}/\text{d}$  are plotted semi-logarithmically against time in days after inhalation with the maximum value of 1.6  $\mu\text{Ci}/\text{d}$ .

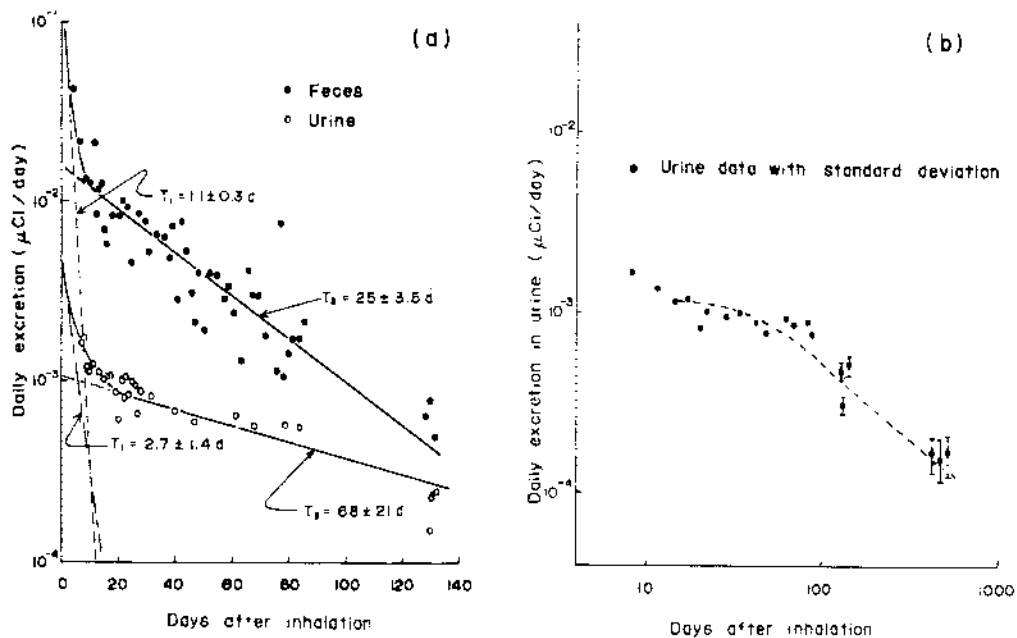


Fig.2. Excretion data of thulium-170, plotted semi-logarithmically (a) and logarithmically (b).

In this figure thirty plots from 47 measurements were taken to illustrate the excretion. With the aid of creatinine determination the samples with insufficient contents were excluded from the plots. A least squares best fit to a sum of two exponentials was obtained as shown in Fig. 2(a) with effective half-lives of  $2.7 \pm 1.4$  days and  $68 \pm 21$  days.

In order to see the long-term trend of urinary excretion, the data of the sample taken during the period from 445 to 447 days added to the graph of urinary excretion rates on a log-log scale. This is shown in Fig. 2(b). The plots in this figure are shown along with a curve which is arbitrarily drawn to show the long-term pattern. This pattern of urinary excretion was similar to those calculated from the radionuclide in the lungs, when the biological half-lives in the lungs were 30 and 300 days (these correspond to effective half-lives of 23 and 90 days shown in Fig. 1(a), respectively) and the exponent of the power function of urinary excretion was assumed to be  $-1.0 \sim -1.7$ . This calculation method is shown in the ICRP Publication 10A.<sup>8</sup>

During the 134 days of observation, there were 49 fecal samples in total. Over the period from 4 to 85 days all outputs of feces except one were collected. The results of determination are presented semi-logarithmically in Fig. 2(a) with the maximum value of 43 nCi/d. A good fit to a sum of two exponentials was again obtained with the effective half-lives of  $1.1 \pm 0.3$  days and  $25 \pm 3.5$  days.

The total activity of  $^{170}\text{Tm}$  excreted daily in feces and urine was obtained by combining the fecal and urinary data. In this case the daily feces and the total of a single output collected on the same day as 24-h urine samples, were combined together. Here again a good fit to a sum of two exponentials was obtained. The exponentials had half-lives of about 5.4 days and about 31 days, respectively. Since there was no data for the time earlier than 7 days after inhalation, the estimated half-life for the rapid component of excretion (5.4 days) might have some ambiguity.

The daily excretion of  $^{170}\text{Tm}$  in feces collected from 4 to 134 days after inhalation was consistently higher than that in urine collected for the same period. The ratio of urinary to total excretion changed from 0.06 to 0.3 during the above observation period. Whether this ratio changed with time was tested by

regression analysis. The coefficient of correlation obtained was 0.86 and this correlation was significant at the 1 per cent level (linear correlation was supposed until 134 days).

#### 4. Discussion

##### 4. 1. Half-lives

It may be interesting to compare the three sets of effective half-lives obtained from the data of urine, feces and chest. The shortest half-life appeared in the short-term component of fecal excretion (about 1.1 days). On the other hand, the longest half-life appeared in the long-term component of chest clearance (about 90 days) and the intermediate half-lives of similar length, in chest and feces (about 23 days and 25 days each). Therefore, the behavior of  $^{170}\text{Tm}$  inhaled in the body as the oxide was presumed as follows.

(1) Within the first ten days there was a very rapid clearance, though, unfortunately, no data supporting this were obtained on the deposition and excretion for the first 4 days. The major part of deposition in the naso-pharyngeal region and in the ciliated area of the tracheo-bronchial region was excreted in feces via the gastro-intestinal tract and the minor part of the deposition was excreted in urine with a biological half-life of less than 3 days after being absorbed into the blood.

(2) In the next period of about 4 months, there was a rapid decrease with a biological half-life of about 30 days. During this period, the fecal excretion involved 1) the material initially deposited in the pulmonary region, subsequently moved up the bronchial tree and swallowed, though a certain quantity of the material was removed through the very rapid clearance phase above mentioned, and 2) the material initially deposited in the pulmonary region, absorbed in blood and afterward excreted via bile and others. These exogenous and endogenous component of excretion could not be separated by measurements.

(3) In the third period, the deposition in the chest decreased exponentially with effective half-lives of  $90 \pm 11$  days which corresponded to the biological half-lives of 140 to 450 days with a mean of about 300 days. The activity detected in the chest would be mainly due to the non-transportable  $^{170}\text{Tm}$  deposited in the lungs though the measurements by chest counting included the radionuclide in the lungs, lymph nodes and chest wall. In addition, as described in the results, the urinary excretion data (Fig. 2(b)) were explained by the two components which had about 23 and 90 days half-lives which were the same as those in the chest.

In short, in this case study we assumed that the retention of  $^{170}\text{Tm}$  in the chest fitted a sum of three exponential terms having effective half-lives of about 3, 23 and 90 days, respectively, though the half-life of 3 days was not distinctly determined. On these assumptions, it was estimated that at least 1.5  $\mu\text{Ci}$  of  $^{170}\text{Tm}$  were initially deposited in the lungs of the subject.

##### 4. 2. Distribution

The nature of the work caused the inhalation and the subsequent study concerning the aerosols strongly supported that the inhaled material was thulium-170 sesquioxide ( $^{170}\text{Tm}_2\text{O}_3$ ) which was insoluble in water. Since it was impossible to make observation on the material inhaled, particle size investigation was carried out on the other thulium oxide pellet in which thulium-170 was previously formed by neutron activation. Thulium aerosol produced on the various conditions was measured with an electron microscope and a cascade impactor. Repeated measurements on the aerosols showed that the activity median aerodynamic diameter (AMAD) of this particle size distribution changed with the duration time of arc welding and with time after welding, and fluctuated between 0.01  $\mu\text{m}$  with a geometric standard deviation,  $\sigma_g$ , of 5 and 0.5  $\mu\text{m}$  with  $\sigma_g$  of 1.4. Although there was no evidence that this was the case in the incident, we estimated that submicron particles were inhaled in the incident.

Using the compartment model proposed by the Lung Dynamics Task Group,<sup>9</sup> the depositions of thulium-170 in the lungs of the subject were evaluated, as a class Y compound. The percentage depositions obtained were about 1% for the

naso-pharynx region (N-P), about 8% for the tracheo-bronchial region (T-B) and about 60% for the pulmonary region (P). The dominant deposition in the pulmonary region predicted the presence of the long-term component of the retention in the chest as seen in Fig. 1(a). Moreover, the characteristics of the non-transportable submicron particle might be responsible for the rapid excretion at early stage and the subsequent slow excretion in urine as in Fig. 2(a).

Human exposure to thulium-170 oxide has been reported in literature. Eakins and Morgan,<sup>10</sup> and Strambi and Testa<sup>11</sup> investigated different inhalation cases and studied excretion patterns, but no thulium-170 could be detected in urine.

Thomas and Kingsley<sup>12</sup> studied inhalation of  $^{171}\text{Tm}_2\text{O}_3$  in beagle dogs and reported that 63% of the sacrificed body burden was in the skeleton, 18%, in the lungs and 11%, in the liver 128 days post exposure. However, on our observations, the depositions in the skull and in the femur were not clarified though the possibility remained.

#### 4. 3. Urinary and fecal excretion in relation to chest retention

The urinary, fecal and total excretion rates of  $^{170}\text{Tm}$  in  $\mu\text{Ci}/\text{d}$  observed during the period from 7 to 126 days after inhalation were plotted against time in days after inhalation, on log-log paper, after being corrected for radioactive decay. Each set of values could be described by a power function,  $y = At^{-B}$ , where  $y$  is the activity excreted per day in  $\mu\text{Ci}/\text{d}$ ,  $t$ , time in days and  $A$  and  $B$ , constants. The values of  $A$  and  $B$  were 0.002, 0.058, 0.085 ( $\mu\text{Ci}/\text{d}$ ) and 0.25, 0.65, 0.71 for urinary, fecal and total excretion, respectively.

Integration of these power functions from  $t = 7$  to 126 days gave the total amounts of urine, feces and total excreta during the period. They were 0.1, 0.57, 0.69  $\mu\text{Ci}$ , respectively. On the other hand, according to the direct chest counting, about 0.75  $\mu\text{Ci}$  were lost through biological routes from the chest region during the period from 7 to 126 days after inhalation. Although it was impossible to take into account the rapidly decreased component of the excretion which might appear for the first 10 days, this value, 0.75  $\mu\text{Ci}$ , agreed fairly well with the total amount (0.69  $\mu\text{Ci}$ ) excreted during the same period. This attempt was also made on the exponential expression of the same data, and the similar result obtained.

#### Acknowledgements

Grateful acknowledgements are extended to the subject for his patience and co-operation and to Dr. I. Miyanaga, Head, Health Physics and Safety Division, and Dr. M. Fujita, Chief, Health Physics Laboratory, for their valuable suggestions and guidance.

#### References

- 1) 'Directory of Whole-body Radioactivity Monitors', JA 1.1, IAEA, Vienna (1970)
- 2) C. M. Lederer, et al.: 'Table of Isotopes', 6th Edition, John Wiley & Sons, New York (1968)
- 3) 'International Directory of Radioisotopes', IAEA, Vienna (1962)
- 4) R. J. Everett: SC-RR-66-2579 (1967)
- 5) T. Shirotani, M. Fujita: J. Nucl. Sci. Tech. 9, 165 (1972)
- 6) T. Shirotani, M. Fujita: J. Nucl. Sci. Tech. 10, 301 (1973)
- 7) D. Ramsden, D. A. Waite: 'Assessment of Radioactive Contamination in Man', 65, IAEA, Vienna (1972)
- 8) 'The Assessment of Internal Contamination Resulting from Recurrent or Prolonged Uptakes', ICRP Publication 10A, Pergamon Press, Oxford (1971)
- 9) ICRP Task Group on Lung Dynamics: Health Physics, 12, 173 (1966)
- 10) J. D. Eakins, A. Morgan: 'In Assessment of Radioactivity in Man', I, 230, IAEA, Vienna (1964)
- 11) E. Strambi, C. Testa: RT/PROT-(57)4 (1966)
- 12) R. L. Thomas, B. L. Kingsley: Health Physics, 21, No. 6, 15 (1971)

## GASTRO-INTESTINAL ABSORPTION OF CERIUM IN SUCKLING MICE

Y. Feige, A. Naharin, E. Lubin and T. Sadeh

Radiation Safety Dept.,  
Soreq Nuclear Research Centre,  
Yavne, Israel.

### Abstract

The absorption of lanthanides from the gastro-intestinal tract is known to be extremely low in adult mammals. Our previous studies as well as some recent publications revealed that the absorption of various heavy metal ions, including lanthanides, from the G.I. tract is relatively high during the early stages of life in mice and rats, and decreases gradually during the suckling period. We investigated specifically and quantitatively the variations with age of the absorption of cerium from the G.I. tract in young mice during their suckling period.

Post partum albino mice were injected intraperitoneally with  $^{144}\text{Ce}^{+3}$  citrate at different times during the lactation period. Twenty four hours after the injection to the mothers, the suckling litters were sacrificed and the  $^{144}\text{Ce}$  content in their digestive tract, liver and carcass determined by gamma counting. The internal deposition, via gastrointestinal transfer as determined from the ratio between the  $^{144}\text{Ce}$  activities found in the liver and carcass to that ingested with the milk during 24 hours of suckling, was about 4.5% during the first 24 hours of life and about 2% for the whole suckling period. These values, obtained under normal physiological conditions, are about two orders of magnitude above the currently accepted values for gastrointestinal absorption of lanthanides in adult mice, and confirm previous findings regarding the age dependence of the gastro-intestinal absorption of heavy metal ions during the early period of life. Various suggestions put forward to explain these findings are discussed. The assumption that the discriminatory properties of the G.I. tract in mammals are not fully developed at birth seems consistent with all recent experimental findings.

### Introduction

In spite of the non-metabolic role of the rare earths, small but significant fractions of lanthanides are absorbed from the gastrointestinal tract of very young mammals.

We investigated specifically and quantitatively variations in the absorption of cerium from the gastrointestinal tract of suckling mice, under normal physiological conditions<sup>1</sup>, and confirmed earlier findings that the absorption of  $^{144}\text{Ce}$  during the suckling period is age dependent and much higher than in the adult mouse<sup>2-6</sup>.

The relatively high fraction of lanthanides transferred from the gastrointestinal tract into the blood stream in the very young may have some



practical consequences. The Maximum Permissible Intake values generally used are based on experiments with adult mammals whose absorption of lanthanides is extremely low. These values, though safe and acceptable for adult humans, could cause unacceptably high internal exposures when fed to infants.

#### Methods

Gravid albino mice were divided into seven experimental groups of four. Every mouse received an intraperitoneal injection of 5  $\mu\text{Ci}/0.2$  ml.  $^{144}\text{Ce}^{+3}$  citrate during the lactation period. The seven groups were injected at different times: 3 hours after delivery, and the 2nd, 4th, 7th, 10th, 12th and the 16th day after delivery. By the 16th day the litters already consume a significant amount of solid food in addition to milk.

Twenty-four hours and 28 days after dose administration to the dams 4 neonates of each litter were sacrificed and radioassays performed on the digestive tract, liver and the remaining carcass. The first day was chosen on the assumption that during this short time there would be no excretion by the young of the contaminant ingested through the milk.

Measurements were carried out in a 3" well-type NaI crystal, connected to a single channel analyzer. For calibration of the counting system  $5 \times 10^{-4} \mu\text{Ci}$  of  $^{144}\text{Ce}$  diluted in 10 ml of water was used as a standard. The counting efficiency at 133 keV was about 1%.

#### Results

The transfer and internal deposition of  $^{144}\text{Ce}$  in neonates of various ages after suckling for 24 hours from contaminated mothers is presented in Table I. Earlier experiments <sup>2,3</sup> showed that the content of the carcass could be taken as representing the retention of the skeleton. Ce transfer via milk during the first 24 h was practically equal to the activity determined in the G.I. tract while the internal deposition due to gastrointestinal transfer was calculated as the ratio between the combined  $^{144}\text{Ce}$  activities found in the liver and carcass to that ingested during the first 24 hours.

The transfer of cerium through the intestinal wall decreases from 4.4% at birth to less than 1% at weaning. From the transfer ratios one finds that about 2.3% of the G.I. tract content is internally deposited during the entire lactation period, which corresponds to about  $4.5 \times 10^{-5}$  of the  $^{144}\text{Ce}$  injected to the dam. The total fraction of contaminant transferred from the mother to her suckling offspring during lactation is so low that no radiation hazard to the breast fed infant is involved, provided lactating mothers consume food contaminated with radioactive lanthanides not exceeding the currently accepted Maximum Permissible Intake values.

The last column of Table I shows the retention of  $^{144}\text{Ce}$  in the neonates that were allowed to suckle freely, 28 days after the dose was administered to the mother. The internal contamination of neonates is higher the earlier the injection of the dam occurred, mainly because of the longer suckling period.

#### Discussion

Many factors such as dietary characteristics (solid or liquid) and the chemical state (unbound or chelated) of the food, as well as the degree of peristaltic motion of the intestine are known to affect the absorption of heavy metal ions from the G.I. tract. Shiraiishi and Ichikawa suggested that pinocytosis could be the main transfer mechanism of dissolved cerium ions in suckling mammals<sup>5</sup>. This mechanism requires the operation of chelation or polymerization processes which form aggregates or large colloids. Only in such

form could cerium ions be transported across the membrane of the G.I. tract by pinocytosis. This hypothesis, however, seems to conflict with the conclusions of Inaba and Lengemann who showed that cerium remained in the epithelial cells of the villi of the G.I. tract until eliminated by the process of sloughing<sup>6</sup>. Clearly the role of pinocytosis as a route of transfer cannot be regarded as proven so far. Their suggestion that the ileum might be regarded as a possible site of entry due to the previous accumulation of cerium within the epithelial cells<sup>6</sup> is also not convincing. Limiting ourselves to measuring the transfer for 24 hrs suckle only, any accumulation factor is obviously eliminated without a significant reduction on the transfer of cerium into the neonates as compared to our earlier experiments<sup>2</sup>.

Factors other than the formation of colloid or macromolecules may influence the absorption of polyvalent ions from the intestine. Some data indicate that the association with milk facilitates the absorption of calcium and strontium from the G.I. tract<sup>7-9</sup>. Taylor suggested that the high absorption of radionuclides in very young animals may be caused by the inhibition of an existing block by lactose or other factors present in the milk diet<sup>10</sup>. If this interpretation were correct one might expect a positive correlation between the amount of milk ingested and the fractional absorption from the G.I. tract. Although milk production increases steadily during the first week of lactation, the fractional absorption of cerium, (as shown in Table I) decreases continuously from the day of birth. The enhanced absorption of lanthanides from the G.I. tract in the very young cannot therefore be accounted for by the "milk effect" alone according to our experiments.

Plutonium and actinides resemble lanthanides in their extremely low absorption from the G.I. tract in adult mammals. Studies on the intestinal absorption of ingested Pu also showed an increased absorption in the very young and a gradual decrease in the G.I. transfer with age<sup>11-14</sup>. However, it was demonstrated by Finkel<sup>13</sup> that milk was not the main factor contributing to the enhanced absorption of Pu from the G.I. tract of suckling rats.

The mechanism responsible for the discrimination of the G.I. tract against heavy metal ions is thus not well understood and needs further study. It seems to take some time for an active transport mechanism favoring calcium ion absorption on the one hand, and the establishment of an effective exclusion mechanism against metabolically useless metal ions on the other, to develop after birth. This selective barrier becomes fully operative only after weaning, when the diet becomes varied and the need for a discriminatory mechanism arises.

#### References

1. A. Naharin, Y. Feige et al: Internal Deposition of Ingested Cerium in Suckling Mice, to be published in Health Phys. (1974).
2. A. Naharin, E. Lubin and Y. Feige, Health Phys. 17, 717 (1969).
3. D.D. Mahlum, M. R. Sikov, Health Phys. 14, 127 (1968).
4. N. Matsuoka, J. Inaba, et al: Proc. of 9th Hanford Symp. Ed: M.R. Sikov and D.D. Mahlum, USAEC Symposium Series, 17, 217 (1969).
5. Y. Shiraishi and R. Ichikawa, Health Phys. 22, 373 (1972).
6. J. Inaba and F.W. Lengemann, Health Phys. 22, 169 (1972).
7. C.L. Comar and R.H. Wasserman, in Mineral Metabolism, Ed: C.L. Comar and F. Bronner, Academic Press, Vol. II, Part A, P. 539 (1964).

8. C.E. Bugg, J. Amer. Chem. Soc. 95, 908 (1973).
9. P. Charley and P. Saltman, Science 139, 1205 (1963).
10. D.M. Taylor, Proc. of the 1st Int. Symposium on Strontium Metabolism, Ed: M.A. Lenihan, p. 175, (1961).
11. M.R. Sikov and D.D. Mahlum, Health Phys. 22, 707 (1972).
12. Y.E. Ballou, Proc. Soc. Exp. Biol. Med. 98, 726 (1958).
13. M.P. Finkel, The absorption of ingested Pu, ANL, Int. C.R. Report 9 - 18 (1947).
14. The Metabolism of Compounds of Plutonium and other Actinides, ICRP Publication 19, p. 32, Pergamon Press (1972).

TABLE I. Transfer and Internal Deposition of  $^{144}\text{Ce}$  in Mouse Neonates at Various Ages

Age of neonate at $^{144}\text{Ce}$ injection to dam (days)	After first 24 hr of suckling		28 days later
	Transfer via milk = body burden (a)	Internal deposition (b) (%)	Body burden (a)
0	8.0 ± 0.3	4.4	9.6
2	9.8 ± 0.3	3.3	9.0
4	10.8 ± 0.4	2.5	7.0
7	11.7 ± 0.5	1.9	3.4
10	12.8 ± 0.5	1.7	2.1
12	13.9 ± 0.5	1.5	0.8
16	3.8 ± 0.3	< 0.6	0.6

$$(a) \quad 10^6 \times \left[ \frac{\text{Body burden of offspring}}{\text{Dose injected to dams}} \right]$$

$$(b) \quad 100 \times \left[ \frac{\text{carcass} + \text{liver}}{\text{G.I. tract} + \text{carcass} + \text{liver}} \right]$$

## A HUMAN METABOLIC MODEL FOR $^{14}\text{C}$ -LABELLED METABOLITES

### USEFUL IN DOSE ESTIMATION\*

S. R. Bernard  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee, USA

#### Abstract

An average individual (70-kg man) takes in some 350 g of carbon per day, and his total body content of carbon is about 14 kg. Assuming uptake to blood is essentially complete, this corresponds to a biological half-time ( $T_b$ ) of 28 days. Metabolic models used in ICRP Publication 10 allow only for  $T_b \cong 12$  days for glycine and of less than 1 day for  $\text{CO}_2$ . To be in agreement with the stable carbon data, one or more terms of longer half-life are needed. Such a model is defined in this paper and is more conservative than that for glycine in ICRP Publication 10 by about a factor of 3. Data of D. L. Buchanan on uptake of  $^{14}\text{CO}_2$  in mice indicate that the equilibrium level approached  $10^{-4}$  times the specific activity of  $^{14}\text{CO}_2$  in air, suggesting uptake of about 1% to blood with further dilution by dietary stable carbon of  $\sim 0.01$ . G. V. LeRoy *et al.* have determined the retention of  $^{14}\text{CO}_2$  in man during the first day, but need for a compartment of long biological half-life is indicated here also. When these compartments are included in the models given in ICRP Publication 2 and ICRP Publication 10, it is found that the former model overestimates dose to the total body by a factor of  $\sim 30$ , while the latter underestimates it by a factor indicated to be of least 16 and which might be as high as  $\sim 100$ .

#### Introduction

The purpose of this report is to present a mathematical model for  $^{14}\text{C}$  metabolism which can be used in health physics for estimating radiation dose to man from intake of  $^{14}\text{C}$ . This model is needed for an adequate estimation of radiation dose received by research workers or others who work with  $^{14}\text{C}$ -labelled compounds and incidentally or accidentally take  $^{14}\text{C}$  into their bodies. In this paper, experimental data provided by others will be used and interpreted with a mathematical model.

#### Experimental Data

D. L. Buchanan<sup>1</sup> obtained data from a study of  $^{14}\text{CO}_2$  inhalation by mice in stable  $\text{CO}_2$  levels ranging from 0.03% to  $\sim 5\%$   $\text{CO}_2$  in air. He let the mice inhale  $^{14}\text{CO}_2$  for as long as 40 days, and then he studied the retention out to 50 days after the end of exposure. By serial sacrifice, both during exposure and following the cessation of inhalation exposure, he took many tissues (but not bone) and plotted the results expressed as the ratio of specific activities

---

\* Research sponsored by the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.

in tissues to the specific activity of air. He called this the air carbon fraction, and from his graphs, the value increased from zero at time zero up to  $\sim 10^{-4}$  at 40 days or so. Thus, the equilibrium level of the specific activity was only 0.01% of the corresponding level in the air breathed by the mice.

Other data are available also, e.g., the paper by Skipper<sup>2</sup> which deals with the trapping of  $^{14}\text{C}$  in bone following injection into blood of  $^{14}\text{C}$ -labelled  $\text{Na}_2\text{CO}_3$ . Here the bone was seen to retain a small fraction for a long time, but there seemed to be no chronic accumulation of C in bone.

Data are available from another experiment of Buchanan<sup>3</sup> involving continuous feeding of a  $^{14}\text{C}$ -labelled diet of sucrose and yeast for a period of 40 to 50 days and serial sacrifice of mice and rats from which tissues were obtained. These data show the ratios of labelled to unlabelled C in tissues and foods applied and equilibrium set in at  $\sim 40$  days after intake began.

ICRP Publication 2<sup>4</sup> refers to data of Nardi for its  $T_b$  value of 10 days for total body. A  $T_b$  value for bone based on studies on mice and a value for fat are given in Publication 2 also. Note that all of these were obtained on small animals (mice and rats).

#### Metabolic Model for $^{14}\text{C}$

Denote by  $R(t)$  the fractional retention of dietary  $^{14}\text{C}$  in the body at time  $t$  after uptake of a unit amount into the bloodstream. Let  $E(t)$  denote the cumulative excretion (via all paths), and we write

$$E(t) = 1 - R(t). \quad (1)$$

It is assumed that

$$R(0) = 1 \quad \text{and} \quad R(\infty) = 0,$$

that is, the life span of man is considered to be long enough so that retention at 50 to 70 years  $\rightarrow 0$ . The mean residence time (MRT) in the body is

$$\begin{aligned} \text{MRT} &= \int_0^{\infty} t \frac{dE(t)}{dt} dt \\ &= - \int_0^{\infty} t \frac{dR(t)}{dt} dt \\ &= -tR(t) \Big|_0^{\infty} + \int_0^{\infty} R(t) dt \\ &= 0 + \int_0^{\infty} R(t) dt. \end{aligned} \quad (2)$$

Thus it has been proved (and this was also noted by Bergner<sup>5</sup> and probably others) that the integral of the retention function over all time is the mean of the probability distribution of residence times for an atom to be released from the body after the initial introduction of one atom at time zero. Now, one additional expression is needed to prove the MRT is given by the ratio of the body burden,  $q(t)$ , to the daily uptake into blood. Snyder<sup>6</sup> noted that for chronic intake with  $f_1 d\tau$  units of the isotope entering the body in  $d\tau$  units of time, the retention at a time  $t$  is given by,  $\tau \cong t$ ,



$$q(t) = \int_0^t f_1 I d\tau R(t - \tau). \quad (3)$$

By changing the variables, i.e., by letting  $t - \tau = T$  and  $dT = -d\tau$ , then (3) becomes

$$q(t) = \int_0^t f_1 I d\tau R(t - \tau) = f_1 I \int_0^t R(T) dT. \quad (4)$$

Rearranging the equation, one obtains

$$\frac{q(\infty)}{f_1 I} = \int_0^\infty R(T) dT = \text{MRT}. \quad (5)$$

### Application of the Model

In applying the model to man, it is assumed that body carbon is 14 kg and the diet contains 350 g/day of carbon<sup>3</sup> such that

$$\text{MRT} = \frac{14,000 \text{ g}}{350 \text{ g/day}} = 40 \text{ days},$$

which corresponds to a biological half-life of  $0.693 \times 40 \cong 28$  days. For mouse and rat, it is assumed that 1/5 of the body weight (and food weight per day) is carbon. Thus, for a 25-g mouse ingesting 4 g of food per day,

$$\text{MRT}_{\text{mouse}} = \frac{1/5 \times 25 \text{ g}}{1/5 \times 4 \text{ g/day}} = 6 \text{ days};$$

and for a 200-g rat eating 20 g of food per day,

$$\text{MRT}_{\text{rat}} = \frac{200}{20} \text{ day} = 10 \text{ days}.$$

In the above estimates, it is assumed that complete (100%) uptake of dietary C occurs.

The above values for mouse and rat agree with Buchanan's tissue data (from the experiment using a diet labelled with <sup>14</sup>C) in that the ratio of specific activity of organs and tissues to specific activity in diet is approximately given by

$$\frac{q^*/q}{I^*/I} = 1 - e^{-\lambda_b t}$$

which approaches 1 at large times (40 to 50 days in the case of rats and mice) where the asterisk denotes <sup>14</sup>C. Then we can show

$$1 - \frac{q^*/q}{I^*/I} = e^{-\lambda_b t}$$

which indicates that the exponential decreases with the reciprocal mean residence time as the decay constant. Buchanan's feeding data are in approximate accord with this equation. However, in some cases (brain and muscle, for example) more than one exponential is indicated, but the MRT for most tissues is 4 to 10 days which is approximately correct. Now Buchanan's inhalation experiment can be interpreted.

### New Model for $^{14}\text{CO}_2$ Inhalation

The daily intake of C in inhaled air is needed. For a mouse inhaling 0.15 cc per breath (the tidal volume) and 163 breaths per minute,<sup>8</sup> 23 cc of air are taken in per minute which corresponds to an intake of 33 liters per day. Air normally has 0.03%  $\text{CO}_2$ , and since  $\text{CO}_2$  has 44 g/mole and 1 mole of any gas occupies 22.4 liters, then

$$3 \times 10^{-4} \times \frac{33 \text{ liters}}{\text{day}} \times (22.4)^{-1} \frac{\text{mole}}{\text{liter}} \times \frac{12 \text{ g C}}{\text{mole}} = 0.005 \text{ g C/day}$$

is inhaled by the mouse. Since he eats 4 g of food per day with 1/5 being C, he ingests

$$4 (1/5) \text{ g C/day} = 0.8 \text{ g C/day.}$$

So he inhales  $\sim 10^{-2}$  as much carbon as he ingests. Thus the inhaled radioactive  $^{14}\text{CO}_2$  is diluted 100 times by ingestion of uncontaminated C present in food. This accounts for  $10^{-2}$  of the  $10^{-4}$  fraction observed by Buchanan. Evidently only 1% of the inhaled C is taken up by the blood in a form metabolically similar to C in food. This could be related to the fact that alveolar air has 3 to 5%  $\text{CO}_2$  concentration or 100 times the air inhaled. Thus the specific activity of the inhaled air is diluted by another factor of 100 to produce the equilibrium level in tissue.

It is believed that this is an important parameter to use in the case of inhalation intake of  $^{14}\text{CO}_2$  by a man who had inhaled, accidentally or otherwise,  $^{14}\text{CO}_2$ .

To get  $\mu\text{Ci}$ -days residence, we use

$$0.01 \mu\text{Ci} \int_0^{\infty} e^{-0.693t/28} dt \text{ days} = 0.4 \mu\text{Ci-days}$$

where 0.01  $\mu\text{Ci}$  represents uptake of 1% of 1  $\mu\text{Ci}$  inhaled by man. Compare this to the ICRP Publication 2 value of

$$0.75 \int_0^{\infty} e^{-0.693t/10} dt \mu\text{Ci-days} = 10.8.$$

Thus from their model, one overestimates the dose (given by

$$D = 51 \times \left( \frac{q}{m} \frac{\mu\text{Ci}}{\text{g tissue}} \right) \times \left( \frac{6 \text{ MeV}}{\text{dis}} \right) \times (\text{MRT days})$$

so dose is proportional to  $\mu\text{Ci}$  days) by a factor of

$$\frac{10.8}{0.4} = 27.$$

When the above is compared to the model in Publication 10 of ICRP,<sup>7</sup> care must be taken because Publication 10 only gives  $\mu\text{Ci}$ -days residence for intake to the blood. There, for  $\text{NaH}^{14}\text{CO}_3$ , they give a value of 0.58  $\mu\text{Ci}$ -day for bone, but this is for uptake to bone of 1  $\mu\text{Ci}$  and not for inhaled  $^{14}\text{C}$ . For the case of injection into blood, that fraction  $f$ , which has the metabolism of C taken in food, would give

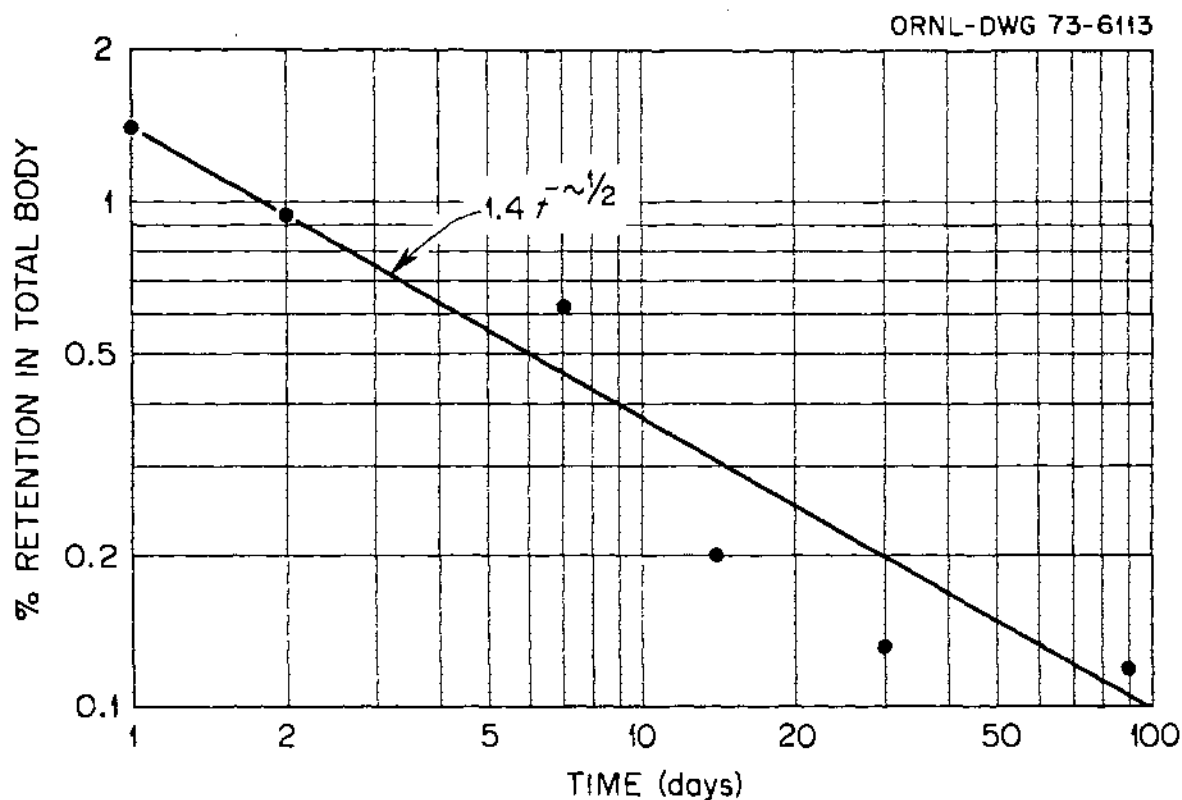
$$f \times \int_0^{\infty} R(t) dt = 40 f \mu\text{Ci-days.}$$

Included here is a logxlog graph of Skipper's data on  $^{14}\text{CO}_2$  total body retention in mouse after a single injection of  $\text{NaH}^{14}\text{CO}_3$  (Fig. 1). From here it can be noted that the power function will fit the data, and the MRT out to 900 days is only 1 day, somewhat lower than the earlier estimate of 6 days. This might indicate rapid exhalation of 5/6 of the total activity injected. Also, although the power function implies some trapping, it should be noted that there is no excessive concentration of C in bone or other organs in the body. More data on larger animals are needed here. Meanwhile, the value  $f = 1/6$  is suggested from the above data on mice.

Thus, from the above considerations, it can be seen that a model for  $^{14}\text{CO}_2$  inhalation of a single intake has been derived. Note, however, the value of 1% uptake of  $^{14}\text{C}$  into blood from inhalation is based on a very small animal--a mouse. The author knows of no data on a larger animal, such as a dog, from which the 1% could be verified. Note also in the above that the mean residence time factor indicates how fast the specific activities rise for the case of continuous intake. Also, when more than one exponential is involved in the retention, then the integral of the retention function gives the MRT; hence, it can be said, conversely, where the MRT is known, one can infer the approximate correctness of the retention function. For example, Publication 10 gives the retention function for injection of  $\text{NaH}^{14}\text{CO}_3$  into the blood of man as

$$R(t) = 0.7 e^{-0.693t/0.05} + 0.3 e^{-0.693t/0.4}, \quad t \text{ in days,}$$

and from this



Skipper's  $^{14}\text{CO}_2$  Data Injection of  $\text{NaH}^{14}\text{CO}_3$  into Mice.

Fig. 1.



$$\int_0^{\infty} R(t) dt = \text{MRT} = \frac{0.7 \times 0.05 + 0.3 \times 0.4}{0.693} \text{ days} = 0.22 \text{ days}$$

which is much below the value of 40 days. This suggests the existence of longer-term exponentials. If one exponential is added in the amount of 2% with a 1400-day half-life, then

$$R(t) = 0.7 e^{-0.693t/0.05} + 0.28 e^{-0.693t/0.4} + 0.02 e^{-0.693t/1400}$$

and then

$$\text{MRT} \cong 43 \text{ days.}$$

Although this MRT concept implies longer-term exponentials, it does not tell how many to add, and only experiments will indicate that.

#### A Model for $^{14}\text{C}$ -Labelled Glycine

While the above is for  $^{14}\text{CO}_2$  (or  $\text{NaH}^{14}\text{CO}_3$ ), a model is given in Publication 10 for  $^{14}\text{C}$ -labelled glycine (an amino acid) injected into blood of man. There it is recommended that

$$R(t) = 0.2 e^{-0.693t/0.12} + 0.2 e^{-0.693t/0.9} + 0.3 e^{-0.693t/6} + 0.3 e^{-0.693t/35}$$

From this function, the MRT can be seen to be  $\sim 15$  days, lower by a factor of 3 than the value above of 40 days. Thus it would be recommended that a component with a coefficient of 0.02 and a  $T_{1/2}$  of  $\sim 1200$  days be included to increase the MRT up to  $\sim 40$  days.

#### Conclusions and Recommendations

From the above it is concluded that in mice, 1% of the inhaled  $^{14}\text{CO}_2$  enters into long-term retention in the body. The mean residence time of C in the diet of mouse is 6 days, while for man, it is 40 days. It is recommended that the 1% uptake to blood be used rather than 75% (ICRP Publication 2 value) and the residence time of 40 days be used for metabolites (compounds which are involved in the buildup and breakdown of cells and tissues) ingested into the body of man. For a single injection of  $\text{NaH}^{14}\text{CO}_3$  into blood, some studies indicate a lower residence time in mice ( $\sim 1$  day) than for food labelled with  $^{14}\text{C}$  (6 days). When the above models are used in dose estimation for  $\text{CO}_2$ , it is found that ICRP Publication 2<sup>9</sup> overestimates the dose to the total body by a factor of  $\sim 30$ , while the ICRP Publication 10 model underestimates it by a factor indicated to be at least 16 and which might be as high as  $\sim 100$ . Probably the single exponential is an oversimplification and should be replaced by several exponentials of fairly long half-times, but to determine these, further data on man, or on several species of animals, would be desirable.

#### References

1. D. L. Buchanan, J. Gen. Physiol. **34**, 371 (1951).
2. H. E. Skipper, Nucleonics **10**, 40 (1952).
3. D. L. Buchanan, Biochem. Biophys. **94**, 500 (1961).
4. As appears in Health Phys. **3** (1960).
5. P. E. Bergner, Personal Communication.
6. As applied by W. S. Snyder in Publication 6 of ICRP (1962).
7. Report of Committee IV on Evaluation of Radiation Doses to Body Tissues from Internal Contamination due to Occupational Exposure (Pergamon Press, London, 1968).
8. W. S. Spector, Handbook of Biology Data (W. B. Saunders Co., Philadelphia, 1956).
9. Report of ICRP Committee 2 on Permissible Dose for Internal Radiation, Health Phys. **3** (1960).

## METABOLIC BALANCES OF $^{210}\text{Pb}$ AND $^{210}\text{Po}$ IN UNEXPOSED MEN\*

R. B. Holtzman, H. Spencer†, F. H. Ilcewicz, and L. Kramer†  
Argonne National Laboratory  
Argonne, Illinois 60439

### Abstract

The metabolic balances of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  were measured in each of 12 men maintained on a metabolic ward. These nuclides were determined in urine and feces from each subject collected for one month or more from each. Representative diets, drinking water and atmospheric levels in the ward were also sampled. The mean levels ( $\pm$ S.E.) were for  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , respectively, in diets over a period of five months ( $1.25 \pm 0.04$ ) and ( $1.63 \pm 0.05$ ) pCi/day, in urine ( $0.275 \pm 0.026$ ) and ( $0.269 \pm 0.033$ ) pCi/day, and in feces ( $1.333 \pm 0.062$ ) and ( $1.89 \pm 0.10$ ) pCi/day.

The mean overall balances (the difference between diet and excreta) of ( $-0.235 \pm 0.075$ ) pCi  $^{210}\text{Pb}$ /day and ( $-0.337 \pm 0.100$ ) pCi  $^{210}\text{Po}$ /day show that over the collection period, larger amounts were excreted than taken in the diet. Atmospheric intake accounts for an additional  $^{210}\text{Pb}$  intake of 0.07 pCi/day, but it contributes a  $^{210}\text{Po}$  intake of only about 0.02 pCi/day. The contribution of cigarette smoke and dietary levels of calcium to the balances is discussed.

### Introduction

The metabolic properties of  $^{210}\text{Pb}$  and its decay product,  $^{210}\text{Po}$ , in people exposed only to normal environmental levels of these nuclides are important because they contribute a large fraction of the dose from internally deposited nuclides.<sup>1,2</sup> The  $^{210}\text{Pb}$  produces essentially no dose, but with its 22-year physical half life, it can accumulate in the body. On the other hand, the  $^{210}\text{Po}$  with its 5.3-Mev alpha particle contributes 90 to 95% of the dose from this series ( $^{210}\text{Pb}$ - $^{210}\text{Bi}$ - $^{210}\text{Po}$ ), but its 138-day half life allows only a limited accumulation from sources other than the  $^{210}\text{Pb}$  in the body.

Metabolic balance studies may indicate other routes of intake and excretion. Thus, if the balance is negative (based on diet and excreta), one may look for other sources of intake which may be difficult to measure directly, such as atmospheric aerosols and cigarette smoke.

Some dietary and excretion data have been reported in unexposed populations, but the studies were generally of a limited nature. Glöbel et al.<sup>3</sup> studied the metabolic balances of these nuclides on one person over a one-month period. Magno et al.<sup>4</sup> measured the  $^{210}\text{Pb}$  in "typical" diets from several cities, and Morse and Welford<sup>5</sup> determined these levels in a composite New York diet. Some urinary excretion rates have been measured in people with potentially high exposures.<sup>6-9</sup>

---

\* Work performed under the auspices of the U. S. Atomic Energy Commission.

† Metabolic Research Unit, Veterans Administration Hospital, Hines, Illinois.

We report here on a study of the metabolic balances of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in men maintained on a metabolic ward for four weeks or more.

### Experimental Methods and Materials

Twelve fully ambulatory patients in good physical condition were studied under strictly controlled conditions on the Metabolic Research Ward.<sup>10,11</sup> Their ages were from 42 to 64 years with an average age ( $\pm$ S.E.) of  $(52.0 \pm 1.7)$  years, and their weights were from 62.6 to 87.0 kg with a mean of  $(76.0 \pm 2.0)$  kg. Each consumed a constant diet daily which weighed about 2 kg and contained 2200 calories, about 200 mg of calcium and 800 mg of phosphorus. Each subject chose his own daily consumption of water intake (average 2.8 liters/day) which was then maintained constant over the period of study. In some of the studies the calcium intake was increased to 800 or 2400 mg/day by addition of either milk, calcium lactate or calcium gluconate tablets to the above low calcium diet. Most of the patients had been maintained on the ward on the same diet for many months prior to these studies.

Complete urine and stool collections obtained for the study were usually pooled on a six-day basis, although some were combined into four-, eight-, or ten-day pools. Aliquots of representative diets were taken for analysis once a week over a period of 27 weeks. Two sets of drinking water samples of 1 liter each were measured. The atmospheric concentration of these nuclides in the ward was estimated from collections made at several periods during the study.

The  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in the samples were determined by the previously described procedure<sup>1</sup> of wet ashing and plating the  $^{210}\text{Po}$  onto a silver disk which was then alpha counted. Calcium was determined by atomic absorption analysis on a Perkin Elmer Model 303 Atomic Absorption Spectrophotometer.<sup>12</sup> The accuracy of the analytical results was generally  $\pm 5\%$  or better, except that those of the  $^{210}\text{Po}$  were about 10%.

### Results and Discussion

The metabolic pathways of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are illustrated in Fig. 1. Intake is from food, water, air and cigarette smoke, while output is through excretion and other pathways, such as loss of hair and desquamation of skin.<sup>13</sup> Both nuclides may enter the circulation from the gut and be excreted into the gut. One large contribution (and probably the major one) to the  $^{210}\text{Po}$  pool is that from the decay of  $^{210}\text{Pb}$  present in the body.

The largest source of intake is diet. The levels from the representative 6-day samples are shown in Fig. 2; the overall mean values ( $\pm$ S.E.) are  $1.248 \pm 0.029$  and  $1.630 \pm 0.048$  pCi/day for  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , respectively. The coefficient of correlation between the  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  data is essentially zero ( $R = -0.20$ ,  $P \gg 0.05$ ). However, the mean ratio of  $^{210}\text{Po}$  activity to that of  $^{210}\text{Pb}$ ,  $1.33 \pm 0.06$  is more important, since correlations may be neither evident nor important in data which vary little about their means.

These data show some systematic variations of the  $^{210}\text{Pb}$ , such as minima at the 3rd and 8th 6-day periods and maxima at the 5th and 17th periods. The variations are about 20% of the mean. The  $^{210}\text{Po}$  shows similar structure but it lags 3 to 5 periods behind the  $^{210}\text{Pb}$ . Some compensation for these variations is made in the balances by using dietary levels extant at the collection times. However, the means of the 27 measured diets and of the diets of the individuals were essentially identical.

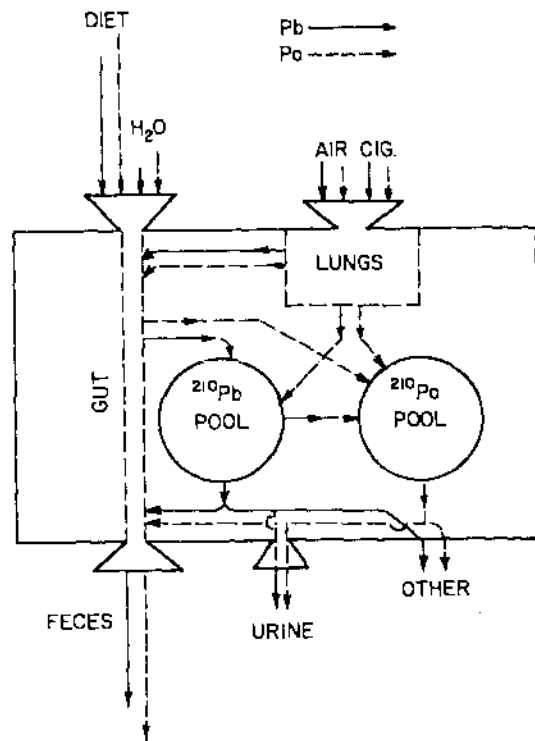


Figure 1. A model of the metabolic patterns of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ .

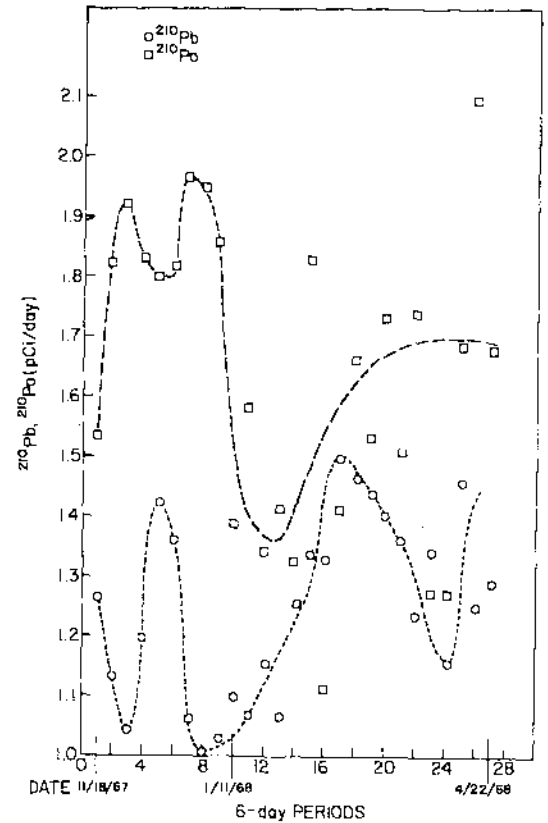


Figure 2. Contents of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in the diets of the subjects measured in representative 6-day samples.

The mean  $^{210}\text{Pb}$  content of these diets is lower than that found by Morse and Welford<sup>3</sup> in New York City of  $1.40 \pm 0.08$  pCi/2 kg and than the results of Magno et al.<sup>4</sup> for four U. S. cities of about 1.7 pCi/2 kg. The differences may be due to the lack of some constituents high in  $^{210}\text{Pb}$  present in the standard diet, but not present here, such as fresh vegetables and milk. The seasonal variations observed in these data suggest that these values may be at least as representative of a U. S. diet as the single and composite diets reported elsewhere. Drinking water contributes in addition about 0.11 and 0.10 pCi/day to the  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  intakes, respectively.

The contributions of inhalation to intake are more difficult to assess because they were not measured directly, and they will be estimated only for an average subject. This is estimated from the breathing rate and the atmospheric concentration. Because of the more sedentary condition of these men relative to working men, we assumed the average man's daily breathing rate to be  $15 \text{ m}^3/\text{day}$  rather than the  $20 \text{ m}^3/\text{day}$  of the ICRP "Standard Man".<sup>14</sup> The atmospheric concentrations of the nuclides are shown in Fig. 3. The mean specific activities were  $(10.9 \pm 0.9)$  and  $(1.76 \pm 0.16)$  pCi/1000 SCM (Standard Cubic Meter) for  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , respectively. The total intake from the atmosphere is then 0.16 pCi  $^{210}\text{Pb}$  and 0.026 pCi  $^{210}\text{Po}$  per day.

The mean  $^{210}\text{Pb}$  excretion rates for each subject, along with his respective dietary and water intake and the respective balances are shown as a function of increasing balance in Fig. 4. Because of the large uncertainties and because they were not measured directly, the contribution of inhalation are not included in these figures. The  $^{210}\text{Pb}$  values indicate that the balances depend to a large

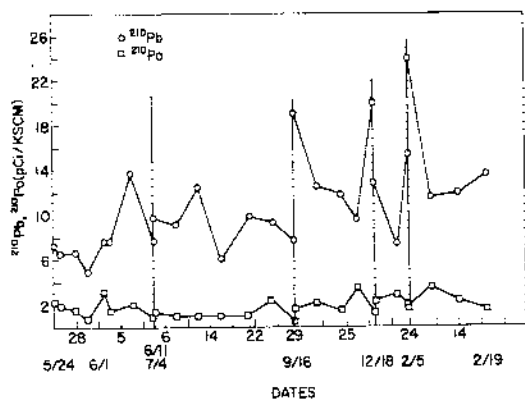


Figure 3. Concentrations of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in atmospheric aerosols sampled in various periods from 5/24/68 to 2/19/69. Each date is the mean date of the sampling period.

extent on the fecal values, which in most cases are about equal to the dietary intake. The more negative balances show more fecal output than dietary input and the more positive ones less fecal output. There also appears to be a general downward trend of both fecal and urinary levels as the balance increases. The calcium balances and high calcium diets (noted by the numbers in mg/day intake next to the calcium balance points) do not appear to affect the  $^{210}\text{Pb}$  balances.

The  $^{210}\text{Po}$  results were similar to those of  $^{210}\text{Pb}$  but their values scatter more and show fewer associations, such as little correlation between diet and feces. The order of the subjects by balance is different than that for  $^{210}\text{Pb}$ .

The mean ( $\pm$ S.E.) excretion rates and ranges are shown in Table 1.

The contribution of cigarette smoke to the nuclide intake is shown in Fig. 5 by balance vs. number of cigarettes smoked per week for the seven subjects on whom we measured cigarette consumption. For  $^{210}\text{Pb}$  the linear regression of balance (B) vs. cigarettes/week (C) is

$$B = (-0.10 \pm 0.17) - (0.0015 \pm 0.00043)C$$

and the correlation coefficient is  $-0.59$  ( $P \approx 0.10$ ). While neither the intercept nor the slope is statistically significant, they do give an indication of the values. The intercept is equivalent to the atmospheric intake of  $0.1$  pCi/day and the slope is about  $0.21$  pCi/day, in a person smoking one pack per day. (These seven people smoked an average of 22 cigarettes per day.) For  $^{210}\text{Po}$  the levels of significance are even poorer, although, if the intercept is forced through zero, the slope of  $-0.0015$  pCi/day is significant ( $P \approx 0.05$ ). Previous studies on the nuclide content of cigarette smoke<sup>15,16</sup> lead to an estimated total intake of  $0.30$  pCi  $^{210}\text{Pb}$  and  $0.72$  pCi  $^{210}\text{Po}$  per day.

The results in Fig. 5 are compatible with the directly measured values of intake from smoke, if one assumes that retention of inhaled nuclides in the respiratory tract is 50% as indicated by various data in the literature.<sup>14,17-19</sup> Daily deposition would then be  $0.23$  pCi ( $0.08$  from air and  $0.15$  from cigarettes) and that from  $^{210}\text{Po}$  would be  $0.37$  pCi ( $0.01$  from air and  $0.36$  from cigarettes) for persons smoking one pack/day.

The balance based on intake from diet and water only, is negative, but it becomes essentially zero, if we add to the intake the contribution from inhalation, as shown in Table 2. In this table the dietary intakes are the average of our representative 6-day samples, the inhaled values are those estimated for inhalation alone, and the excretion values are the overall averages. The different means, taken from the overall averages in Table 2 (diet + water + inhala-

Table 1. Mean excretion rates and balances of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ .

	$^{210}\text{Pb}$ (pCi/day)	$^{210}\text{Po}$ (pCi/day)
Urine	$0.275 \pm 0.026$ (0.11 to 0.41)	$0.269 \pm 0.033$ (0.079 to 0.52)
Feces	$1.333 \pm 0.062$ (0.99 to 1.83)	$1.89 \pm 0.10$ (1.54 to 2.46)
Balance*	$-0.235 \pm 0.075$ (-0.73 to +0.25)	$-0.337 \pm 0.100$ (-0.95 to +0.25)

\* diet + water - excreta

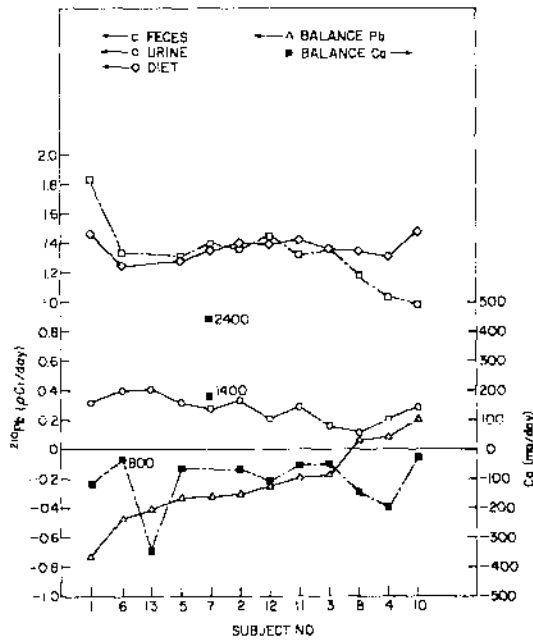


Figure 4. The mean daily levels of  $^{210}\text{Pb}$  for each subject in diet, feces and urine and the mean daily  $^{210}\text{Pb}$  and calcium balances. The subjects are listed in order of  $^{210}\text{Pb}$  balance. The calcium intakes greater than the standard 200-mg per day level are given in mg/day for Subjects 6 and 7.

of 600 pCi (the  $^{210}\text{Pb}$  body content of an average man<sup>20</sup>) and with a mean balance of  $^{210}\text{Po}$  of less than 0.1 pCi/day, the implied half life is very long and comparable to the 7-year values observed in people high in  $^{226}\text{Ra}$ .<sup>21</sup> The  $^{210}\text{Po}$  activities are then essentially in radioactive equilibrium with the  $^{210}\text{Pb}$  (> 90% of the  $^{210}\text{Pb}$  activity).

Thus, the low ratios of  $^{210}\text{Po}$  to  $^{210}\text{Pb}$  in diet and excreta confirm the previous estimates<sup>20</sup> that only a small fraction of the  $^{210}\text{Po}$  in the body is supported by diet when the ratio of the activity of  $^{210}\text{Po}$  to that of  $^{210}\text{Pb}$  is about unity *in vivo*. Even though the residence time of  $^{210}\text{Po}$  is much longer than the 25 days assumed earlier,<sup>20</sup> the limiting factor would be the 138-day physical half life. With no excess of excretion over intake, the accumulation from the diet would be only about 24 pCi, compared to 500 to 600 pCi in the body (if the  $^{210}\text{Po}$  is nearly in radioactive equilibrium with the  $^{210}\text{Pb}$ ).

tion - excreta), is -0.02 pCi/day for  $^{210}\text{Pb}$  and -0.06 pCi/day for  $^{210}\text{Po}$  which is essentially identical to that of individual balance taken from the mean of the balances for each individual based on his diet, water and excreta (-0.275 and -0.269 pCi of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , respectively) to which has been added the estimated mean daily inhalation intake of 0.23 pCi of  $^{210}\text{Pb}$  and 0.37 pCi of  $^{210}\text{Po}$ . Thus, the mean balances approach zero to within 3% of the intake values.

These results are lower than those of Glöbel et al.<sup>3</sup> in Germany. Their diet (and fecal excretion) had more than twice the nuclide content of ours, 4.65 pCi of each, and the  $^{210}\text{Po}/^{210}\text{Pb}$  ratio was about unity compared to our ratio of 1.33. The urinary excretion of  $^{210}\text{Po}$  of 0.3 pCi/day was comparable to ours, but the  $^{210}\text{Pb}$  excretion was twice this value, in line with the higher dietary levels. The values of Glöbel et al. are probably not typical of the U.S., either because they studied a particular person or because their foods were different.

The balance of the  $^{210}\text{Po}$  is important to estimates of radiation dose, while the amount in the body under normal intake depends mainly on the  $^{210}\text{Pb}$  content of the body rather than on the intake. For an internal source of  $^{210}\text{Pb}$

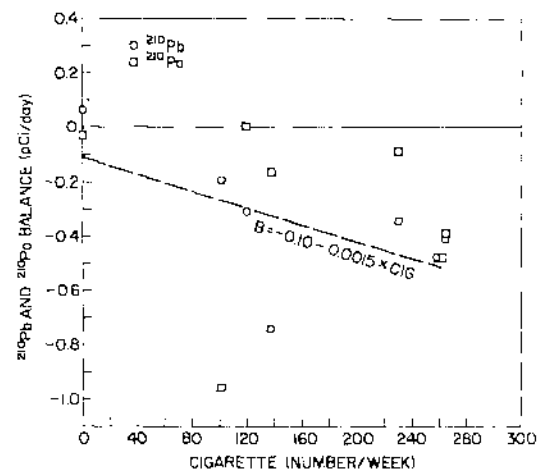


Figure 5.  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  balances plotted against the weekly cigarette usage for each of the seven subjects for whom this quantity was determined.

Table 2. Summary of components of metabolic balances.

I. Intake		
A. Ingestion		
Diet	1.25	1.63
Water	<u>0.11</u>	<u>0.10</u>
	1.36	1.73
B. Inhalation (50% retention)		
Air	0.07	0.02
Cigarette Smoke*	<u>0.15</u>	<u>0.36</u>
	0.24	0.37
C. Total	1.60	2.10
II. Output		
Excretion		
Feces	1.33	1.89
Urine	<u>0.28</u>	<u>0.27</u>
	1.61	2.16
III. Balances		
A. This Table	-0.02	-0.06
B. Individual (see text)	0.00	+0.03

The values of  $^{210}\text{Po}/^{210}\text{Pb}$  ratios of 1.07 in urine and 1.47 in feces make it possible to estimate the amounts of one nuclide given a measurement of the other. However, because of the large variability of these ratios, this estimate is reliable only to within a factor of 2 or so.

In summary, the  $^{210}\text{Pb}$ - $^{210}\text{Po}$  content of the diets appear to be representative of those in the U.S. when compared to the data of others. They exhibit seasonal variations, a phenomenon not noted in other measurements.<sup>3-5</sup> Both the diets and fecal excretions had a 30 to 40% excess of  $^{210}\text{Po}$  over  $^{210}\text{Pb}$ , while urinary excretion exhibited equal amounts of each. When account is taken of the contributions of inhalation to intake, this group is in material balance with respect to both nuclides. This result tends to justify the assumption on inhalation used in arriving at the intake values.

\*1 pack/day

#### References

- Holtzman, R. B., Health Phys. 9, 385 (1963).
- United Nations of Scientific Committee on the Effects of Atomic Radiation. Ionizing Radiation: Levels and Effects. (United Nations, New York, 1972), p. 72.
- Gibbel, B., H. Muth and E. Oberhausen, Strahlentherapie 131, 218 (1966).
- Magno, P. J., P. R. Groulx and J. C. Apidianskis, Health Phys. 18, 383 (1970).
- Morse, Robert S. and George A. Welford, Health Phys. 21, 53 (1971).
- Holtzman, R. B. and F. H. Ilcewicz, Argonne National Laboratory Radiological and Environmental Physics Division Annual Report ANL-7860-II, (July 1970 through June 1971), p. 307.
- Black, S. C., V. E. Archer, W. C. Dixon and G. Saccomanno, Health Phys. 14, 81 (1968).
- Bale, W. F. and J. V. Shapiro, Proceedings of the International Conference on Peaceful Uses of Atomic Energy. Vol. 13, p. 233, P/276 (Geneva, United Nations, 1956).
- Cohen, Norman and Theo. J. Kneip, Health Phys. 17, 125 (1969).
- Spencer, Herta, Joan A. Friedland and Vernice Ferguson, (Biological Mineralization. I. Zipkin, ed. (John Wiley and Sons, Inc., New York, 1973) p. 689.
- Spencer, Herta, Lois Kramer, Joseph Samachson, Edward P. Hardy and Joseph Rivera, Health Phys. 24, 525 (1973).
- Perkin-Elmer Corporation. Analytical Methods for Atomic Absorption Spectrophotometry. 1968.
- Jaworowski, Z., Nukleoneka IX, 57 (1964).
- International Commission on Radiological Protection, Committee II on Permissible Dose for Internal Radiation (1959). Alao, Health Phys. 3, 1 (1960).
- Holtzman, R. B. and F. H. Ilcewicz, Science 153, 1259 (1966).
- Ferri, Esther S. and Howard Christiansen, Public Health Reports 82, 828 (1967).
- George, A. and A. J. Breslin, Health Phys. 17, 115 (1969).
- Aurand, Karl, Wolfgang Jacobi, Hermann Muth and Alfred Schraub, Strahlentherapie 112, 262 (1960).
- International Radiological Protection Commission Task Group on Lung Dynamics, Committee II, Health Phys. 12, 173 (1966).
- Holtzman, Richard B., Health Phys. 10, 763 (1964).
- Holtzman, R. B., Argonne National Laboratory Radiological Physics Division Annual Report ANL-7760-II (July 1969 through June 1970), p. 35.

LONG-TERM METABOLISM OF  $^{90}\text{Sr}$  IN RHESUS MONKEYS (*Macaca mulatta*)\*  
Patricia W. Durbin, Nylan Jeung, Marilyn H. Williams, and Marshall W. Parrott\*\*  
Donner Laboratory and the Division of Biology and Medicine,  
Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

ABSTRACT

The plasma content ( $P_t$ ) and daily excretion rate ( $E_t$ ) were measured, and the whole-body retention ( $R_t$ ), was calculated for 7 to 15 yr after a single injection of  $^{90}\text{Sr}$  in young (2.3-3 yr), adolescent (3.1-5 yr), and adult (> 5 yr) rhesus monkeys. Half-times of 6-term exponential equations of  $P_t$  and  $E_t$ —0.4-0.5 d, 1-2 d, 3.5-7 d, 40-80 d, 300-650 d, and 2400-3600 d—are similar within each age group and also among the 3 age groups. The half-times of the first 5 terms of equations of  $R_t$  are similar to those of  $P_t$  and  $E_t$ , but those of the 6th terms are longer, 4200-5600 d, presumably representing re-deposition of  $^{90}\text{Sr}$  recirculated from remodeled bone. The coefficients of the metabolic equations demonstrated greater early  $^{90}\text{Sr}$  excretion by the older monkeys and greater late retention of  $^{90}\text{Sr}$  by the growing monkeys. When age at injection was considered,  $^{90}\text{Sr}$  retention in monkeys agreed with retention of  $^{85}\text{Sr}$  or  $^{90}\text{Sr}$  in other simians, beagles, and human adults.

INTRODUCTION

Even before the first thermonuclear weapons were exploded in the Marshall Islands (March 1954), it was recognized that  $^{90}\text{Sr}$  would be a hazardous constituent of the radioactive fallout, but there was little information on  $^{90}\text{Sr}$  metabolism in species other than rodents.<sup>1-3</sup> In 1954, urged by Dr. Willard F. Libby, then a member of the U.S. Atomic Energy Commission, and the late Dr. Joseph G. Hamilton, director of the Crocker Laboratory, we fed or injected animals from the laboratory's small colony of rhesus monkeys with low doses of  $^{90}\text{Sr}$ .<sup>4-6</sup>

As new techniques were developed,<sup>7,8</sup> metabolic information began to be available from human beings directly, and interest in study of other species declined. Controlled long-term study of the alkaline earths in human beings is not possible, because there are no long-lived  $\gamma$ -emitting isotopes of these elements. Furthermore, the long-term data from the human  $^{226}\text{Ra}$  cases are equivocal, because of late radiation effects and uncertainty in their  $^{226}\text{Ra}$  exposure levels. Therefore, we maintained and gradually added to the  $^{90}\text{Sr}$ -injected monkey colony to obtain from individual monkeys over a significant part of their 30-yr life span metabolic data that could be used to test long-term models of alkaline earth metabolism.<sup>9</sup> This report summarizes our study of  $^{90}\text{Sr}$  in monkeys and compares their long-term retention of  $^{90}\text{Sr}$  with that in other species.

METHODS AND MATERIALS

Rhesus monkeys caught in the wild were purchased from animal dealers. Ages were determined from dentition<sup>10</sup> and skeletal roentgenograms.<sup>11</sup> The animals have been assigned to 3 age groups: young, 2.3-3 yr, most epiphyses open, skeleton and body weights increasing rapidly, sexually immature; adolescent, 3.1-5 yr, some epiphyses open, skeleton and body weights increasing slowly, sexually mature; adult, > 5 yr, epiphyses closed and permanent teeth erupted, skeleton and body weights stable. Colony management was described earlier.<sup>4</sup> A high-Ca diet (0.03-0.12 g/d/kg body weight) was fed to suppress reutilization of  $^{90}\text{Sr}$ .

Strontium-90 diluted in 3% Na citrate, was given in a single i.v., i.m., or i.p. injection. The only differences in  $^{90}\text{Sr}$  behavior related to route of injection were in the early plasma concentrations. Experimental protocol is shown in Table 1.

The monkeys are housed in individual noncommunicating stock cages from which, with careful cleaning, we obtain complete excreta collections. Excreta were collected frequently during the first year and thereafter in two 7-d pools every 8 wk. In some cases urine and feces were collected separately for the first few weeks. Blood samples were drawn frequently in the first weeks and at least twice yearly



Table 1. Injection and retention data for <sup>90</sup>Sr-injected monkeys

Injection data							
Monkey <sup>a</sup> no., sex	Age (yr)	Weight (kg)	<sup>90</sup> Sr ( $\mu$ Ci)	Injn. date	Days to death	<sup>90</sup> Sr in body (%)	Cause of death or reason for euthanasia
<u>Young monkeys (2.1 to 3 yr)</u>							
37F	2.6	4.4	12.8	2/15/60	1	45.4	Test animal
36F	2.3	3.1	12.8	2/15/60	4	56.2	Test animal
52F	2.3	2.8	130.5	11/13/61	21	48.5	Test animal
53F	2.7	2.6	130.5	11/13/61	66	69.3	Test animal
29F	3.0	3.3	41.6	9/10/58	280	34.8	Positive TB test
51F	2.7	2.8	26.1	11/13/61	441	26.6	Test animal
20F	2.7	3.6	49.8	1/15/57	707	13.8	Amoebic dysentery
50F	2.7	3.1	26.1	11/13/61	1211	24.8	Test animal
34F	2.9	3.6	56.2	2/21/58	1921	15.4	Accident
35F(1) <sup>b</sup>	2.4	4.0	56.2	2/21/58	2037	~10	Reinjected by mistake <sup>b</sup>
28F	2.6	2.9	41.6	9/10/58	2087	15.5	Accident
<u>Adolescent monkeys (3.1 to 5 yr)</u>							
10F	3.3	3.4	56.3	11/10/55	94	45.1	Test animal
64F	4.3	4.2	33.1	3/27/67	427	16.9	Test animal
33F	3.2	3.8	56.2	2/21/58	2247	7.3	Leptospirosis
9M	4.3	9.1	38.0	4/21/54	2520	18.0	Cyst on neck
31F	4.1	4.1	61.1	10/27/57	2639	5.6	Intussusception
27M	3.6	3.2	41.6	9/10/58	3159	11.2	Cage paralysis
65F	4.4	4.5	33.1	3/27/67	---	5.1 <sup>c</sup>	Alive, 2242 days p.i.
63F	4.1	5.4	46.9	1/9/67	---	5.8 <sup>c</sup>	Alive, 2319 days p.i.
32F	4.2	4.5	61.1	10/27/59	---	7.1 <sup>c</sup>	Alive, 5360 days p.i.
21F	3.2	4.2	49.8	1/15/57	---	5.7 <sup>c</sup>	Alive, 6010 days p.i.
<u>Adult monkeys (&gt;5 yr)</u>							
G8M	6.8	8.9	4.4	9/9/69	2	37.0	Test animal
G11M	~7	7.0	5.0	6/24/70	16	50.7	Test animal
98F	9.3	5.6	3.5	10/28/69	35	17.9	Test animal
40F	8.8	8.5	70.4	1/9/67	98	7.0	Pneumonia
35F(2) <sup>b</sup>	7.9	5.2	50.1	9/23/63	147	~8	Test animal <sup>b</sup>
7M	>6.5	4.2	37.5	3/16/54	181	21.4	Cage paralysis
23M	>7	5.0	49.8	1/15/57	3183	7.8	Accident
38F	5.4	3.2	50.1	9/23/63	3411	2.4	Uterine tumor
8F	~5	5.5	39.9	4/1/54	3507	2.9	Leptospirosis
39F	5.5	3.2	50.1	9/23/63	---	1.3 <sup>c</sup>	Alive, 3599 days p.i.
61M	6.7	6.8	117.6	2/25/63	---	3.8 <sup>c</sup>	Alive, 3809 days p.i.
62M	12	9.4	168.3	2/25/63	---	1.6 <sup>c</sup>	Alive, 3809 days p.i.

a In Ref. 4 monkeys 7M, 8F, 9M, and 10F were identified with pet names as follows: 7M, Tony; 8F, Rosy; 9M, Stupe; 10F, Pat.

b Monkey 35F was reinjected (by mistake) with <sup>90</sup>Sr 2037 d after her original injection. When she was killed 147 d after the second injection, the measured <sup>90</sup>Sr in excreta, bones, and tissues was compatible with the approximate <sup>90</sup>Sr body contents shown.

c Body count 4 to 7 mo before the closing date of this report, Aug. 1, 1973.

after the first year. After 1968 whole-body bremsstrahlung were measured periodically. Tissues and bones of dead monkeys were analyzed for  $^{90}\text{Sr}$ .

Samples were dry-ashed, dissolved in acid, and analyzed for  $^{90}\text{Sr}$  by one of the following methods: (a) samples of  $> 10^4$  dis/min—small aliquots were dried on glass planchets; (b) samples of  $10^2$ - $10^4$  dis/min—large aliquots were concentrated by coprecipitation with  $\text{CaC}_2\text{O}_4$ ; <sup>12</sup> and (c) samples of  $< 10^2$  dis/min—large aliquots or entire samples were analyzed by a commercial laboratory using a  $^{90}\text{Y}$ -extraction process. <sup>12</sup>

## RESULTS AND DISCUSSION

We have chosen to describe plasma activity,  $P_t$ , daily excretion rate,  $E_t$ , cumulative excretion,  $E_t^*$ , and whole-body retention,  $R_t$ , as sums of first-order exponentials, of the form

$$A_t = \sum_{i=1}^{i=n} A_i \exp(-0.693t/Ta_i). \quad (1)$$

Half-times,  $Ta_i$ , and time after injection,  $t$ , are in days. The data for  $P_t$  and  $E_t$  were plotted semi-logarithmically for each monkey. Curves were fitted by the method of least squares in 6 straight-line segments over the intervals shown in Table 2, and the coefficients of their exponential equations,  $P_i$  (%) and  $E_i$  (%/d), and the half-times,  $Tp_i$  and  $Te_i$ , were determined graphically. Cumulative excretion was calculated for each monkey by integration of its equation of  $E_t$  at the postinjection times shown by the data points in Fig. 1. Whole-body retention was calculated from the relationship

$$R_t (\%) = (100\% - E_t^*) = (100\% - \int_0^t E_t dt). \quad (2)$$

Tabulations were made of the values of  $P_t$  and  $E_t$  read from the individual monkey's curves and of their calculated values of  $R_t$  at the times shown by the data points in Figs. 1-3. Mean values of  $P_t$ ,  $E_t$ , and  $R_t$  (the data points shown in the figures) were calculated for the 3 age groups. The individual values usually varied over a 2- to 4-fold range. The mean curves shown in the figures were also analyzed into 6 exponential terms, and their coefficients and half-times are collected in Tables 2-4.

Total excretory clearances of the 3 monkey groups (see curves of  $P_t$  and  $E_t$  in Figs. 2,3) ranged over 4.5-9 plasma volumes/d and tended to be lower in the younger monkeys. The best current estimate of excretory clearance of  $^{90}\text{Sr}$  in adult man<sup>9</sup> is 4.3 plasma volumes/d (13.6 liters/d for a 70-kg man with an average daily intake of 1 g Ca, 0.014 g Ca/kg body weight). The greater clearance in the monkeys is probably related to their 2- to 10-fold greater Ca intake.

The half-times of the comparable terms of the equations of  $P_t$ ,  $E_t$ , and  $R_t$  (Tables 2-4) were similar within the 3 age groups, which is to be expected, because retention, plasma activity, and excretion rate are all dependent on bone metabolism. However, in the equations of  $R_t$  for the adolescent and adult monkeys,  $Tr_6$  was 4200 to 5600 d, while in their equations of  $P_t$  and  $E_t$ ,  $Tp_6$  and  $Te_6$  were 2400 to 3640 d. Although the data are insufficient to demonstrate a significant difference, such a finding is compatible with redeposition in the skeleton of some  $^{90}\text{Sr}$  recirculated during bone resorption.

The coefficients,  $P_{1-3}$ ,  $E_{1-3}$ , and  $R_{1-3}$ , representing  $^{90}\text{Sr}$  uptake in bone, are smaller in the metabolic equations of the young monkeys than in those of the adults. The coefficients,  $P_{4-6}$ ,  $E_{4-6}$ , and  $R_{4-6}$ , representing turnover of  $^{90}\text{Sr}$ -labeled bone, are greater in the equations of the young monkeys than in those of the adults. The relative magnitudes of the coefficients of the equations of  $P_t$ ,  $E_t$ , and  $R_t$  in the 3 age groups demonstrate again the dependence of  $^{90}\text{Sr}$  uptake and retention on bone-growth status. <sup>15-18</sup>

The half-times of 6-term exponential equations of  $^{90}\text{Sr}$  retention in young-adult beagles<sup>14</sup> and a 57-yr old woman<sup>7</sup> were similar to those in the retention equations of the monkeys, when all retention curves were analyzed over the same postinjection intervals (Tables 2,4). The finding of common half-times in exponential equations of  $^{90}\text{Sr}$  retention in monkeys with varying bone-growth status, young-adult beagles, and a middle-aged woman supports the view that the dynamics of skeletal metabolism are qualitatively similar among mammals.<sup>15</sup>

If age at injection is considered,  $^{90}\text{Sr}$  retention in our monkeys agrees with the retention of  $^{85}\text{Sr}$  or  $^{90}\text{Sr}$  in young and adolescent rhesus monkeys studied elsewhere,<sup>19</sup> young and adolescent monkeys of 3 species of *Cercopithecus* (West African monkeys of small body size),<sup>20,21</sup> 1.4- to 9-yr-old beagles,<sup>14,16,18</sup> and in adult human subjects during the first 100 d after injection. For example, 15 d after injection, retention or  $^{85}\text{Sr}$  of  $^{90}\text{Sr}$  was  $27 \pm 9\%$  and  $34.3 \pm 10.2\%$ , respectively, in 8 adult rhesus monkeys and 9 human beings 27-61 years old.<sup>7, 22-24</sup> By 350 d, retention of  $^{85}\text{Sr}$  in 9 human adults

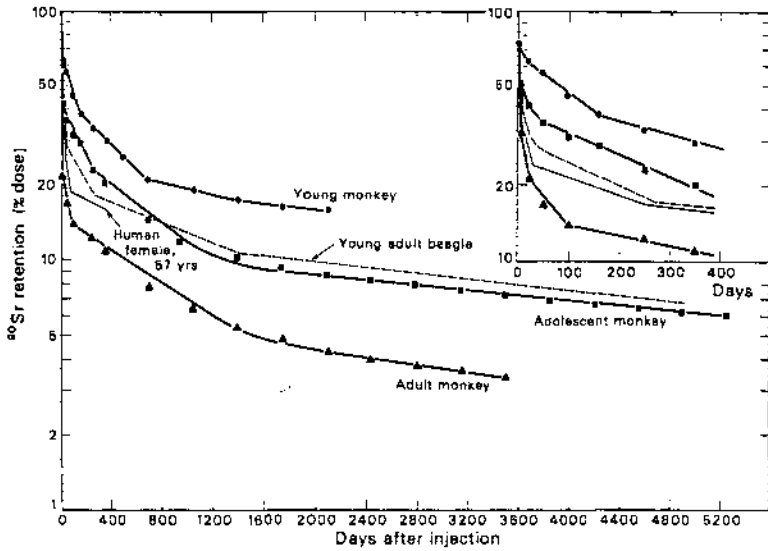


Fig. 1. Retention of a single injection of  $^{90}\text{Sr}$  in rhesus monkeys, young-adult beagles,<sup>14</sup> and a 57-yr-old woman.<sup>7</sup> Parameters of the exponential equations appear in Table 4.

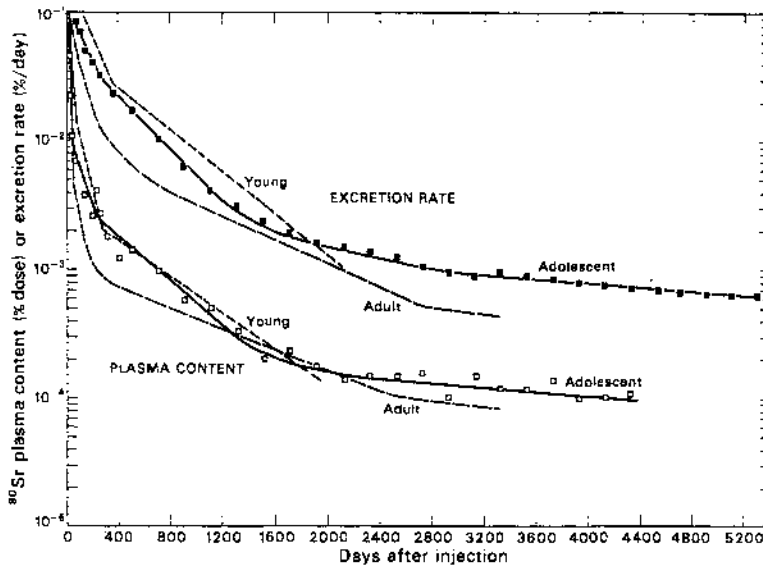


Fig. 2. Plasma content and excretion rate (urine and feces combined) after a single injection of  $^{90}\text{Sr}$  in rhesus monkeys. Parameters of the exponential equations appear in Tables 2,3.

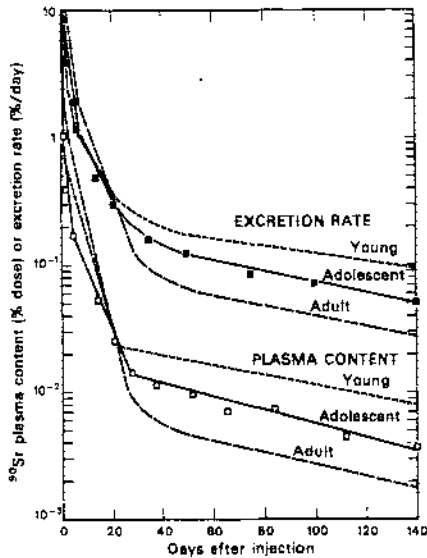


Fig. 3. Expanded time scale of Fig. 2.

Table 2. Plasma content: Coefficients ( $P_i$ ) and half-times ( $TP_i$ ) of the exponential equations that describe the  $^{90}\text{Sr}$  content of the plasma<sup>a</sup> after a single injection in rhesus monkeys.

Exponential term	Fitting interval		Young		Adolescent		Adult	
	$t_1$ (d)	$t_2$ (d)	$t_f = 1900$ d <sup>b</sup>		$t_f = 4600$ d <sup>b</sup>		$t_f = 3450$ d <sup>b</sup>	
			$P_i$ (%)	$TP_i$ (d)	$P_i$ (%)	$TP_i$ (d)	$P_i$ (%)	$TP_i$ (d)
1 <sup>c</sup>	0	1	—	—	—	—	—	—
2	1	5	1.0	.65	1.2	1.1	1.7	.53
3	5	25	.88	4.4	.28	5.8	1.8	3.3
4	25	250	.026	60	.019	51	.007	40
5	250	1500	.0032	420	.0038	300	.0008	640
6	> 1500	—	—	—	.00024	3500	.0002	2400

a Total plasma  $^{90}\text{Sr}$  was calculated from  $^{90}\text{Sr}$  in weighed samples of whole blood and the measured hematocrit, assuming a blood density of 1.056g/ml and a plasma volume of 36.4 ml/kg body weight in rhesus monkeys.<sup>13</sup>

b Time of last available measurement.

c The earliest components of the blood curves have not yet been analyzed.

Table 3. Excretion: Coefficients ( $E_i$ ) and half-times ( $TE_i$ ) of the exponential equation of the rate of  $^{90}\text{Sr}$  excretion, and intercepts ( $E_i^*$ ) of the equations of cumulative excretion of  $^{90}\text{Sr}$  after a single injection in rhesus monkeys.

Exponential term	Young			Adolescent			Adult		
	$t_f = 1900$ d			$t_f = 5250$ d			$t_f = 3300$ d		
	$E_i$ (%/d)	$TE_i$ (d)	$E_i^*$ (%)	$E_i$ (%/d)	$TE_i$ (d)	$E_i^*$ (%)	$E_i$ (%/d)	$TE_i$ (d)	$E_i^*$ (%)
1 <sup>a,b</sup>	16	.40	9.2	41	.46	27	31	.42	19
2	5.9	1.7	14	7.9	1.3	15	21	1.2	36
3	1.5	7.1	15	2.0	6.5	19	4.1	5.5	32
4	.19	80	22	.094	41	5.5	.091	88	12
5	.053	360	28	.058	300	25	.0086	570	7.2
6	—	—	—	.0017	3640	8.9	.0011	2400	3.8

a The parameters of the first terms of the equations of  $E_t$  were obtained from curves in which 0-1 d excretion was plotted at 0.5 d, and fecal lag was neglected. Such curves artificially depict constant renal excretion during the first 24 h, and gastrointestinal excretion as both constant and instantaneous.

b Same fitting intervals as in Table 2.

Table 4. Retention: Coefficients ( $R_i$ ) and half-times ( $TR_i$ ) of the exponential equations of whole-body retention of  $^{90}\text{Sr}$  after a single injection in rhesus monkeys. Parameters of  $^{90}\text{Sr}$  retention curves of young-adult beagles and of a woman are shown for comparison.<sup>a</sup>

Exponential term	Monkey								Human subject	
	Young		Adolescent		Adult		Beagle			
	$t_f = 2100$ d		$t_f = 5250$ d		$t_f = 3450$ d		$t_f = 4900$ d		$t_f = 397$ d	
	$R_i$ (%)	$TR_i$ (d)	$R_i$ (%)	$TR_i$ (d)	$R_i$ (%)	$TR_i$ (d)	$R_i$ (%)	$TR_i$ (d)	$R_i$ (%)	$TR_i$ (d)
1	11	.50	23	.38	28	.41	19	.6	22	.44
2	10	2.1	18	2.0	24	2.2	32	2.7	31	2.5
3	11	17	17	10	24	7.0	18	14	22	14
4	20	66	14	100	9.2	22	11	120	4.6	130
5	26	300	17	500	8.8	540	6.7	540	22	1000
6	21	5400	11	5600	6.0	4200	13	5300	—	—

a Data from LLOYD *et al.*<sup>14</sup> for beagles injected with  $<30 \mu\text{Ci/kg}$  of  $^{90}\text{Sr}$ , and data from COHN *et al.*<sup>7</sup> for a 57-yr-old woman patient MOS were replotted, and the retention curves were fitted over the same intervals (Table 2) used to analyze the monkey curves.

(some ill but with apparently normal skeletons) was  $15.9 \pm 4.7\%$ <sup>7,23</sup>--apparently greater than in 5 adult monkeys,  $11 \pm 4.3\%$ . However, by 350 d, decay and excretion of <sup>85</sup>Sr introduce large errors in whole-body  $\gamma$ -ray measurements, which, added to individual variability, render any difference insignificant. Thus, rhesus monkeys of varying ages seem to be a suitable model system in which to evaluate long-term metabolism of the alkaline earth elements.

#### ACKNOWLEDGMENTS

The indispensable assistance of the Lawrence Berkeley Laboratory animal care staff, W. Moore, Jr., D. Brooks, R. Frazier, and F. Crenshaw, and the Technical Editor, Mary Wildensten, are gratefully acknowledged.

#### FOOTNOTES AND REFERENCES

\*Prepared for the U.S. Atomic Energy Commission under Contract No. W-7405-eng-48.

\*\*Present address: Oregon State Board of Health, Portland, Oregon 97201.

1. R.A. DUDLEY, Biological hazard of radioactive strontium, in: *Proc. 2nd Ann. Conf. on Plutonium, Radium and Mesothorium*, Salt Lake City, Utah, 1954, U.S.A.E.C. Doc. TID-7639.
2. A. ENGSTROM, R. BJÖRNERSTEDT, C.-J. GLEMEDSON, and A. NELSON, *Bone and Radiostrontium*. John Wiley and Sons, Inc., New York (1957).
3. U.S. CONGRESS, JOINT COMMITTEE ON ATOMIC ENERGY, Hearings on fallout from nuclear weapons tests, May 27-29, June 3, 1957. U.S.G.P.O., Washington, D.C.
4. P.W. DURBIN, M.W. PARROTT, M.H. WILLIAMS, M.E. JOHNSTON, C.W. ASLING, J.G. HAMILTON, N. JEUNG, and S.A. COLE, Metabolic studies with strontium-90 in the rhesus monkey, in: *The Shorter-Term Biological Hazards of a Fallout Field*. (Edited by G.M. DLNNING and J. A. HILCKEN), p. 173-187, Proc. of conf., Washington, D.C. U.S.G.P.O., Washington, D.C.
5. P. DURBIN and H. JONES, Estimation of the turnover equation of strontium-90 for human bones, in: *Proc. 2nd Int. Conf. Peaceful Uses of Atomic Energy*, United Nations, Geneva. 23, 140-141 (1958).
6. P.W. DURBIN, M.H. WILLIAMS, M.W. PARROTT, and H.B. JONES, Retention of Sr<sup>90</sup> and Ca<sup>45</sup> in rhesus monkeys, *Radiology* 72, 98-99 (1959). Abstract.
7. S.H. COHN, H. SPENCER, J. SAMACHSON, and J. ROBERTSON, The turnover of strontium-85 in man as determined by whole-body counting, *Radiat. Res.* 17, 173-185 (1962).
8. J.L. KULP, W.R. ECKELMANN, and A.R. SCHULERT, Strontium-90 in man, *Science* 125, 219-225 (1957).
9. INTERNATIONAL COMMISSION ON RADIATION PROTECTION, *Alkaline earth metabolism in adult man*, Report of Task Group of Committee II, J.H. MARSHALL, chmn., ICRP Publication 20, Pergamon Press, Oxford (1973).
10. V.O. HURME and G. VAN WAGENEN, Basic data on the emergence of permanent teeth in the rhesus monkey (*Macaca mulatta*), *Proc. Am. Phil. Soc.* 105, 105-140 (1961).
11. G. VAN WAGENEN and C.W. ASLING, Roentgenographic estimation of bone age in the rhesus monkey (*Macaca mulatta*), *Am. J. Anat.* 103, 163-186 (1958).
12. D.N. SLNDERMAN and C.W. TOWNLEY, *The radiochemistry of barium, calcium, and strontium*, Nucl. Sci. Series, NAS-NS-3010 (1960).
13. M.I. GREGERSON, H. SEAR, R.A. RAWSON, S. CHIEN, and G.L. SAIGER, Cell volume, plasma volume, total blood volume, and F<sub>cells</sub> factor in the rhesus monkey, *Am. J. Physiol.* 196, 184-187 (1959).
14. R.D. LLOYD, C.W. MAYS, and G.N. TAYLOR, Retention and skeletal dosimetry of injected <sup>90</sup>Sr in beagles, Radiobiol. Lab. Univ. of Utah, Report C00-119-241, p. 79-96 (1970).
15. E.A. GUSMANO, J.N. CONCANNON, S.R. BOZZO, and S.H. COHN, Evaluation of the parameters of strontium metabolism in the rat as a function of age, *Radiat. Res.* 33, 540-553 (1968).
16. B.W. GLAD, C.W. MAYS, and W. FISHER, Strontium studies in beagles, *Radiat. Res.* 12, 672-681 (1960).
17. T.W. SPECKMAN and W.P. NORRIS, The age dependence of strontium retention in rats and mice, *Radiat. Res.* 23, 461-474 (1964).
18. C.F. DECKER, L.V. KASPAR, and W.P. NORRIS, The variation of strontium metabolism with age in the dog, *Radiat. Res.* 23, 475-490 (1964).
19. S.A. GÖKSEL, The toxicity of strontium-90 in the monkey skeleton, Thesis, Univ. of Rochester (1962).
20. A.H. WARD, Retention and excretion of radiostrontium in monkeys, *J. Nucl. Energy* 5, 192-202 (1957).
21. A.H. WARD, Retention and toxicity of radiostrontium in monkeys, in: *Radioisotopes in Scientific Research*. Proc. of UNESCO conf., Paris, 1957 (Edited by R.C. EXTERMANN) 4, p. 221-231. Pergamon Press, Oxford.
22. M.A. VAN DILLA, S. WALLACH, and J.S. ARNOLD, Sr<sup>85</sup> tracer studies in humans, Radiobiol. Lab., Univ. of Utah, Prog. Rep., U.S.A.E.C. Doc. TID-16459, p. 48-58 (1956).
23. M. BISHOP, G.E. HARRISON, W.H.A. RAYMOND, A. SUTTON, and J. RUNDO, Excretion and retention of radioactive strontium in normal men following a single intravenous injection, *Int. J. Radiat. Biol.* 2, 125-142 (1960).
24. G.E. HARRISON, T.E.F. CARR, and A. SUTTON, Distribution of radioactive calcium, strontium, barium, and radium following intravenous injection into a healthy man, *Int. J. Radiat. Biol.* 13, 235-247 (1967).

A STUDY OF THE TRANSLOCATION OF RADIOSTRONTIUM  
FROM WOUNDS AND THERAPY BY LOCAL INSOLUBILIZATION

Roger DUCOUSSO, André CAUSSE and Christian PASQUIER.

Groupe de Recherche du Service de Santé des Armées  
auprès du Commissariat à l'Energie Atomique.

92260 - Fontenay-aux-Roses - FRANCE

Abstract

The translocation of  $^{85}\text{Sr}$  was followed by a 2 h external counting of the wound and homologous bone and by measurement of the blood radioactivity. Two types of wounds were simulated on Maccacus monkeys : puncture wounds and lacerations.

The same experimental procedure was applied in order to test therapy through Sr insolubilization by rhodizonate (K),  $\text{Mg SO}_4$ , alginate (Ca) and aluminium phosphate gel, on laceration wounds either 5 or 15 min after the contamination.

It was verified that local or IV administration of DTPA did not result in an increased absorption of Sr.

Introduction

Wounds can be classified into three classes : abrasion, laceration and puncture. The first will not result in any significant absorption after contamination<sup>1</sup> ; the second one only can be treated by local insolubilization of the radionuclides. Most authors have studied wounds contaminated by untranslocable nuclides (Pu and Am), whereas our study was concerned with strontium, a translocable fission product with a high radiotoxicity. For safety purposes and easier measurement,  $^{85}\text{Sr}$  was chosen.

Material and methods

Twenty eight Maccacus monkeys weighing 5 kg were used. Wounds were performed on the postero-external side of the thigh. Puncture wounds were simulated by IM injections ; lacerations were made by incision and ecrasement of the muscle 2 cm long and 2.5 cm wide. A catheter was introduced into the arteria femoralis of the opposite leg for blood samples. From 10 to 100  $\mu\text{Ci}$  of  $^{85}\text{Sr}$  ( $0.1 \text{ cm}^3$ ) were deposited on the wound and the radioactivity of the wound and the homologous area on the other leg was measured by NaI (Tl) detectors with the same

counting efficiency. The radioactivity of the blood samples (1 cm<sup>3</sup>) was measured in a well-type crystal. The various treatments were applied 5 or 15 min after exposure.

Results

Effect of the type of wound on absorption.

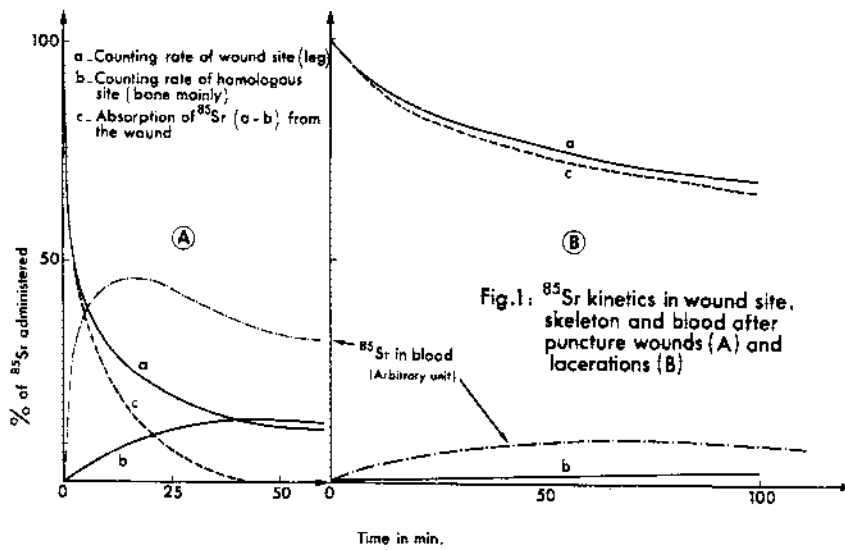


Fig. 1: <sup>85</sup>Sr kinetics in wound site, skeleton and blood after puncture wounds (A) and lacerations (B)

Following puncture, all the Sr was absorbed within 40 min (fig. 1 A, average on 2 animals). Blood concentration was highest within 20 min, then slowly decreased. Bone uptake seemed to stop after 50 min's time. Following laceration (fig. 1 B, average on 6 animals) these processes were slower and not so complete.

Sr insolubilization tests on lacerations. (17 animals).

As shown by fig. 2, therapy must be early : when administered 5 min after exposure, all the insolubilizing reagents resulted in a more or lest significant

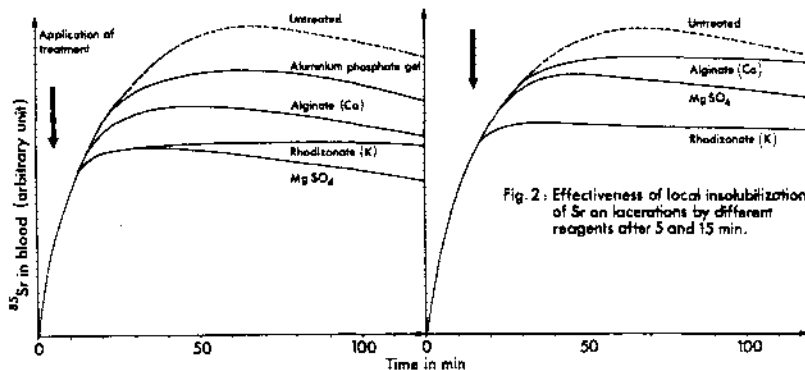
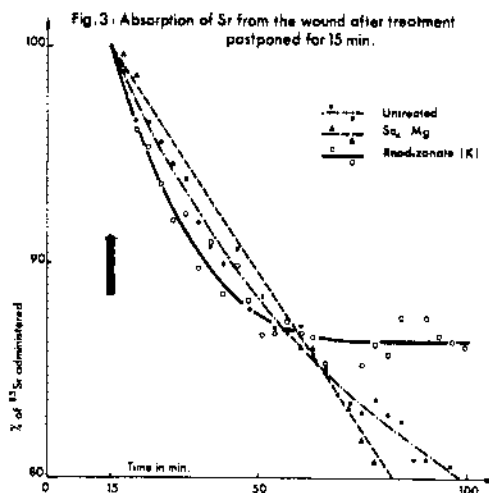


Fig. 2: Effectiveness of local insolubilization of Sr on lacerations by different reagents after 5 and 15 min.

decrease of absorption ; when administration occurred 15 min later, only K rhodizonate (powder) and Mg SO<sub>4</sub> (saturated solution) had a significant effect. Counting of the wound showed rhodizonate to be more efficient (fig. 3), which was verified by measurement of urinary excretion (table 1).



Effect of DTPA on Sr absorption. (3 animals).

In case of wounds contaminated by a mixture of fission products, DTPA (used as DTPA Ca Na<sub>3</sub>) can be used in order to chelate the lanthanons. Table 1 shows that Sr absorption was not enhanced by DTPA whether insolubilized or not.

Treatment after 15 min	Absorption from the wound % of deposit	Urinary excretion during 24 h. % of deposit
Untreated	22	7
Mg SO <sub>4</sub>	33	2,5
Rhodizonate	10	1,5
DTPA	23	5
Rhodizonate + DTPA	10	1
Rhodizonate + DTPA + DTPA (iV)	20	2,5

Table.1: Influence of treatment on absorption and urinary excretion of Sr deposited on laceration



### Discussion and conclusion

Lacerations are very difficult to standardize and result in widely dispersed data (slide number 5).

As a conclusion, laceration wounds contaminated by Sr can be treated by insolubilization whereas puncture wounds cannot, because of access and delay. Rhodizonate seemed to be the best insolubilizing reagent. DTPA did not act on Sr absorption.

### References

1. SCHOFIELD (G.B.) - Clin. Rad., 15, 1, 50-54 (1963).

## OPERATIONAL HEALTH PHYSICS - II

### INSTRUMENTS AND METHODS FOR DAY-TO-DAY MONITORING OF TRITIUM AND CARBON-14 IN PRODUCTION

M.I.Balonov, E.I.Dolguirev, A.S.Karasik, O.N.Prokofiev

Institute of Radiation Hygiene

The RSFSR Ministry of Public Health

Leningrad

USSR

#### Abstract

A complex of instrumentation and techniques for dosimetric monitoring at industrial enterprises using unshielded beta-emitters of T and  $^{14}\text{C}$  is offered. Air-borne radionuclides are assayed by automatic monitors developed on the basis of gas counters. A portable device for detection of radioactive contamination of surfaces is worked out. Scintillation and gas counters have been used to measure  $^{14}\text{C}$  and T content in personnel. The sensitivity of the methods is sufficient for monitoring of all categories of exposure.

#### Introduction

The problems of protection from tritium and  $^{14}\text{C}$  radiation arise in the manufacture and application of tracer compounds, luminophores, tritium-titanium targets as well as in spheres involving the use of the energy of nuclear fission and fusion. Certain difficulties, as far as the dosimetry of these radionuclides is concerned, are due both to their specific radiation characteristics and insufficient knowledge of the radiotoxicity of their compounds<sup>1,2</sup>.

This paper does not deal with the biophysical aspects of the indirect dosimetry of T and  $^{14}\text{C}$ ; it describes instrumentation and methods for detecting low-energy beta-emitters in the human body and environment.

#### Monitoring of Radionuclides in Air

Gas filling counters are employed as detecting devices in all the monitors developed. The instruments also incorporate devices for passive and active shielding of the counters from external radiation, systems for intake, processing and regulation of the flows of assayed air and filling gas, electronic registering systems, control and signalling assemblies as well as high- and low-voltage power supplies for the counters and transistorized circuits, respectively. The instruments are designed as portable double-tier columns with detecting device at the bot-

tom and gas systems and electronic blocks at the top.

A mixture of aerosol-free air (0.1 l/min) and methane (0.5 - 1 l/min) is fed to the detectors of instruments A and B. The air piping and proportional counter bodies are made of Teflon. The operating volume of Counter A (Fig.1a) of 0.15 l is limited by a cylindrical cathode consisting of 22 metal wires.<sup>3</sup> The de-

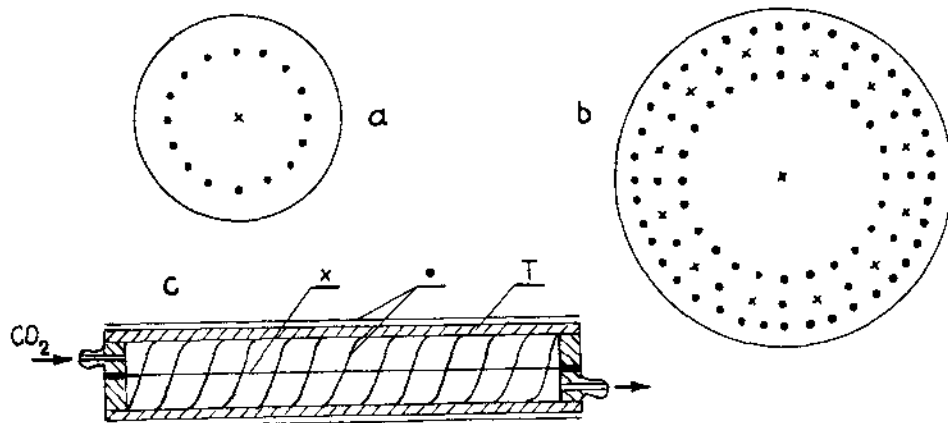


Fig. 1. Schematic constructions of gas-flow counters.  
x - anodes; . - cathodes; t - quartz tube.

detector and pre-amplifier are surrounded by a 5 cm thick lead shield. Pulses are registered by an integral discriminator with a sensitivity of 0.5 mV. To calibrate counters A and B, a flow of methane bubbler-saturated with T-ethanol vapours is passed in succession through the multi-wire counter and a calibration counter with known parameters. The counting efficiency is calculated with due regard to the ratio of the detector volumes and the known efficiency of the calibration counter.

The detection efficiency for T and  $^{14}\text{C}$  beta-radiation for counter A operation in air-free methane is over 90%. A 500V - long counting plateau has no appreciable slope. Introduction of air affects the counting characteristics considerably. For a 5% admixture of air, the plateau length is reduced to 250V and its slope is 2%/100 V. The count curve for a mixture containing 10% of air in the region of 3.4 to 3.6 kV has a slope of 8%/100 V. The efficiency of beta particle detection in the working point (3.5 kV) is 85%. The count curve becomes still less stable as the air portion increases. The counter background of 150-180cpm remains stable up to 4.0 kV. The detector and gas system are cleaned from THO vapours and gaseous tritium exponentially ( $\lambda = 1 \text{ min}^{-1}$ ), from the moment radioactivity intake has ceased.

The 0.5 l counter B with a multi-wire cathode is enclosed in a ring of protection counters located in the same volume of gas (Fig.1b). A two-channel panel selects coincidence and anticoincidence pulses.

The counter B anticoincidence background is 35 cpm in methane, 70 cpm in 10% air mixture and 85 cpm at 20% content of air, respectively. The count curves for T and  $^{14}\text{C}$  have a plateau slope of about 5%/100V at 10% and 20% air in the mixture. The counting efficiency is about 90%.

The assay of the energy composition of air-borne radioactivity is based on the correlation between the number of coincidence counts in the measuring and protection counters and the path of

primary ionizing particles<sup>4</sup>. Thus, the ratio of coincidence and anticoincidence counts for T is 0.1, whereas for <sup>14</sup>C - 1.6. The counter B measurement results may be used for estimating the ratio of nuclide concentrations in air.

Counter C (Fig.2) is intended for a selective determination

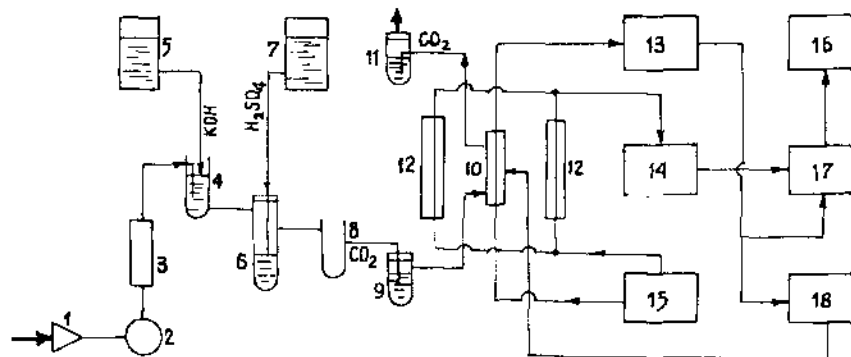


Fig.2. Block-diagram of air-borne <sup>14</sup>C monitor.  
 1 - air intake; 2 - compressor; 3 - oven; 4 - bubbler;  
 5 - vessel with KOH; 6 - reaction vessel; 7 - vessel with  
 H<sub>2</sub>SO<sub>4</sub>; 8 - dessicator; 9 - bubbler with cyclohexan; 10 - CO<sub>2</sub>-  
 counter; 11 - liquid seal; 12 - shielding counters; 13,14 -  
 input blocks; 15 - high voltage block; 16 - recorder; 17 -  
 measuring block; 18 - electronic suppressor.

of air-borne as carbon dioxide or carbonate aerosols. Air under assay is bubbled through a flow of a 10% solution of KOH to absorb CO<sub>2</sub>. The solution of KOH and K<sub>2</sub>CO<sub>3</sub> is passed to a reaction vessel with a 25% solution H<sub>2</sub>SO<sub>4</sub>. Carbon dioxide resulting from the reaction:  $K_2CO_3 + H_2SO_4 \rightleftharpoons K_2SO_4 + H_2CO_3$ ;  $H_2CO_3 \rightarrow H_2O + CO_2 \uparrow$  is passed through a desiccator and a quenching agent (cyclohexan) bubbler to the counter. CO<sub>2</sub> acts both as an operating gas in Geiger counter and a radiocarbon carrier.

The cathode of the CO<sub>2</sub> -counter is provided with a 0.3 mm-dia. platinum wire coil wound at a pitch of 5 mm inside the inner surface of a 18 mm-dia. quartz tube (Fig.1c). The counter volume is 30 cm<sup>3</sup>; the anode diameter 0.1 mm.

The count curve in measuring crude CO<sub>2</sub> is quite good due to the incorporation of an external suppressor assembly with a pulse height of 1.5 kV, duration 2 msec. The plateau length is 400 V; the slope - 2%/100 V; the working point - 3600 V. The background of the counter surrounded by a 5 cm-thick lead shield at anticoincidence with a ring of shielding Geiger counters is 4-5 cpm. The counter radiometric characteristics for air monitoring are given in the Table. The minimal detectable concentration (MDC) in the atmospheric air is defined as a level capable of inducing a count rate equal to that of the background. The simplest monitor A ensures a reliable detection of maximum permissible concentrations for personal of THO vapours (5.10<sup>-9</sup> Ci/l<sup>6</sup>) and <sup>14</sup>C compounds (3.5 x 10<sup>-9</sup> Ci/l). The model B is capable of detecting concentrations lower than the above by an order as well as differentiating beta - radiation with respect to energy. The provision for the chemical selec-

Table of Parameters of Air-Borne Radioactivity Monitors

Counter	Gas to be assayed	Sample volume, cm <sup>3</sup>	Background cpm	MDC, Ci/l of air	Response time min.
A	Air	15	150	$5 \times 10^{-9}$	1
B	Air	100	85	$4 \times 10^{-10}$	5
C	CO <sub>2</sub>	30	4	$2 \times 10^{-14}$	30

tion of CO<sub>2</sub> makes counter C applicable for monitoring of air-borne <sup>14</sup>C for all categories of exposure<sup>6</sup>. These instruments intended for operation under various conditions offer a still wider range of uses:

- (a) automatic recording and signalling of emergency situations;
- (b) simultaneous sampling of air from several checking points;
- (c) differentiation of gaseous tritium and THO vapours by means of the desiccator assembly;
- (d) continuous catalytic combustion of <sup>14</sup>C and tritium-labelled organic compounds in air to form CO<sub>2</sub> and THO.

#### Monitoring of Radioactive Contamination of Surfaces

National standards treat of radioactive contamination of surfaces in terms of particle output from a unit of area regardless of radiation energy<sup>6</sup>. To comply with this, the efficiency of our portable instrument hardly depends on radiation energy. Detection is effected by a four-section gas-flow proportional counter with an open window (Fig.3). Anode wires are designed to run parallel to the examined surface. The outside cathode made of 0.6x0.6mm mesh brass net is separated by a 2mm clearance from the surface.

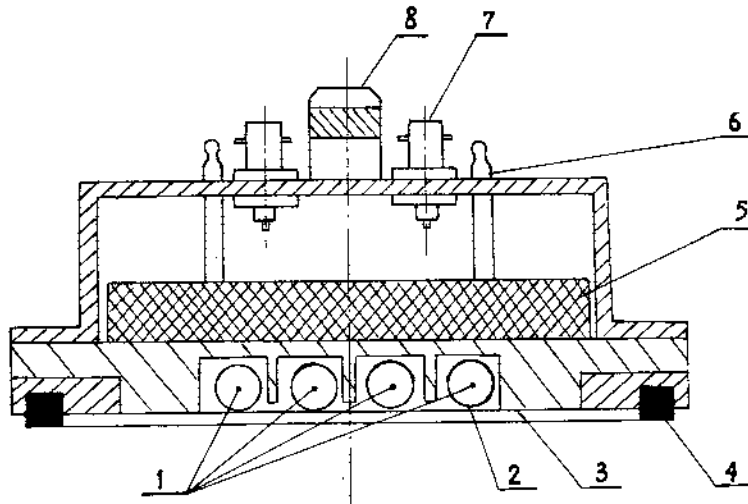


Fig.3. Surface contamination detector.

1-anodes; 2-insulators; 3-net; 4-rubber ring; 5-lead disk; 6-gas connection pipe; 7-plug connector; 8-grip.

The sensitivity area is 40 cm<sup>2</sup>. The detector comprises a transistor pre-amplifier of signals. The gas (methane) flow rate is less than 0.5 l/min.

The measuring panel incorporates an integral discriminator,

a rate meter, a pointer indicator and a power supply.

Solid sources of beta radiation of tritium,  $^{14}\text{C}$  and  $^{137}\text{Cs}$  are used for calibration purposes. Plateaux of 200 to 300 V length with a slope of 5%/100 V have been obtained. The counting efficiencies at the point of operation are: 70% - for tritium, 80% - for  $^{14}\text{C}$  and 90% - for  $^{137}\text{Cs}$ . The proximity of the above values is due to the peculiarity of open-window counter operation, which detect a great portion of ionization electrons released in the gas layer bounded by the examined surface and the cathode. The counter background is 6 cps. The instrument reliably detects contamination under 0.01 of the maximum permissible levels for operation surfaces ( $2.000 \text{ particle/cm}^2 \text{ min}^6$ ).

#### Measurement of Tritium and $^{14}\text{C}$ Content in Human Body

The internal contamination of the human body induced by such beta-emitters as T and  $^{14}\text{C}$  may be determined only indirectly on the basis of the results of biological sample assays.

T and  $^{14}\text{C}$  concentrations in the body liquid samples and exhaled air were measured by scintillation and gas counters. Samples of urine, saliva, blood and exhaled vapour condensate were mixed with a liquid scintillator containing 8 g PPO + 0.2g POPOP + 100 g naphthalene per 1 l dioxane. Different methods of preparation of tritium-containing samples with respect to the degree of purification, such as distilled, activated coal-treated (urine only) and untreated samples, were tested. Precipitates were separated by filtration.

The beta-radiometry of the samples was carried out on a scintillation coincidence counter with two venetian-blind photomultipliers Ф3Y-81A. A 30 ml cuvette for samples is made of Teflon and provided with quartz windows. The electronic recording system selects time-coinciding (within 100 nsec) pulses fed from the two photomultipliers and analyses them with respect to amplitude by means of a differential discriminator with a threshold ratio of 10:1. The measurements were carried out under balance conditions. The instrument was calibrated with the aid of a solid emitter of X-rays simulating tritium radiation<sup>7</sup>.

The best sensitivity is ensured in measuring samples containing 20% water or 8 to 10% urine, with the tritium counting efficiency being 12 to 15%; the counter background is 50 cpm. The sensitivity threshold to tritium in water is  $3 \cdot 10^{-9}$  and in untreated human urine -  $7 \cdot 10^{-9} \text{ Ci/l}$ , respectively (measurement time - 30 min; relative error - 25%).

An analysis of the instrument background shows that 65% is contributed by internal processes occurring in the photomultipliers and generating light impulses; 30% - by radioactive and cosmic radiation, and 5% - by random coincidence of dark current pulses and the phosphorescence of samples.

The minimal detectable concentration of tritium is four orders below the "initial levels of the examination" of personnel in contact with tritium oxide<sup>8</sup>. The scintillation counter sensitivity to  $^{14}\text{C}$  concentration in urine or water samples is  $10^{-9} \text{ Ci/l}$  which is quite sufficient for the purposes of industrial dosimetry.

Another modification of the indirect dosimetry method is provided by the measurement of THO vapours and  $^{14}\text{CO}_2$  in exhaled air by means of gas monitors of types B and C. The air to be assayed is continuously sampled from a through-flow vessel where exhaled air comes. The  $\text{CO}_2$  counter is capable of measuring  $^{14}\text{C}$  concentrations up to  $2 \cdot 10^{-12} \text{ Ci/l}$  in exhaled air ( $\text{CO}_2$  content - 3%). This makes it possible for  $^{14}\text{C}$  excretion to be reliably monitor-

ed during one week after one "initial level of the examination" of  $\text{NaH}^{14}\text{CO}_3$  incorporation. During the first days after exposure this level can be detected by instrument B as well. For a similar THO incorporation the body fluids tritium concentration is  $35 \mu \text{Ci/l}^8$ , whereas this parameters for exhaled air is  $1.5 \cdot 10^{-9} \text{Ci/l}$  at the body temperature. However, when air is transferred to the detector, the air temperature is equalized to that of the device parts. This leads to the condensation of some vapours and the residual concentration of THO in the assayed air is determined by the ambient temperature. Thus,  $5 \cdot 10^{-10} \text{Ci/l}$  of exhaled air corresponds to the "initial level of the examination" at  $20^\circ\text{C}$ . This concentration is reliably detected by counter B.

So, the above complex of instrumentation and techniques guarantees an all-round monitoring of exposure conditions of operation in contact with unshielded tritium and radiocarbon compounds. Depending on the process used and scale of production of different radioactive substances, the dosimetrical service may be supplied with some of the instruments of this complex or a suitable combination of them. The counters have been tested for a long period of time at the works of manufacturers specializing in the large-scale production of various radionuclide items, such as labelled organic and inorganic substances, luminous compounds and devices, metallic targets with tritium, etc. The test results have shown the described equipment and methods to comply with the requirements of day-to-day dosimetry.

#### References

1. "Assessment of Air-Borne Radioactivity", IAEA, Vienna, 1967.
2. Vennart J. *Hlth. Phys.* 16, 429 (1969).
3. Балонов М.И. и др. Приборы и техника эксперимента, 1973, в печати.
4. Ehret R. In "Assessment of Air-Borne Radioactivity", IAEA, Vienna, p. 563.
5. Карасик А.С. Приборы и техника эксперимента, № 2, 68 (1966).
6. "Нормы радиационной безопасности НРБ-69", Москва, Атомиздат, 1970.
7. Балонов М.И. Приборы и техника эксперимента, № 4, 82 (1971).
8. Recommendations of ICRP, Publication 10, Pergamon Press, 1968.

## SAMPLING FOR TRITIATED WATER VAPOUR

by

R.V. Osborne

Biology and Health Physics Division, Chalk River Nuclear Laboratories  
Atomic Energy of Canada Limited, Chalk River, Ontario, Canada.

### ABSTRACT

Collection of tritiated water vapour  $(\text{HTO})_v$  from air by bubbling the air through non-tritiated water and subsequent measurement of the accumulated activity by liquid scintillation counting is often used as a simple method of monitoring for  $(\text{HTO})_v$  in air.

Expressions for the collection efficiency of a model collecting device of this kind are derived here in terms of the initial mass of water in the collector, the total air flow, the humidity in the air entering and leaving the collector, the intrinsic efficiency of the collector and the ratio of the relative isotopic concentration  $[T/H]$  in the vapour phase to that in the liquid phase at the effective operating temperature of the device.

Predictions from the model are compared to the measured efficiencies of collectors with and without fritted glass air dispersers, for water masses from 50 g to 200 g, air volumes 0.1 to 20 m<sup>3</sup>, air flow rates up to 160 cm<sup>3</sup>/s and water temperatures in the range 5°C–35°C. Intrinsic efficiencies greater than 95% and agreement between predictions and results to within a few percent is demonstrated for a practicable range of the variables.

### Introduction

A simple method of collecting a sample of tritiated water vapour  $(\text{HTO})_v$  from air is to bubble the air through water as shown in figure 1. This is, of course, a particular application of the general laboratory technique of gas washing. The comparative collection efficiencies of various kinds of gas washing bottles were investigated experimentally many years ago<sup>(1)</sup>.

The method has been widely applied for tritium monitoring since the collected activity may be easily measured in a liquid scintillation counter<sup>(2)</sup>. Collection efficiencies observed in particular sampling systems have been reported<sup>(3–5)</sup>. In the last reference a theoretical expression ignored the sampling conditions and was of very limited applicability.

Here, the influence of relevant variables upon the collection efficiency is investigated theoretically. The experimental determinations of some of the parameters are reported and the experimentally observed dependence of the collection efficiency on some of the parameters is compared to the theoretical predictions.

### Theory

Suppose that in the model bubbler shown in figure 2, air containing water ( $\text{H}_2\text{O}$ ) at  $X$  g/m<sup>3</sup> and  $(\text{HTO})_v$  at  $C$   $\mu\text{Ci}/\text{m}^3$  is being bubbled through water. Let  $\epsilon$  be the fraction of the air from which the ingoing tritiated water is removed and assume that this fraction is saturated by water vapour from the bubbler. Suppose that the saturated vapour density at the bubbler water/air





Figure 1: Laboratory and field bubbler: Nominal volume is 250 cm<sup>3</sup>.

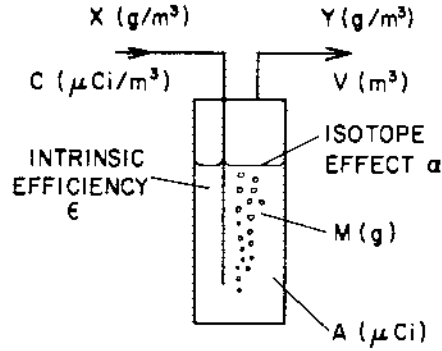


Figure 2: Model bubbler.

interface is  $Y \text{ g/m}^3$  and that the ratio of specific activity of HTO in the vapour phase to that in the liquid phase at the water/air interface under the particular dynamic conditions is  $\alpha$ , the isotope effect coefficient.

Let the mass of water in the bubbler be  $M \text{ g}$  and the activity of the tritium in it,  $A \text{ } \mu\text{Ci}$ .

The rate of change of mass of water in the bubbler with air volume  $V \text{ m}^3$  passed through is given by

$$\frac{dM}{dV} = \epsilon (X - Y) \quad (\text{i})$$

and, assuming that the activity is uniformly distributed in the bubbler water, the rate of change of activity in the bubbler with air volume is given by

$$\frac{dA}{dV} = \epsilon \left( C - \frac{\alpha AY}{M} \right) \quad (\text{ii})$$

Equation (i) may be integrated directly to give

$$M = M_0 - \epsilon (Y - X)V \quad (\text{iii})$$

where  $M_0$  is the initial mass of water in the bubbler.

Assuming that  $\alpha$  is independent of  $V$  and  $M$ , if  $X \neq Y$  the solution for  $A$  can be shown to be

$$A = \frac{MC}{(Y-X)W} \left[ 1 - \left( \frac{M}{M_0} \right)^W \right] + A_0 \left( \frac{M}{M_0} \right)^{W+1} \quad (\text{iv})$$

where  $A_0$  is the activity initially in the bubbler.

$$W = \left( \frac{\alpha Y}{Y - X} \right) - 1$$

If  $X = Y$  then

$$A = \frac{M_0 C}{\alpha X} \left[ 1 - \exp \left( -\frac{\epsilon \alpha X V}{M_0} \right) \right] + A_0 \exp \left( -\frac{\epsilon \alpha X V}{M_0} \right) \quad (\text{v})$$

In the trivial case of  $Y = 0$ ,

$$A = \epsilon C V + A_0 \quad (\text{vi})$$

while in the special limiting case of  $X = 0$  and  $\alpha = 1$ ,

$$A = A_0 - \frac{CM}{Y} \ln \left( \frac{M}{M_0} \right) \quad (\text{vii})$$

The fraction of the sampled activity that is retained in the bubbler is the number of practical interest. This is the overall collection efficiency ( $E$ ) and when, as is usually the case,  $A_0 = 0$ ,

$$E = A/CV. \quad (\text{viii})$$

Figures 3–5 illustrate how variations in  $X$ ,  $Y$ ,  $\epsilon$  and  $\alpha$  affect the variation of collection efficiency with air volume sampled. For convenience the inefficiency ( $1-E$ ) is shown, expressed as a percentage. The initial mass of water  $M_0$  is 100g in all these figures. However, since the variables  $X$ ,  $Y$  and  $M_0$  could have been combined as  $(X/M_0)$  and  $(Y/M_0)$  in the model, the curves shown can be interpreted for other values of  $M_0$  by normalising the values of  $X$  and  $Y$  to the new  $M_0$ ; i.e. by multiplying by  $(M_0/100)$ .

When the mass of water introduced into the bubbler and the mass of water lost from the bubblers are both small compared to the mass of water in the bubbler, the efficiency may be approximated by the expression

$$E = \epsilon - \frac{\epsilon^2 \alpha YV}{2M_0} \quad (\text{ix})$$

When  $W = 1$ , (i.e.,  $X/Y = 1-(\alpha/2)$ ),  $(1-E)$ , calculated from equations (iv) and (viii), is linearly dependent upon volume  $V$  and is identical to the value calculated from equation (ix). A convenient set of conditions that produce a value of unity for  $W$  ( $X = 10\text{g/m}^3$ ,  $Y = 20\text{g/m}^3$ ,  $\alpha = 1$ ) is repeated on figures 3–5. Hence in figure 3, the linear relation ( $X = 10$ ) is also the approximation for all values of  $X$  as given by equation (ix). In figure 4,  $W = 1$  for the three cases where  $\alpha = 1$  so that equation (ix) predicts  $(1-E)$  identical to the value from equations (iv) and (viii). In the other case ( $\alpha \neq 1$ ) and in figure 5, linear approximations from equation (ix) are shown by the dashed lines.

Note from figures 3–5 that the inefficiency is most sensitive to the values of  $Y/M_0$  and  $\epsilon$  for the ranges of variables shown. Clearly, equation (ix) is adequate for predicting bubbler performance over a wide range of sampling conditions and values of  $\epsilon$ . Determination of values of  $\epsilon$ ,  $\alpha$  and the effects of the sampling conditions upon the overall collection efficiency of a particular bubbler are considered separately in the following sections.

#### Intrinsic efficiency ( $\epsilon$ ) of a bubbler

In general,  $\epsilon$  will depend upon the extent of the dispersion of the two phases and their time of interaction, the air flow rate and the temperature. A high value for  $\epsilon$  is desirable, from the point of view of overall collection efficiency.

Values of  $\epsilon$  are difficult to predict *ab initio*. However the measured value of  $E$  is the lower limit of  $\epsilon$  which might be expected to be the dominant limiting parameter determining  $E$  when

$$\frac{\alpha YV}{2M_0} < 1 - \epsilon \text{ and } \ll 1 \quad (\text{x})$$

The intrinsic efficiency,  $\epsilon$ , may be estimated therefore from the measured  $E$  without the estimate's being very dependent upon the  $\alpha$  and  $Y$  which are difficult to measure accurately.

To determine experimentally when  $\epsilon$  decreases below, say, 0.98, the sampling conditions should be chosen so that  $\alpha YV < .04 M_0$ . For the conditions  $M_0 > 50 \text{ g}$ ,  $\alpha = 1$  and  $Y = 20 \text{ g/m}^3$ ,  $V$  should therefore be less than  $0.1 \text{ m}^3$ . Figure 6 shows the method used to measure  $\epsilon$  for various kinds of bubblers, masses of water, temperatures and air flow rates. In these experiments input humidity  $X$  was  $3 \text{ g/m}^3$  and the ambient air temperature was  $\sim 23^\circ\text{C}$ .

Figures 7 and 8 summarize the results of the measurements. The efficiencies predicted from the sampling conditions, using equation (ix), with  $Y$  (taken from psychrometric tables) corresponding to the temperature of the bulk of the bubbler water and values for  $\alpha$  and  $\epsilon$  of unity are also shown. Because this represents a lower limit to the efficiency that could be attributed to the sampling conditions, where the experimental points drop below this curve, the intrinsic

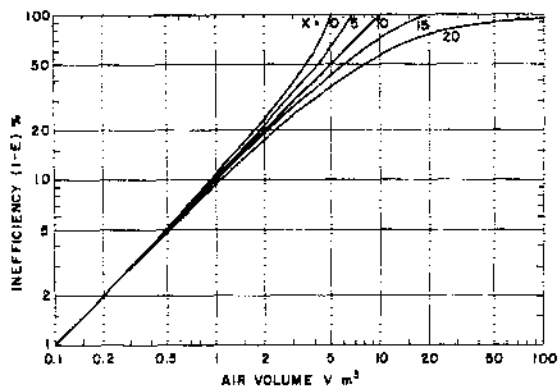


Figure 3: Calculated variations of collection inefficiencies with air volume for different values of sample humidity. Fixed parameters are  $Y = 20 \text{ g/m}^3$ ,  $M = 100\text{g}$ ,  $\alpha = 1$ ,  $\epsilon = 1$ . Values of sample humidity ( $X \text{ g/m}^3$ ) are given on the appropriate curves.

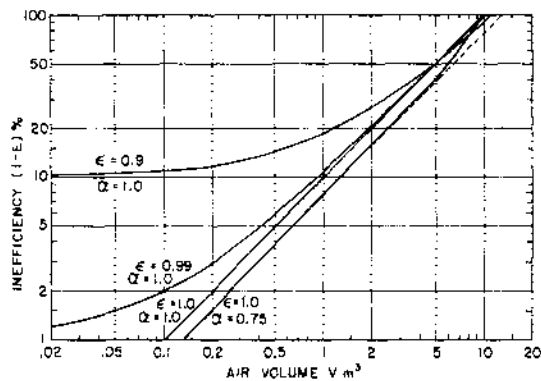


Figure 4: Calculated variations of collection efficiencies with air volume for different values of intrinsic efficiency  $\epsilon$  and isotope effect coefficient  $\alpha$ . Fixed parameters are  $X = 10 \text{ g/m}^3$ ,  $Y = 20 \text{ g/m}^3$ ,  $M = 100\text{g}$ . Values of the intrinsic efficiency  $\epsilon$  and isotope effect coefficient  $\alpha$  are given on the appropriate curves. The dashed linear extension when  $\alpha \neq 1$  is calculated from equation (ix).

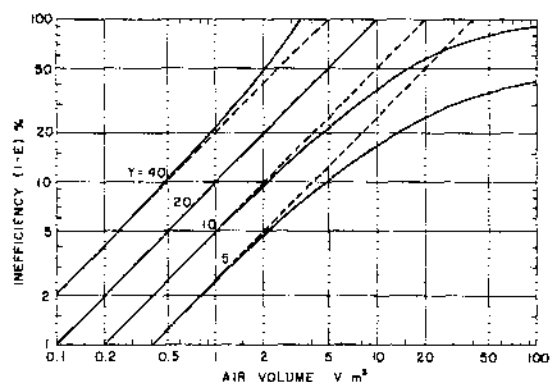


Figure 5: Calculated variations of collection efficiencies with air volume for different values of effluent air humidity. Fixed parameters are  $X = 10 \text{ g/m}^3$ ,  $M = 100\text{g}$ ,  $\alpha = 1$ ,  $\epsilon = 1$ . Values of effluent air humidity ( $Y \text{ g/m}^3$ ) are given on the appropriate curves. The solid lines are calculated from equations (iv), (v) or (vii) with (viii). The linear dashed extensions are from the approximate equation (ix).

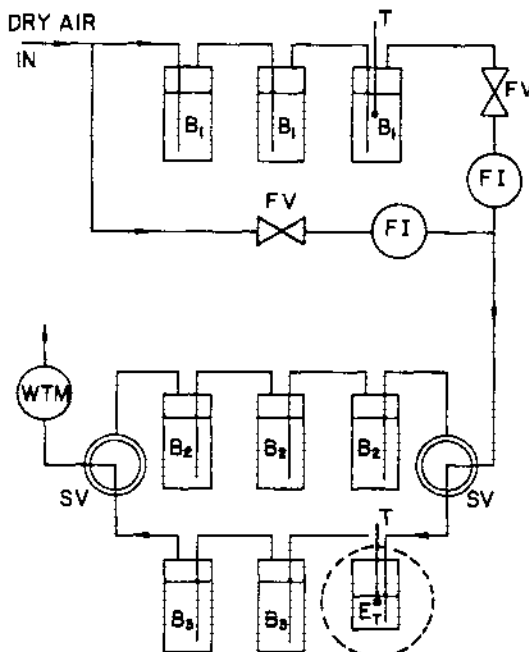
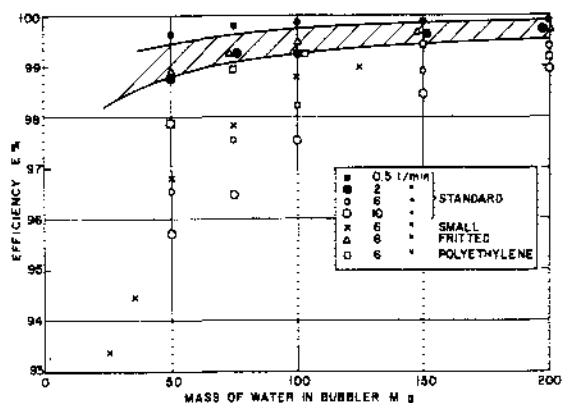


Figure 6: Outline of experimental arrangement for measuring properties of various bubblers. Tritiated water vapour was introduced into an air stream by passage through a series of bubblers ( $B_1$ ) filled with tritiated water then diluted with dry air to give the requisite relative humidity. A metered flow was passed firstly, through the bubbler ( $E_1$ ) under test and secondly, through a series of bubblers ( $B_3$ ) to collect the activity escaping from the test bubbler. A measured volume of  $0.096 \text{ m}^3$  was used throughout. The valves ( $FV$ ) and rotameters ( $FI$ ) were used for setting up the flow rates and for maintaining steady conditions. The wet-test meter ( $WTM$ ) was used to measure the total volume of air passed by the test bubbler. During set up of the flows and after completion of a particular sample, the tritiated flow was diverted by valves ( $SV$ ) through the second series of bubblers ( $B_2$ ). The water temperature ( $T$ ) in the last tritiated source bubbler and the test bubbler were measured during each sample. The activity in the test bubbler and the activities in the series of bubblers collecting the escaped activity were determined by diluting the water in each bubbler to a known volume and measuring the activity in an aliquot with a liquid scintillation counter. The experimental efficiency was then  $A/(A + \sum B_3)$ . Sufficient members of the  $B_3$  series were included in the measurement to ensure that the activity not accounted for was less than 1% of the total lost. Generally, 2 were sufficient.



**Figure 7:** Measured efficiencies with different quantities of sampling water for various bubblers at a fixed air flow rate and for one bubbler at various flow rates. The band defined by the hatched area is the range of minimum efficiencies calculated from equation (ix) for all conditions used assuming an intrinsic efficiency  $\epsilon = 1$ . The standard bubbler is the one illustrated in figure 1. 'Small' refers to a similar one of volume  $125 \text{ cm}^3$ . 'Fritted' refers to the standard type with a fritted stone air disperser. The polyethylene bubbler was similar in size to the standard with a coarse frit.

efficiency was the limiting parameter. This occurs with the smaller water masses and at the higher flow rates with the standard bubbler as might be expected. Nevertheless, even with only 25g of water  $\epsilon$  was greater than 0.93. In all cases with 200g of water as collector, the intrinsic efficiency was greater than 0.99. With the fritted bubblers which more finely dispersed the air in the water a reduction in  $\epsilon$  with decreasing  $M_0$  was not observed at the flow rate used. Indeed, the total efficiency was better than predicted from the sampling conditions alone. In this case and at the lowest flow rate in the standard bubbler, agitation and mixing of the water in the bubbler was noticeably less than in the other cases. The effective temperature of the water at the air/water interfaces may therefore have been lower than that of the bulk of the water, resulting in a lower effective value for  $Y$ , and accounting, in part, for the high collection efficiency observed.

Clearly, the intrinsic efficiencies of even simple bubblers are high enough and are sufficiently independent of mass of water and flow rate for most practical purposes when used with air flow rates below 10 l/min and water masses above 50g in these types of bubbler.

#### Isotope effect coefficient ( $\alpha$ )

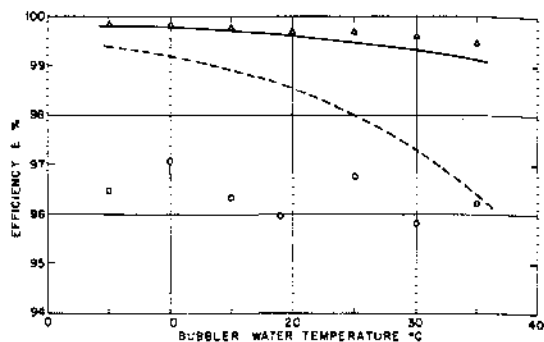
The specific activity of  $(\text{HTO})_v$  is known to be less than that of the water phase with which it is in equilibrium. The coefficient ranges from 0.86 at  $0^\circ\text{C}$  through 0.91 at  $20^\circ\text{C}$  to 0.94 at  $50^\circ\text{C}$  (6,7). However equilibrium conditions are not necessarily attained in a bubbler; the high efficiencies observed above (figure 7) are not completely explained by  $\alpha$ 's having equilibrium values. Appropriate values of  $\alpha$  can be estimated for particular conditions from the initial rate of change of efficiency ( $E$ ) with air volume ( $V$ ) since, from equation ix,

$$\alpha = -\left(\frac{dE}{dV}\right) \frac{2M_0}{\epsilon^2 Y} \quad (\text{xi})$$

This is demonstrated in the next section.

#### Variation of collection efficiency with sampled air volume

Figure 9 shows the results of two experiments in which the efficiencies of two bubblers were measured for various sampled air volumes under different conditions using the apparatus outlined in

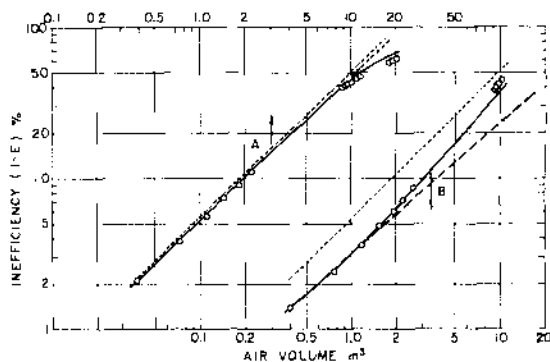


**Figure 8:** Measured efficiencies of bubblers at various water temperatures.

$\Delta$  - Bubbler with fritted glass disperser and  $M_0 = 200\text{g}$ .

$\circ$  - Laboratory bubbler as in figure 1 with  $M_0 = 50\text{g}$ .

The solid and dashed curves are the lower limits to the respective efficiencies calculated assuming that  $\epsilon = 1$  and  $\alpha$  is the equilibrium value for the measured temperature.



**Figure 9:** Measured inefficiencies of bubblers with  $M = 200\text{g}$ , sampling at  $6\text{ l/min}$  for various total air volumes. Series A =  $X \sim 18\text{ g/m}^3$ ,  $Y \sim 23\text{ g/m}^3$ . Series B =  $X \sim 3.4\text{ g/m}^3$ ,  $Y \sim 15\text{ g/m}^3$ .

Solid lines are calculated variations using values for  $\alpha$  determined from the initial values of  $(dE/dV)$ . For series A,  $\alpha = 0.85$ , for series B,  $\alpha = 0.71$ . Dashed lines are calculated from equation (ix) using the same conditions. The dotted lines are calculated using the ambient temperature ( $23^\circ\text{C}$ ) to determine  $Y$  and equilibrium  $\alpha$  in equation (ix).

figure 6 with the samples B3 being replaced periodically. The appropriate values of  $\alpha$  were determined using equation (xi) from the initial slopes established by the experimental points. The curves for the complete experiment were calculated from equation (iv) and are drawn in figure 9. Note that linear approximations also on figure 9, using the ambient temperatures from which to estimate  $Y$  and the equilibrium values of  $\alpha$  predict the efficiencies to within a few percent anyway. Of practical note here is that although carried out under similar ambient conditions, the evaporative self cooling of the series B with the drier input air results in a reduced loss rate and higher collection efficiency than in the A series. The actual bubbler bulk water temperature was about  $3^\circ\text{C}$  below the laboratory air temperature.

## Conclusion

The most important parameters determining the overall collection efficiency for  $(\text{HTO})_v$  of a bubbler are the intrinsic efficiency and the humidity of the effluent air. Deviation of the former from unity may be disregarded for a practicable range of air flow rates, temperatures and bubbler types. The dependence of the efficiency upon the humidity lost in the effluent air may be linearly related to the total air flow with a precision adequate for most monitoring purposes.

## Acknowledgement

Mrs. J. Sterling carried out the experimental work described in this paper. Discussions with Dr. P.J. Barry have proved very beneficial in defining the most appropriate model and interpreting the results.

## References

1. F.H. Rhodes and D.R. Rakestraw, Comparative efficiencies of gas-washing bottles. *Industrial and Engineering Chemistry*, 3, 143-144 (1931).
2. G. Cowper and R.V. Osborne, Measurement of tritium in air in the presence of gamma radiation, "Proceedings of the First International Congress of Radiation Protection", pp 285-293, Pergamon Press, Oxford, 1968.
3. D. McConnon, The use of water as a sampling medium for tritium oxide, Battelle Northwest Laboratory report BNWL-CC-547, Richland, 1970.
4. A.M. Valentine, An investigation of a bubbler tritium sampler, Los Alamos Scientific Laboratory report LA-3916, Los Alamos, 1968.
5. A.K. Dannecker and K.H. Spittel, Discussion in "Assessment of Airborne Radioactivity", pp 519, International Atomic Energy Agency, Vienna, 1967.
6. O. Sepall and S.G. Mason, Vapor/liquid partition of tritium in tritiated water, *Can. J. Chem.* 38, 2024-2025, (1960).
7. F.A. Prantl and E. Robertson, Progress Report, Biology and Health Physics Division, Jan-Mar 31, 1972. Atomic Energy of Canada Limited report AECL 4213, Chalk River, 1972.

A RUGGEDIZED ULTRASENSITIVE FIELD AIR SAMPLER  
FOR DIFFERENTIALLY DETERMINING TRITIUM OXIDE  
AND GAS IN AMBIENT AIR ATMOSPHERE

R. Brown, H. E. Meyer, B. Robinson and W. E. Sheehan  
Mound Laboratory\*  
Miamisburg, Ohio

Abstract

The instrument described is an operational, practical, ruggedized, ultrasensitive, tritium field air sampler assembled for the simultaneous, differential sampling of the environmental air for tritium oxide and elemental tritium. The system uses hardware assembled and packaged in such manner as to facilitate use in the field as well as in the laboratory. The sampling system occupies relatively small space and is simple to operate. The detection sensitivity approaches tritium background levels and is achieved by high volume sampling, efficient removal of tritium oxide and elemental tritium ("tritium gas"), and counting the recovered fractions by liquid scintillation spectrometry.

Introduction

The AEC standard for the maximum permissible concentration of tritium gas in uncontrolled areas is 200 times greater than for tritiated water vapor. Because of this difference, monitoring for tritium in the past at Mound, as well as at other locations, was for tritium as the oxide with the assumption that if the levels did not exceed the standard for the oxide, then the gas would be well within the standard.

When tritium in the environment or releases of tritium to the environment are measured, a measurement of oxide or even total tritium would not be sufficient to determine whether concentration guides have been exceeded; measurement of both tritium gas and tritiated water are necessary. Numerous methods for monitoring tritium oxide have been reported, but the literature is scant concerning tritium gas measurements. Mound Laboratory's goal is to determine background levels of tritium oxide and gas, both from natural and artificial sources, and to establish a baseline to provide a means of measuring any tritium that might be inadvertently added from operations at Mound.

General Description

The Mound Laboratory total system designed by MRC, Dayton Laboratory, consists of a tritium field sampler and a separate laboratory sample recovery system.

Collection System A picture of the collection system is shown in Fig. 1. The collection system with sample inlet, sample outlet, and hydrogen inlet connections, mounted controls, and indicators occupies a space 44 in. wide, 16 in. deep, and 38 in. high. The

\*Mound Laboratory is operated by Monsanto Research Corporation for the U. S. Atomic Energy Commission under Contract No. AT-33-1-GEN-53.

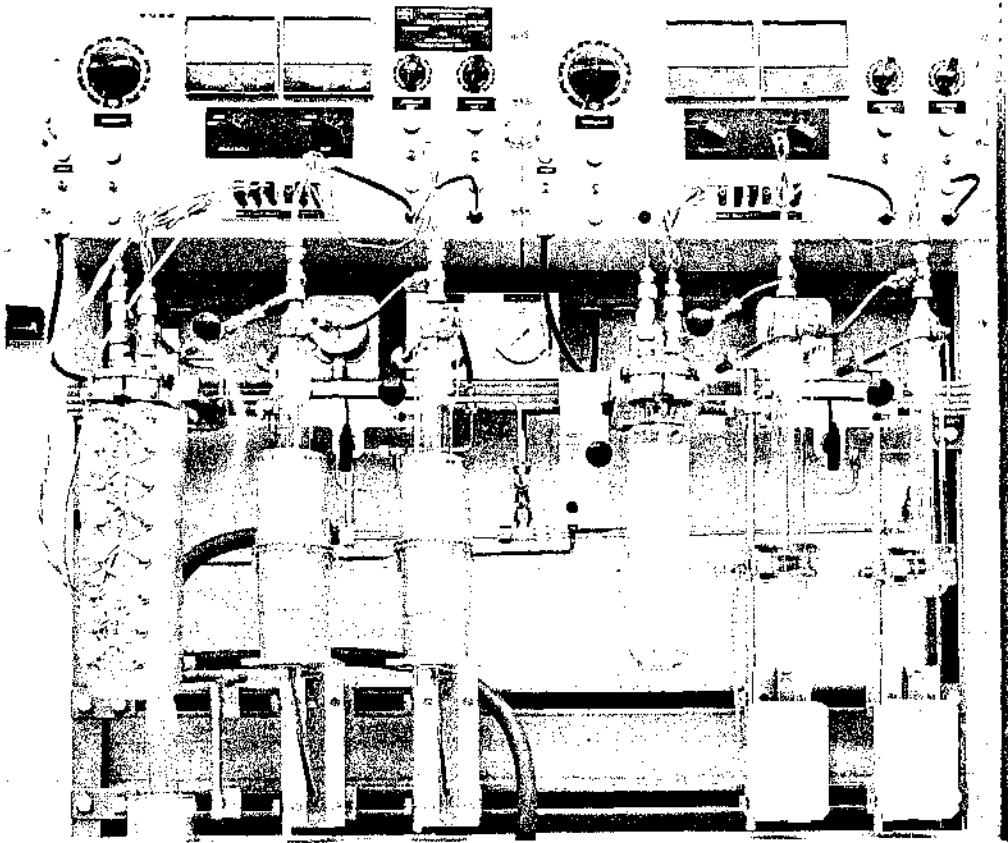


Fig. 1 - Tritium sample collection system.

sample collection/concentration bottles with protective wire-coated grid encasement are 3.88 in. in diameter and 17.55 in. overall length. The weight of the collection system, less bottles, is approximately 135 lb.

The collection system accommodates four sample collection bottles, the first two for the collection of tritium oxide and the other two for the collection of elemental tritium. The system valving permits a wide range of operating modes. Inadvertent introduction of hydrogen into the system (for the collection of elemental tritium) is averted by requiring air to flow past a sensor before a solenoid valve in the hydrogen line can be opened. Maintaining hydrogen supply within the limit of the flow meter provides assurance that the 4% lower explosive limit (L.E.L.), will not be exceeded. For normal air sample system flow rates of 30 to 75 liters/min with carrier hydrogen supplied at the rate of 100 cc/min, the mixture runs 0.2 to 0.4% of the L.E.L. The sensor and solenoid valve shut off the hydrogen inlet line in the event of low or no air flow during sampling.

Recovery System A two-station sample recovery system is shown in Fig. 2. This versatile, two-station system, was provided at Mound Laboratory to determine various operating parameters. The

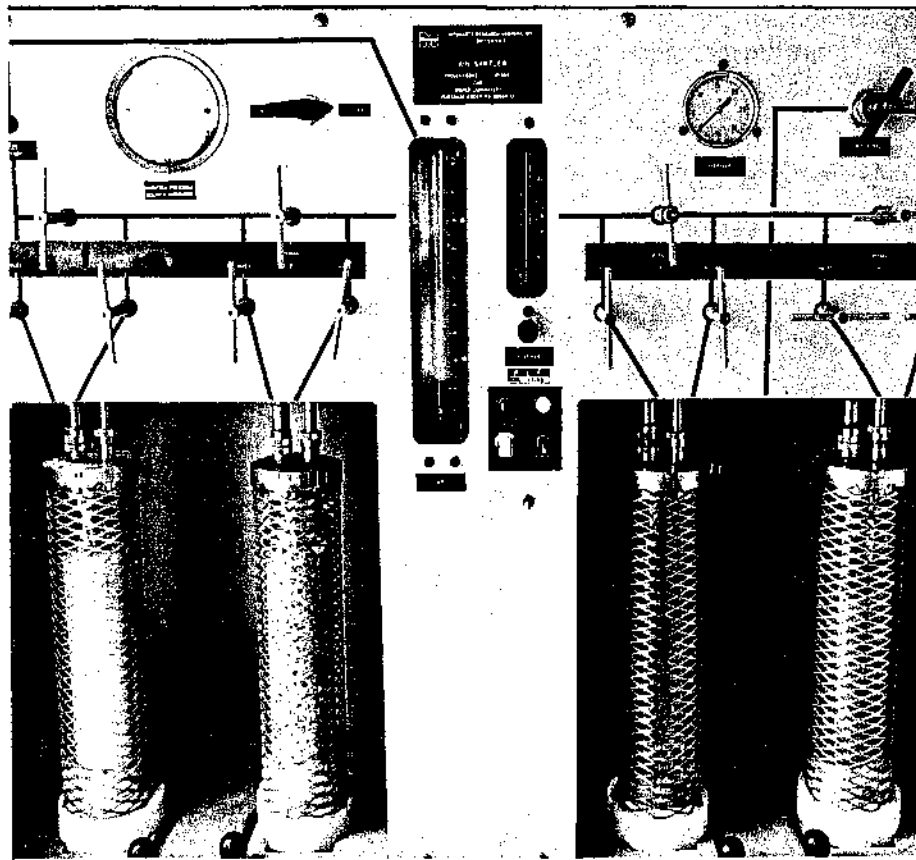


Fig. 2 - Two-station tritium sample recovery system.

recovery system provides a laboratory facility for dry gas displacement or low pressure desorption of a collected sample at selected temperature and pressure into two cold traps in series.

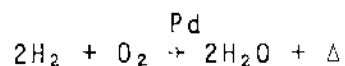
#### Results and Discussion

Experience to date with the system at Mound Laboratory indicates that the silica gel used in the sample collection bottles must be thoroughly dehydrated prior to use. Typical air flow rate used in sampling has been 50 liters/min. At this flow rate, 750 g of silica gel in the collection bottles is adequate for a large volume grab sample in the range of 1500 to 3000 liters even under unfavorable ambient conditions of high relative humidity and temperature. Preliminary results in calibration of the system indicate 97-99% of the water vapor in the sampled air is recovered using one oxide collection bottle.

Seven hundred fifty grams of silica gel is also placed in the elemental tritium or gas collection bottles. The silica gel is coated with palladium black by adding 2 g of palladium powder to the 750-g charge of silica gel and agitating the mixture thoroughly until the palladium powder is completely coated on the surfaces of the silica gel particles. As the ambient air stream passes through this collection bottle, hydrogen gaseous isotopes are reacted with



the palladium to form oxides. The reaction is:



The resulting water is collected by the silica gel just as in the primary oxide collection bottles.

The cryogenic moisture recovery traps employed in the recovery system use liquid nitrogen. Essentially all of the water driven off the catalytic collector is captured in the first cold trap. No visible moisture has occurred in the second cold trap. Moisture is recovered in the system in a simple reverse of the method of collection. Recovery is completed in 30 min with the collection bottle at a temperature at 300°C. The heating mantle and its associated control variac are used to obtain the desired temperature. When the moisture has been driven off the sample, the exit temperature decreases. This temperature change can be monitored as the signal for desorption completion.

The use of palladium black is preferable to copper oxide since palladium reacts with hydrogen at ambient temperatures, whereas copper oxide must be heated before it will react. Since silica gel was found to be satisfactory, no other desiccant was evaluated.

Silica gel, as received from the manufacturer, contains residual moisture, so it must be thoroughly baked out before using at a temperature high enough to condition the silica gel for use in a reasonable period of time but not at a temperature high enough to destroy its sorption properties. Tests performed in the development of this monitoring system showed that a one-hour bakeout at 300°C adequately removed the moisture without affecting sorption properties. Normal residual moisture ranges from 0.03 to 0.07 g of water per gram of new silica gel.<sup>1</sup> To date, silica gel has been reused since there has been no indication that the bakeout reduced the collection efficiency in subsequent runs using known aqueous solutions of tritium oxide.<sup>2</sup>

The efficiency of the procedure for the determination of tritium oxide was tested using the set-up shown in Fig. 3. Known solutions of varying tritium oxide content, assayed by liquid scintillation counting and calibrated against a National Bureau of Standards (NBS) tritium oxide standard, were placed in the container marked "tritium oxide". Air was pulled through the drying trap at a flow rate of 50 liters/min, during which time the standardized tritium oxide was evaporated into the air stream and collected in the oxide collection bottle. Results indicate an overall recovery of 97-99%.

The efficiency of the procedure for the determination of elemental tritium was tested using the setup shown in Fig. 4. The flow rate of the calibration system was adjusted to 50 liters/min with valve #1 open and valves #2 and #3 closed. Hydrogen flow was adjusted to 150 cc/min. The system was operated in this configuration for 20 min after which time valves #2 and #3 were opened and valve #1 was closed. The tritium gas sample standard flask was flushed for 10 min. Gas samples used in calibration were standardized against an NBS gas standard prepared from the NBS tritium oxide standard.<sup>3</sup> At the end of 30 min the set-up was shut down, the

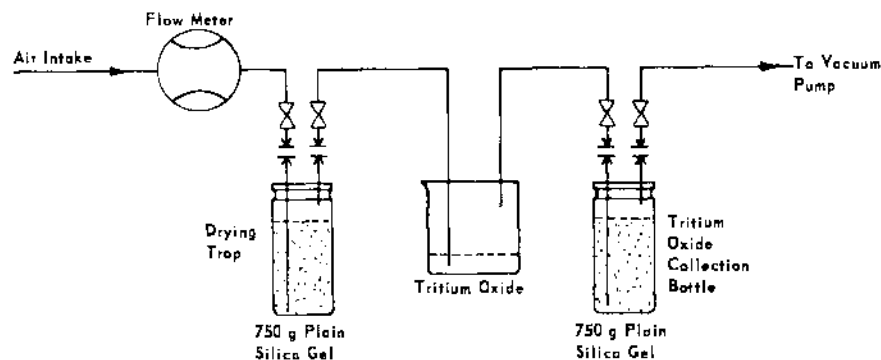


Fig. 3 - Setup used to test efficiency of tritium oxide collection.

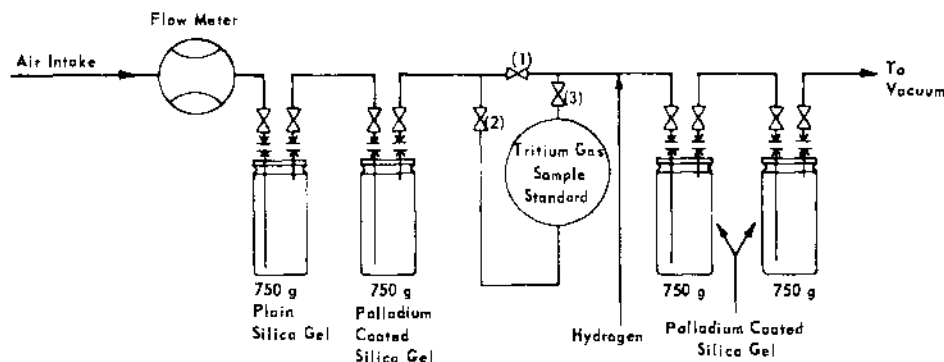


Fig. 4 - Setup used to test efficiency of elemental tritium collection.

collection flask containing the palladium-coated silica gel was removed, and the water was removed using the standard recovery procedure.

Two grams of palladium powder coated on the 750 g of silica gel are required to achieve recoveries in excess of 90% in the collection bottle. One gram of palladium powder achieved only 53 to 80.5% overall recovery in the collection bottle.

The theoretical sensitivity of this sampling system is  $1.6 \times 10^{-12}$   $\mu\text{Ci/cc}$  of air at the 95% confidence level for liquid scintillation counting with a 20 counts/min counter background, a 21% counter efficiency, and a 100 min sample counting time. A total of 1,223 liters of air sampled at a relative humidity of 50%, 68°F, and 760 mm barometric pressure would yield 10 ml of water which is the optimum amount that can be blended with a scintillation liquid for counting. A typical one-hour sample run at a flow rate of 50 liters/min would sample 3000 liters of air and yield some 25 ml of water. If all of the water could be counted as a single sample, the tritium oxide sensitivity would calculate to  $0.6 \times 10^{-12}$   $\mu\text{Ci/cc}$ . The limitation in the system sensitivity is, therefore, not in the air sampling/collecting apparatus but in the liquid scintillation counting since only 10 ml or less of collected moisture can be analyzed. The lower detection limit could be achieved by concentrating the tritium in moisture collected to 10 ml by electrolysis. A sensitivity of  $1.6 \times 10^{-12}$   $\mu\text{Ci/cc}$  of air at the 95% confidence level, however, is in the range of tritium oxide background levels at Miami,

Florida<sup>4</sup> and is adequate for an effective environmental oxide monitoring program.

A sensitivity of  $0.6 \times 10^{-12}$   $\mu\text{Ci}/\text{cc}$  of air is attainable for all samples containing elemental tritium since the total water in the collected gas sample can be controlled to a volume of 10 ml, all of which can be counted. The carrier hydrogen must be used to obtain sufficient water for analysis. Theoretically, 600 cc of hydrogen carrier gas produce 0.5 cc of water.

The use of silica gel, or other desiccants, for air sampling presents a "memory effect" that must be taken into consideration. The memory effect results from the fact that complete moisture desorption of silica gel is not attainable. The tritium content of the residual moisture will affect the results of each air sample collected. This factor has not been completely investigated with this sampling system, but based on limited data obtained to date, it appears that about 1% of the moisture from tritium oxide samples and from 3 to 10% of the moisture from tritium gas samples is not removed. In accurate sampling the memory effect can be corrected for by passing vapor of known tritium concentration through the sample collection bottle while it is being baked out under vacuum. Sufficient moisture is passed on to the silica gel to assure that all residual moisture is "pushed out" and replaced. When the sample collection bottle is used, the sample data are corrected to account for the controlled, known residual or background moisture and its tritium content.

#### References

1. C. L. Mantell, Adsorption, Chemical Engineering Series, 1945, pp. 167-178.
2. J. T. Baker Chemical Co., Phillipsburg, N. J., Product Information Bulletin-68, September 9, 1970.
3. M. L. Curtis, C. T. Bishop, M. M. Bolton, and R. L. Ryan, "Determination of Tritium Leak Rates from Waste Processing Equipment." Presented at Tritium Symposium, August 30 through September 2, 1971, Las Vegas, Nevada. To be published in Symposium Proceedings.
4. H. G. Ostlund, "A Study of Tritiated Hydrogen and Its Compounds in the Environment." Progress Report ML 71027, March 1971.

COMPARISON OF ALBEDO DOSIMETERS AND NUCLEAR  
TRACK DETECTORS FOR NEUTRON MONITORING

E. Piesch, and B. Burgkhardt  
Health Physics Division  
Karlsruhe Nuclear Research Center, Fed. Rep. Germany

1. Abstract

The albedo neutron dosimeters so far used in personal dosimetry provided oversensitive indications of intermediate and thermal neutrons. Therefore, correction factors dependent on the location had to be used to assess the dose equivalent (Harvey<sup>1</sup>, Hoy<sup>2</sup>). An albedo dosimeter tested at the Karlsruhe Nuclear Research Center showed promising results in measuring the dose equivalent of fast neutrons<sup>3, 4</sup>. Intercomparison measurements with nuclear track detectors were performed to indicate the dosimeter response, influence of the energy dependence and of the direction of the radiation incidence on the dosimeter reading.

2. Dosimeter and Radiation Sources

In a LiF-albedo dosimeter the thermal neutrons moderated and backscattered in the body of the wearer are detected by the <sup>6</sup>Li (n,α)-reaction. If a pair of dosimeters is used, the neutron fraction of the reading is obtained from the difference in readings between a TLD-600 dosimeter (neutron + γ-dose reading) and a TLD-700 dosimeter (γ-dose reading).

Albedo dosimeters so far have been used preferably for the detection of intermediate and thermal neutrons. The detection response to fast neutrons is approximately 5 % of the response to thermal neutrons, but still 50 % of the γ-response. In this way, a LiF-albedo dosimeter is capable of detecting the dose equivalent of neutrons and of gamma rays over a dose range between 20 mrem and more than 1000 rem.

The following dosimeters were used for intercomparison measurements (cf. Table 1):

- Albedo neutron dosimeter according to Harvey: Absorption of incident thermal neutrons by means of a boron capsule<sup>1</sup>. A pair of Harshaw ribbons of 3 x 3 x 1 mm<sup>3</sup> size were used.
- Single albedo dosimeter:  
Separate measurement of incident neutrons (D<sub>2</sub>) outside the boron capsule and neutrons back-scattered from the body (D<sub>1</sub>) within the boron capsule by means of one pair of dosimeters each<sup>4</sup>.
- Albedo dosimeter system:  
Reduction of the influence of the body by wearing a dosimeter belt with one single albedo dosimeter each on the front and back of the body<sup>3, 4</sup>. A functional relationship found

Detector	Energy	response
Kodak NTA Film	> 0.7 MeV	2 · 10 <sup>4</sup> tracks/cm <sup>2</sup> · rem <sup>*)</sup>
<sup>237</sup> Np+Makrofol E	> 0.75 MeV	4 tracks/cm <sup>2</sup> · rem <sup>*)</sup>
{40 μg/cm <sup>2</sup> }		
<sup>232</sup> Th+Makrofol E	> 1.2 MeV	37 tracks/cm <sup>2</sup> · rem <sup>*)</sup>
{0.05 mm foil}		
Albedo Dosimeter	n <sub>th</sub> , n <sub>l</sub>	< 10 R/rem
Harvey	and	
Single Albedo	> 100 keV	0.5 R/rem
Dosimeter		
Albedo Dosimeter	-14 MeV	0.54 R/rem
System		

\*) Fluence-Dose conversion factor for track detectors  
2.86 · 10<sup>7</sup> n/cm<sup>2</sup> · rem

Tab.1: Neutron detector characteristic

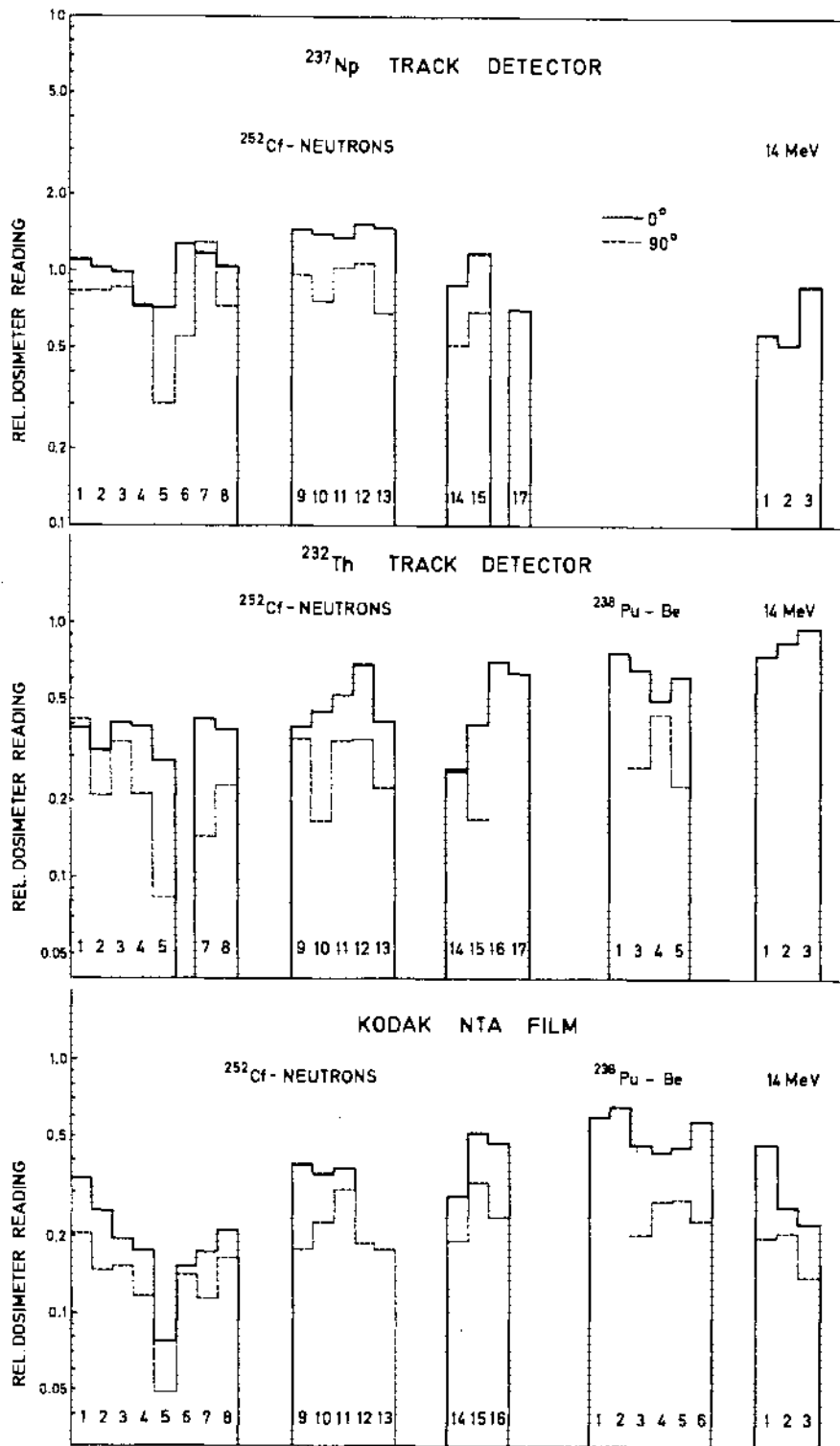


Fig.1: Relative dosimeter reading of nuclear track detectors after phantom irradiations with  $^{252}\text{Pu}$ -Be- and 14 MeV neutrons

Position No.	Source	Distance source-detector
1	<sup>252</sup> Cf in air	2 m
2	+ 5 cm Pb	
3	+ 5 cm Al	
4	+ 5 cm Fe	
5	+ 16 cm Fe	
6	+ 5 cm PVC	
7	+ 11 cm PVC	
8	+ 5 cm Concrete	
9	<sup>252</sup> Cf in air	1 m
10	8 cm wall distance	
11	1 m	
12	2 m	
13	3 m	
14	<sup>252</sup> Cf behind water layers of	2 m
15	4 cm	
16	12.5 cm	
17	43 cm	
1	<sup>238</sup> Pu-Be in air	1 m
2		2 m
3	In a small room:	1 m
4		2.3 m
5		2.2 m
6		3.6 m
1	14 MeV in air	20 cm
2		30 cm
3		54 cm
4		1 m
5		1.5 m
6	behind concrete (40 cm)	2 m
7		3 m

Tab.2: Sources and exposure positions

Table 2 contains further details about the exposure positions. Irradiation with a <sup>252</sup>Cf-source of 1 mg was performed in free air and behind shieldings of PVC, concrete, aluminium, iron and lead. The source was set up at distances between 8 cm and 4 m from a concrete wall (wall effect), the distance of the detector remaining constant, or was suspended into a water tank of 60 cm diameter directly opposite the detector to generate water layers of different thicknesses ranging between 4 and 51 cm.

Irradiations with <sup>238</sup>Pu-Be neutrons were performed in free air and in a small chamber of 2 x 3 m<sup>2</sup>, the source being located in one corner of the room. 14 MeV neutrons were used for irradiation in free air and behind a wall, respectively. Exposures in a heavy water moderated power reactor (Obrigheim Nuclear Power Station) were performed directly on top of the reactor core outside the biological shield. Exposures in the FR 2 research reactor were performed in accessible places between the concrete and paraffin shields for in-pile experiments in horizontal beamholes.

### 3. Measured Results for Fast Neutrons

#### 3.1 Nuclear Track Detectors

Figure 1 is a diagram of the reading of nuclear track detectors referred to the reading of the rem-counter for various exposure conditions. Due to fading within two weeks, the results obtained with the nuclear track emulsion are below of 60 %. An iron shield of 16 cm reduces the average neutron energy of the fission spectrum from 1.9 MeV to 0.88 MeV<sup>5</sup>. Here the NTA-film had higher fading and lower response, thus indicating only 25 % of the free air exposure. Because of the fading, the energy threshold of the NTA film is higher than in the <sup>232</sup>Th-detector. The reading of the <sup>232</sup>Th-detector for fission neutrons is lower by a factor of 2, while for 14 MeV neutrons it is only slightly higher than for Pu-Be

experimentally between D<sub>1</sub> and D<sub>2</sub>/D<sub>1</sub> is used for correction:

$$D = k \times D_1$$

- Kodak NTA Type A nuclear emulsion welded in aluminium-plastic foil for the detection of fast neutrons > 0.7 MeV.
- <sup>237</sup>Np + Makrofol for the detection of fast neutrons > 0.75 MeV and
- <sup>232</sup>Th + Makrofol for the detection of fast neutrons > 1.2 MeV by counting of fission fragment tracks.

The exposures were performed with a human phantom (water bottles of 20 cm diameter, 40 cm height) at 1.4 m above ground with radiation incidence from the front, the sides and the back (0°, 90°, 180°, 270°). The respective dose equivalent were measured with an Anderson-Braun type rem-counter at the point of exposure of the phantom. In the case of 14 MeV neutrons also activation and threshold detectors were used. The rem-counter and the nuclear track detectors were calibrated in free air with an Am-Be-source of known source intensity.

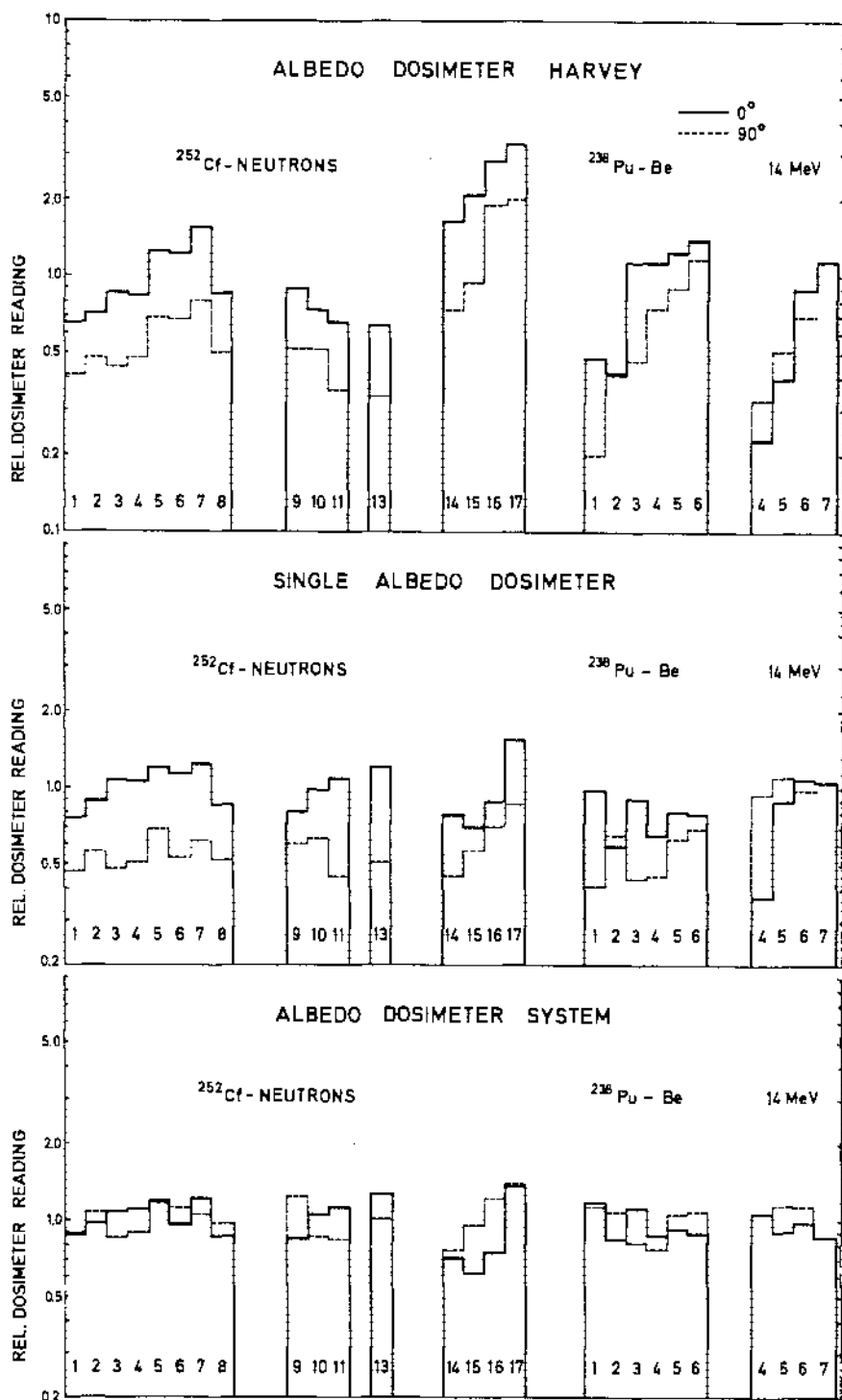


Fig.2: Relative dosimeter reading of albedo neutron dosimeters after phantom irradiations with  $^{252}\text{Cf}$ -,  $^{238}\text{Pu}$ -Be and 14 MeV neutrons

neutrons despite the high (n,f)-cross section, due to the fluence-dose conversion factor.

The deviation of the  $^{237}\text{Np}$  reading from the reading of the rem-counter is between +50 % and -30 %. For 14 MeV neutrons, the deviations are larger because of the fluence-dose conversion factor. For radiation incidence under  $90^\circ$  nuclear track detectors in most cases indicate between 75 % and 45 % of the reading obtained by incidence upon the front.

### 3.2 Albedo Neutron Dosimeter (see Figure 2)

Because of its oversensitivity to thermal and intermediate neutrons, the albedo dosimeter according to Harvey indicates up to a factor of 6 more than the rem-counter. However, a modification of the fission neutron spectrum by 5 cm of shielding results in deviations only within  $\pm 30\%$ . In single albedo dosimeters the energy dependence and the influence of scattered thermal neutrons from the environment are reduced. For front incidence a deviation in readings for fission neutrons is found to be within  $\pm 40\%$ . For 14 MeV neutrons different correction factors were used. As in nuclear track detectors, dosimeter readings between 75 % and 45 % of the reading obtained in free air were found for radiation incidence under  $90^\circ$ . The albedo dosimeter system reduces the influence of the direction dependence to approximately  $\pm 15\%$  for radiation incidences under  $0^\circ$ ,  $90^\circ$ ,  $270^\circ$  and  $180^\circ$ . The total influence of the error due to energy and direction dependence of the dosimeter reading accordingly is  $\pm 30\%$  for a dosimeter belt with two albedo dosimeters.

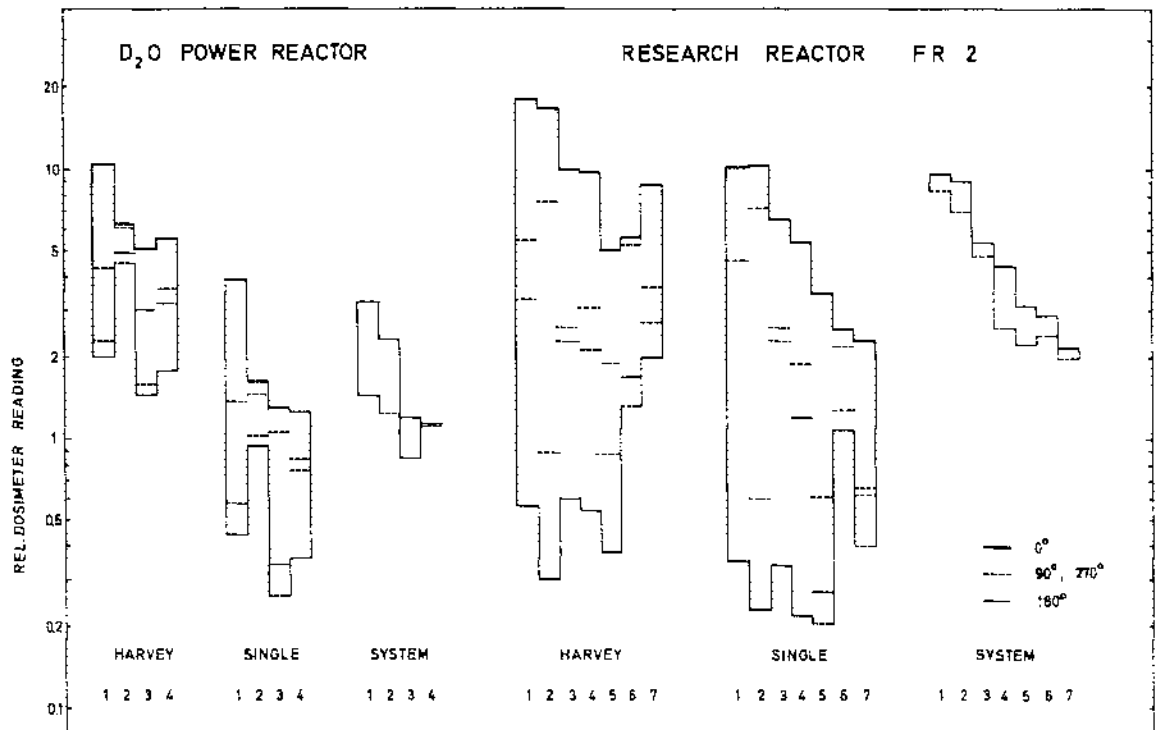


Fig. 3: Relative dosimeter reading of albedo neutron dosimeters at reactor sites after phantom irradiations for different directions of radiation incidence



#### 4. Results Measured at Reactor Sites (see Figure 3)

Chiefly thermal and intermediate neutrons are encountered near reactors, which requires correction factors to be applied in the evaluation of albedo dosimeters which depend on the location<sup>1,2</sup>. The dosimeter reading varies by up to a factor of 2 in the case of incidence on the front near a power reactor and by a factor of up to 4 when measured near a research reactor. Harvey dosimeters have maximum sensitivities of 5 and 9 R/rem, respectively. In single albedo dosimeters this value is reduced to 2 and 5 R/rem, respectively. In albedo dosimeter systems the dosimeter reading shows a maximum variation of a factor of 2.5 except for the first two irradiation positions in the FR 2 research reactor. The maximum sensitivity is 2.5 R/rem. The albedo dosimeter system has the advantage of a non-directional dose reading for all radiation incidences between 0° and 180°. However the more unfavorable conditions existing near beamholes of research reactors are in no way representative for a personnel monitoring at reactors.

#### 5. Summary

Because of the sometimes relatively high dose fraction of intermediate neutrons, location dependent correction factors must be taken into account in personnel monitoring at reactors by means of albedo dosimeters (corrections by up to a factor of 5). Because of the energy threshold, nuclear track detectors cannot be used for this purpose. In extreme cases, corrections by up to a factor of 10 are required near beamholes. This must be anticipated also in the energy range of intermediate neutrons from the calculated response of the dosimeter reading (see<sup>4</sup>, including results from<sup>6</sup>).

Albedo neutron dosimeters can be applied preferably to personnel monitoring near neutron sources in the range of energy between some 100 keV and 14 MeV. The readings are comparable to those obtained from a <sup>237</sup>Np detector with respect to energy and direction dependence, while <sup>237</sup>Th and the NTA-film show more unfavorable results because of the energy dependence and the energy dependence plus fading, respectively. For 14 MeV neutrons, <sup>238</sup>Pu-Be neutrons and <sup>252</sup>Cf-neutrons, a maximum deviation of ± 30 % was ascertained for the albedo dosimeter system, which is due to influences of scattered radiation, the neutron spectrum, and differences in the direction of radiation incidence between 0° and 180°. Compared with nuclear track detectors, albedo dosimeters have the advantage of a broader range of measurement (between 20 mrem and in excess of 1,000 rem), higher measuring accuracy (± 3 %), no energy threshold (with a detection of the correct dose > 100 keV), simple evaluation and simultaneous indication of the gamma dose.

#### Acknowledgments

We wish to thank Mr. J. Vaane for giving us the opportunity to perform irradiations in the laboratories of the European Institute for Transuranium Elements at Karlsruhe. We gratefully acknowledge the assistance of Mrs. B. Baur and Mrs. I. Hofmann in the careful evaluation of the detectors.

#### References

- <sup>1</sup> Harvey, J.R.: Report RD/B/N 827, 1967, Report RD/B/N 1547, 1970
- <sup>2</sup> Hoy, J.E.: Health Physics 23, 385, 1972, Report DP-1277, 1972
- <sup>3</sup> Piesch, E., Burgkhardt, B., Vaane, J.: Report KFK 1666, 1972
- <sup>4</sup> Piesch, E., Burgkhardt, B.: IAEA-Symposium on Neutron Monitoring 1972
- <sup>5</sup> Makra, S.: Private communication
- <sup>6</sup> Alsmiller, R.G., Barish, J.: ORNL-4800, p. 67-69, 1972

НЕКОТОРЫЕ ПЕРСПЕКТИВЫ ПРИМЕНЕНИЯ КРЕМНИЕВЫХ  
ДЕТЕКТОРОВ ИОНИЗИРУЮЩИХ ИЗЛУЧЕНИЙ В ИССЛЕДОВАНИЯХ ПО РАДИАЦИОННОЙ ЗАЩИТЕ

А.А.Петушнов, В.А.Манчук

Институт гигиены труда и профзаболеваний АМН СССР,  
Москва, СССР.

The possibility of application of pulse rate measurements for x- and gamma-ray dosimetry with silicon radiation detectors was investigated. It was shown that this mode of operation ensures the sensitivity of  $10^{-6}$  r.min<sup>-1</sup> for the detectors used in this work, and it is possible to decrease the energy dependence of sensitivity, for use in many radiation protection investigations by increasing the discrimination threshold. The general expression which can be used for rough estimates of sensitivity for these detectors was determined.

The background count rate of surface barrier detectors with different sensitive area from 0.2 to 60 cm<sup>2</sup> in alpha particles energy interval 3-9 MeV was determined. It was suggested for health physics and radiation protection monitoring of alpha-particles and simultaneous spectrometry to use the mosaic probe with large sensitive area (~60 cm<sup>2</sup>) which consists of many parallel united detectors one of which could be eliminated for spectrometric measurements.

Теперь уже хорошо известно, что неоднородные кремниевые детекторы ядерных излучений во многих случаях обеспечивают преимущества при использовании их для дозиметрии и радиометрии в исследованиях по радиационной защите. Тем не менее, ряд направлений возможного их применения изучен еще недостаточно, что и явилось основанием для настоящей работы.

ДОЗИМЕТРИЯ РЕНТГЕНОВСКОГО И ГАММА-ИЗЛУЧЕНИЯ  
С ИСПОЛЬЗОВАНИЕМ ИМПУЛЬСНОГО РЕЖИМА РАБОТЫ  
КРЕМНИЕВЫХ ДЕТЕКТОРОВ.

Служба радиационной безопасности для контроля радиационной обстановки нуждается в малогабаритном дозиметре рентгеновского и гамма излучения, обеспечивающем измерения в широком диапазоне мощностей экспозиционных доз. Однако, интегральный режим работы таких детекторов, подробно изученный рядом авторов (1,2,3 и др.), обладает низкой чувствительностью, ограничивающей использование их для оценки радиационной обстановки. С целью изучения возможности создания дозиметра с более высокой чувствительностью, выполнены исследования счетного режима работы золото-кремниевых

и кремний-литиевых детекторов. Измерялась их чувствительность (выраженная в скорости счета на единицу экспозиционной дозы) и зависимость ее от амплитудного порога регистрации импульсов, а также обсуждается способ оценки величины чувствительности расчетным путем.

Скорость счета на единицу экспозиционной дозы может быть выражена соотношением:

$$\frac{N_0}{PS} = \frac{1 - e^{-\mu_{Si} d}}{E_{\phi} \mu_{\text{км возд.}}} ; \quad (1)$$

где  $N_0$  - скорость счета при уровне дискриминации, равном нулю;  $P$  - мощность экспозиционной дозы;  $E_{\phi}$  - энергия фотонов;  $\mu_{Si}$  - линейный коэффициент ослабления в кремнии;  $\mu_{\text{км возд.}}$  - массовый коэффициент передачи энергии в воздухе;  $d$  - ширина чувствительной области,  $S$  - площадь чувствительной поверхности детектора. Представляет интерес влияние величины порога регистрации (уровня амплитудной дискриминации) импульсов на чувствительность и "ход с жесткостью".

Обозначим через  $N_{\text{п}}$  скорость счета при пороге регистрации  $E_{\text{п}}$ . В интервале энергий фотонов  $E_{\phi} = (0, 1+3) \text{ МэВ}$  в кремнии преобладает комптоновский эффект взаимодействия. Тогда, в случае однократного рассеяния и при толщине чувствительного слоя, превосходящей максимальный пробег вторичных электронов, интегральный спектр импульсов в первом приближении может быть представлен прямой линией, пересекающей энергетическую ось в точке максимальной энергии комптоновских электронов  $E_{\text{max}}$ , а ось скорости счета - в точке  $N_0 \cdot \frac{E_{\text{п}}}{E_{\text{max}}}$ . Аналитически такая линия представляется выражением:  $N_{\text{п}} = N_0 \left(1 - \frac{E_{\text{п}}}{E_{\text{max}}}\right)$ . Тогда зависимость чувствительности от уровня дискриминации можно выразить соотношением:

$$\frac{N_{\text{п}}}{PS} = \frac{1 - e^{-\mu_{Si} d}}{E_{\phi} \mu_{\text{км возд.}}} \left(1 - \frac{E_{\text{п}}}{E_{\text{max}}}\right); \quad (2)$$

Соответствующие экспериментальные исследования выполнены с золото-кремниевыми и кремний-литиевыми детекторами, отличающимися толщиной чувствительного слоя.

Измерялись дифференциальные спектры вторичных электронов, по которым строились кривые зависимости чувствительности от уровня дискриминации для шести различных значений толщины чувствительной области в интервале (0,04+0,2) см.

На рис. 1 приведены экспериментальные результаты для толщины 0,08 и 0,2 см в сравнении с данными, рассчитанными по формуле (2).

Значения чувствительности, полученные в эксперименте для всех использованных толщин чувствительного слоя, отличаются от расчетных на фактор 2 и меньше, а само отклонение носит систематический характер. Такое расхождение определяется, по-видимому, смещением центра тяжести реального спектра импульсов по отношению к идеализированному в область низких энергий вторичных электронов. По этой же причине расхождение проявляет тенденцию к уменьшению при снижении порога регистрации, а значения чувствительности при пороге, равном нулю, полученные экстраполяцией экспериментальных кривых, отличаются от расчетных не более, чем на 30+40%, что находится в пределах экспериментальной ошибки измерения и погрешности в определении величины толщины чувствительного слоя.

Следовательно, формулу (2) можно использовать для грубой оценки чувствительности счетного режима работы кремниевых детекторов (с толщиной чувствительного слоя 0,04 см) в качестве дозиметров.

С целью исследования порога регистрации импульсов на "ход с жесткостью", по формуле (2) строились кривые зависимости чувствительности от энергии фотонов при различных уровнях дискриминации (рис. 2). Как показано на рисунке, "ход с жесткостью" существенно

зависит от уровня дискриминации, что дает возможность понижать его в известных пределах (для интервала энергий  $0,3+3$  Мэв вплоть до  $\pm 25\%$ ), правда, за счет некоторого уменьшения чувствительности, и применять кремниевые детекторы в дозиметрии полей с небольшим градиентом качества. На рис. 2. приведены экспериментальные значения дозовой чувствительности для энергий фотонов  $0,661$  и  $1,25$  Мэв. На рисунке видно, что при переходе от энергии  $0,661$  Мэв к  $1,25$  Мэв с порогом регистрации  $0,15$  Мэв чувствительность уменьшается в  $(2,0 \pm 0,4)$  раза по сравнению с  $1,9$ , полученным в расчете, а при пороге  $0,3$  Мэв - в  $(1,4 \pm 0,3)$  раза против расчетного  $1,3$ .

Одним из критериев, определяющих область применения дозиметра, является интервал мощностей доз, доступных измерению. Максимальный уровень регистрируемых кремниевым детектором в счетном режиме мощностей доз ограничивается разрешающим временем используемых импульсных электронных схем и составляет около  $(0,1-0,01)$  р·мин<sup>-1</sup>. Нижний предел чувствительности определяется наряду с геометрическими параметрами детектора (площадь и толщина чувствительной области), его фоновой скоростью счета. Минимальная мощность экспозиционной дозы, доступная измерению использованными в данной работе детекторами, составляет  $10^{-6}$  р·мин<sup>-1</sup>.

Полученные данные согласуются с теми отрывочными сведениями относительно чувствительности, которые опубликованы в работах 4,5.

Результаты настоящей работы показали, что сочетание интегрального и счетного режимов дает возможность выполнять измерения в диапазоне мощностей экспозиционных доз  $(10^{-6} + 10^2)$  р·мин<sup>-1</sup> с помощью одного и того же детектора (всего малых размеров), а выбором порога регистрации установить "ход с жесткостью", не превышающей  $\pm 25\%$  от среднего значения чувствительности в интервале энергий  $(0,3+3)$  Мэв.

#### РАДИОМЕТРИЯ С ПОМОЩЬЮ ПОЛУПРОВОДНИКОВЫХ ЗОЛОТО-КРЕМНИЕВЫХ ДЕТЕКТОРОВ В ИССЛЕДОВАНИЯХ РАДИАЦИОННОЙ ОБСТАНОВКИ.

Измерения малых активностей и идентификация их изотопного состава связаны с использованием аппаратуры, обладающей хорошим энергетическим разрешением и высокой чувствительностью к конкретному виду излучения. Как известно, чувствительность радиометрической аппаратуры определяется геометрической и физической эффективностью регистрации излучения и фоновой скоростью счета, которая у полупроводниковых детекторов, очевидно, в определенной степени зависит от условий их изготовления и радиационной частоты конструкционных материалов.

Проведено исследование фоновой скорости счета ответственных поверхностно-барьерных золото-кремниевых детекторов с площадью рабочей поверхности ( $S$ ) от  $0,2$  до  $9$  см<sup>2</sup> и мозаичных структур на их основе с площадью до  $60$  см<sup>2</sup>, а также диффузионно-дрейфовых кремний-литиевых детекторов с  $S=0,3+3$  см<sup>2</sup> и толщиной чувствительной области от  $0,4$  до  $2$  мм. Энергетическое разрешение отдельных детекторов при комнатной температуре ( $+20^\circ\text{C}$ ) составляло от  $30$  до  $160$  Кэв.

Обнаружено, что в области энергии  $3+9$  Мэв, являющейся рабочей областью для альфа-спектрометрии радиоактивных изотопов, интегральная фоновая скорость счета как отдельных золото-кремниевых детекторов с площадью рабочей поверхности от  $1$  до  $9$  см<sup>2</sup>, так и группы параллельно соединенных детекторов с общей площадью до  $60$  см<sup>2</sup>, пропорциональна величине  $S$  и составляет  $(0,22 \pm 0,02)$  час<sup>-1</sup>см<sup>2</sup>. Примерно  $1/3$  этой величины обусловлена эманациями и аэрозолями,

содержащимися в воздухе лабораторного помещения.

При увеличении площади рабочей поверхности золото-кремниевых детекторов до 8-10 см<sup>2</sup> их геометрическая эффективность возрастает до 30±35% для источников с площадью активного пятна около 5 см<sup>2</sup>. Разрешающая способность детекторов таких размеров не хуже 2±3% на линии 5,15 Мэв, что обеспечивает возможность идентификации и измерения практически всех естественных альфа-радиоактивных изотопов, находящихся в смеси в малых количествах (порядка 10<sup>-14</sup> кюри в пробе).

В оценке радиационной обстановки при измерении загрязненности рабочих поверхностей необходимы датчики с относительно большим S (~100 см<sup>2</sup>), пригодные для спектрометрического анализа изотопного состава загрязненности непосредственно на месте контроля. Для этой цели не пригодны ни отдельные золото-кремниевые детекторы с большой площадью, ни мозаичные структуры на основе детекторов серийного производства, поскольку разрешающая способность таких систем при комнатной температуре слишком мала, и не может быть повышена до удовлетворительной величины (2±3%) в силу ограничений принципиального свойства.

В данной работе предлагается использовать для измерения поверхностной загрязненности альфа-радиоактивными веществами и их идентификации мозаичный датчик с большой площадью, состоящий из нескольких параллельно соединенных полупроводниковых детекторов, один из которых отключается от остальных для выполнения спектрометрических измерений. Предложенный принцип сочетает достоинства мозаичной структуры, обеспечивающей возможность создания датчика с требуемой площадью чувствительной поверхности (до 100 см<sup>2</sup> и выше), с высокими спектрометрическими свойствами отдельного детектора, площадь которого (5±6 см<sup>2</sup>) не велика по сравнению с площадью мозаики.

Блок-схема прибора приведена на рис. 3. Мозаичный датчик, испытанный в настоящей работе, состоял из 12 поверхностно-барьерных золото-кремниевых детекторов, с площадью рабочей поверхности равной 5 см<sup>2</sup> и энергетическим разрешением близком к 70 кэв, производство которых освоено отечественной промышленностью.

Защиточувствительный преусилитель с большой входной динамической емкостью смонтирован в блоке детектирования вместе с детекторами. Энергетическое разовшение прибора в спектрометрическом режиме работы составило 140 кэв при измерении спектра излучения источника плутония-239 в воздухе. Фоновая скорость счета всей мозаики составила 15 имп·час<sup>-1</sup>.

Отбор детекторов проводился, в основном, по величине максимально допустимого напряжения смещения. Другой критерий - требование идентичности величины оптимального рабочего смещения для всех детекторов мозаики оказался менее критичным, поскольку зависимость энергетического разрешения от напряжения смещения, как правило, у таких детекторов не имеет острого минимума. Годными для использования в мозаике оказались 70% детекторов одной классификационной группы, что говорит о доступности использованного принципа и возможности его широкого применения.

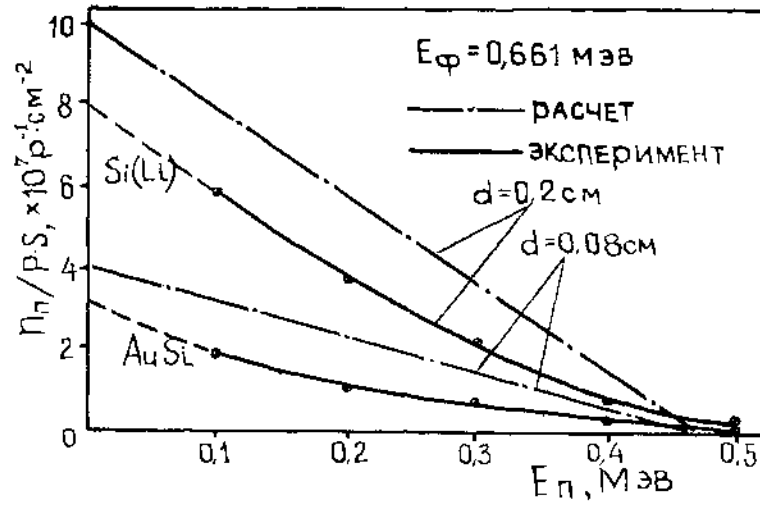


Рис. 1. Дозовая чувствительность в зависимости от порога регистрации импульсов для энергии излучения 0,661 Мэв.

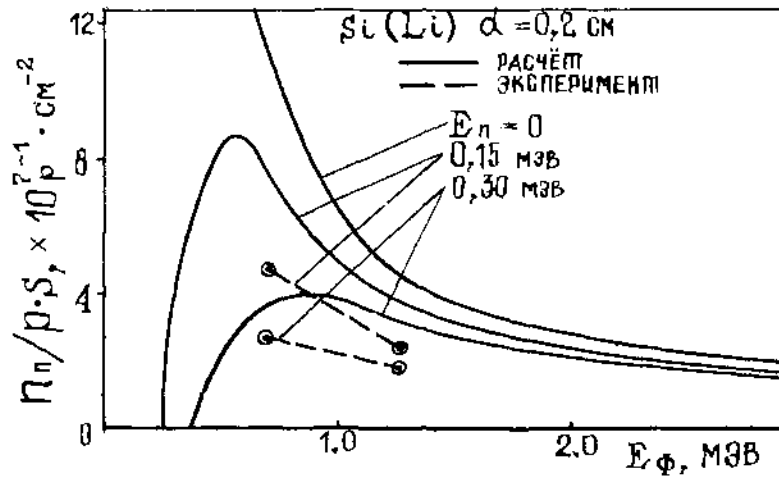


Рис. 2. Зависимость дозовой чувствительности от энергии излучения при различных порогах регистрации.

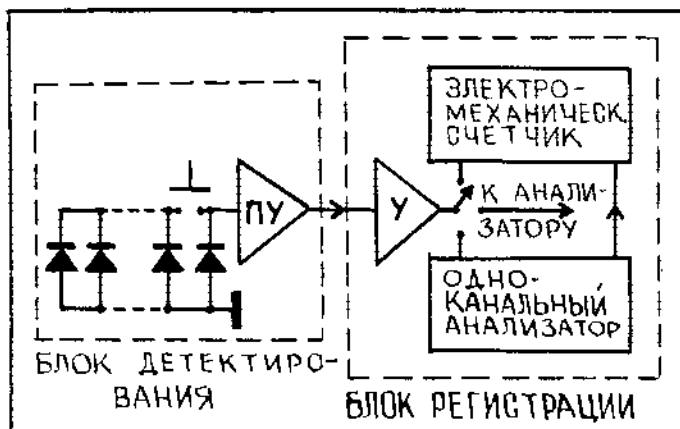


Рис. 3. Блок-схема прибора для измерения поверхностной загрязненности альфа-радиоактивными веществами и их идентификации.

#### ЛИТЕРАТУРА.

1. Scharf, K., 1967, Health Physics, 13, 575.
2. Parker, R.P., Johnson, P.F., and Baker, I.W., 1969, Br. J. Radiol., 42, 69.
3. Машьянов, А.В.; Петушинов, А.А., Глезин Ф.И. 1969, Медицинская радиология, № 1, 53.
4. Jones, A.R., Health Physics, v. 8, № 1, 1.
5. Yablonovith, E. "Health Physics Applications of Thin Silicon Detector", Chalk River, 1967 (AECL-2766).

HIGH ENERGY PHOTON DETECTION  
BY  
FISSION TRACK REGISTRATION

Hiroyoshi Iida and Tokusaburo Koshijima  
National Institute of Radiological Sciences  
Chiba-shi, Japan

Abstract

An attempt is made in this report to detect high energy photons above 5 MeV by photo fission reaction. A solid state fission track detector, in which one face of a solid block was coated with fissionable material, was used as a high energy photon detector.

Typical results obtained with bremsstrahlung at maximum energy of 29 MV are as follows. Fission track density per 100 R was found to be about 1,000 tracks/cm<sup>2</sup>. The number of high energy photons over 5 MeV per 100 R from the total bremsstrahlung was estimated to be  $4 \times 10^9$ /cm<sup>2</sup>, assuming that the effective cross section of natural uranium is approximately 0.05 barn for the bremsstrahlung.

The track detector presented in this report may provide a useful means to detect high energy photons even in the presence of other radiations except neutrons, and is expected to be utilized for the monitoring of high energy photons.

Introduction

For the measurement of high energy X- or gamma-rays above several MV or MeV, nuclear reactions of high energy photons have been applied in some instances in addition to usual measurement by ionizing process. The measurement by means of photo fission has an advantage in that the high energy photons can be distinguished from a mixed radiation field except neutrons.

In the present report, a method was examined for high energy photon detection by solid state fission track detector registering the tracks of fission fragments produced by photo fission reaction. A solid state fission track detector which had been previously developed by us for neutron detection was used to detect the high energy photons.

Experiment

The track detector consists of a solid for registration of fission fragment track and fissionable material. There are several ways of combination, e.g., U-doped glass or a glass block in contact with an uranium foil. The method adopted in the present study was to bring a solid in contact with a fissionable material. Silver activated phosphate glass of Toshiba FD-P8-3,



a commercially available dosimetric glass, was employed as the solid because of its easiness for etching and optically almost perfect surface. Uranylacetate,  $\text{UO}_2(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ , an easily available reagent, was used as a fissionable material. A saturated solution of uranylacetate was made at 30 - 40°C, then 0.1 - 0.2 ml of the solution was applied dropwise and dried on a glass block which was thoroughly pre-rinsed with distilled water, forming a layer of 15 - 30 mg/cm<sup>2</sup> thickness tightly attached on the surface of the glass block (Fig. 1). The layer is thicker than the maximum range of fission fragments, so that there is no effect of thickness on the track density.

The detectors were exposed in the range of 100 to 200 R to X-rays from the NIRS betatron (Toshiba) without any flattening filter at maximum bremsstrahlung energies of 15 to 29 MV. Exposure was measured by the Victoreen thimble ionization chamber, but the chamber could not be irradiated simultaneously with the detector owing to narrow X-ray beam.

To investigate the effect of exposure rate on track density, irradiation was made at variable exposure rate from 25 to 60 R/min at maximum energy of 29 MV.

After irradiation, detectors were cleaned thoroughly by use of an ultrasonic washer to remove the uranylacetate layer, and then radiophotoluminescence of the glass block was measured with a photoluminescence reader. Because of the high energy of X-rays in excess of the applicable range for the glass dosimeter, the reading does not correspond to the absolute exposure but rather its relative value. Finally, the glass blocks were etched by a 30 % solution of sodium hydroxide at 80°C for 10 min to make visible tracks which were counted under a microscope (Fig. 2). Track density was estimated by counting the number of tracks in 4 to 5 fields of 2.5 mm<sup>2</sup> on each glass block. Background was estimated from the opposite side of the glass block.

Experiments were carried out in the same way on thorium-chloride.

### Result

Hereinafter, the value of radiophotoluminescence in roentgen obtained with the reader is expressed by (RPL).

The track density per 100 R of X-ray at maximum energies of 15, 20, 25 and 29 MV is shown in Fig. 3. The sensitivity of the detector for X-ray at maximum energy of 29 MV is about 1,000 tracks/cm<sup>2</sup> per 100 R, which is the average from several independent experiments. Effect of exposure rate on the track density is small as shown in Fig. 5.

In the case of thoriumchloride, significant result could not be drawn due to the insufficient number of fission tracks.

### Discussion

Because of fluctuation in X-ray intensity during irradiation especially at low energy range and of the irradiation process of detector which was not made simultaneously with the ionization chamber, the relative exposure dosimetry with the glass block (RPL) is considered to be more reliable than that with the chamber. Fig. 4 shows the energy characteristic of the detector. The track density per (RPL) tends downward with decreasing energy.

Fig. 6 shows photo fission cross section of uranium and

thorium.<sup>2</sup> Maximum cross section for photons was found to be about 15 MeV for both uranium and thorium. The higher the maximum energy of X-ray, the higher efficiency of detection was observed on the fission track detector as shown in Fig. 4. This seem to be due to the fact that the X-ray spectrum is continuous and that for the same exposure the photons of over 5 MeV is fewer at low maximum energy than at high maximum energy. In mono-energy photon flux such as gamma-rays, the highest efficiency of detection may be obtained at about 15 MeV. In either case, the detection of photons of below 5 MeV is impossible, since the cross section for them is practically zero.

The number of high energy photons contained in 100 R of X-rays at maximum energy of 29 MeV are calculated as below. The relation between track density  $\rho$  and photon flux  $\Phi$  is given by

$$\rho = \Phi \cdot \sigma \cdot N \cdot f$$

where  $\sigma$  is the photo fission cross section of uranium, N, the number of uranium atoms in 1 mg of uranylacetate, and f, the ratio of track density to the number of fissions occurred in 1 mg of uranylacetate layer which has a thickness larger than the range of fission fragments. From the experimental data on neutron irradiation, the value of f is estimated at 10/3 (track  $\cdot$  cm<sup>-2</sup>/fission  $\cdot$  mg<sup>-1</sup>). The number of high energy photons over 5 MeV per 100 R of X-ray was calculated to be approximately  $4 \times 10^9$ /cm<sup>2</sup>, assuming that the effective fission cross section of uranium is 0.05 barn for the X-ray.

The tracks on a glass block indicate only the existence of high energy photons over 5 MeV in radiation field except neutrons. Therefore, the exposure of X-ray which has continuous energy spectrum of photons can not be determined from the track density. In such a case as to investigate the primary effect of high energy photons, the fission track detector may provide a useful means for the counting of high energy photons even in the presence of other type of radiations except neutron, and is expected to be utilized in the monitoring of high energy photons.

#### References

1. H. Iida, T. Koshijima and H. Takahara: "Neutron Detection by Fission Track Registration" *Austl. Radiol.* 16, 427-433 (1972)
2. J. E. Gindler, J. R. Huizenga and R. A. Schmitt: "Photofission and Photoneutron Emission in Thorium and Uranium" *Physical Review* 104, 425-433 (1956)

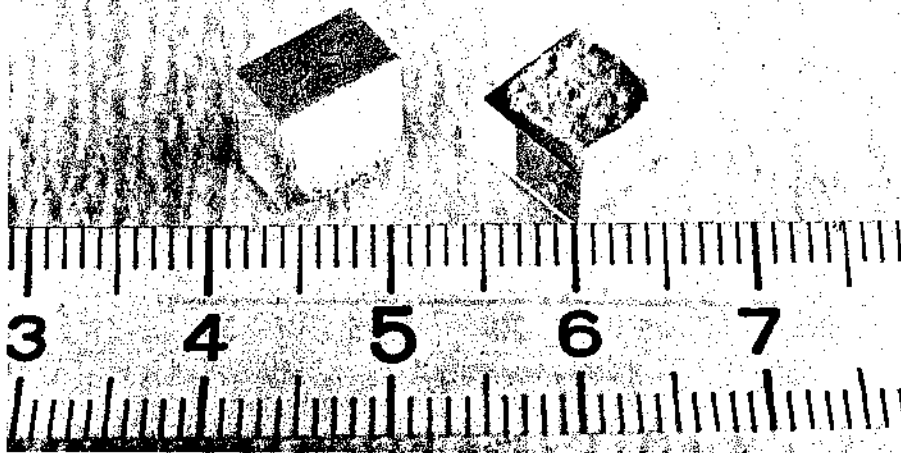


Fig. 1. Detectors, minor unit of scale in mm.

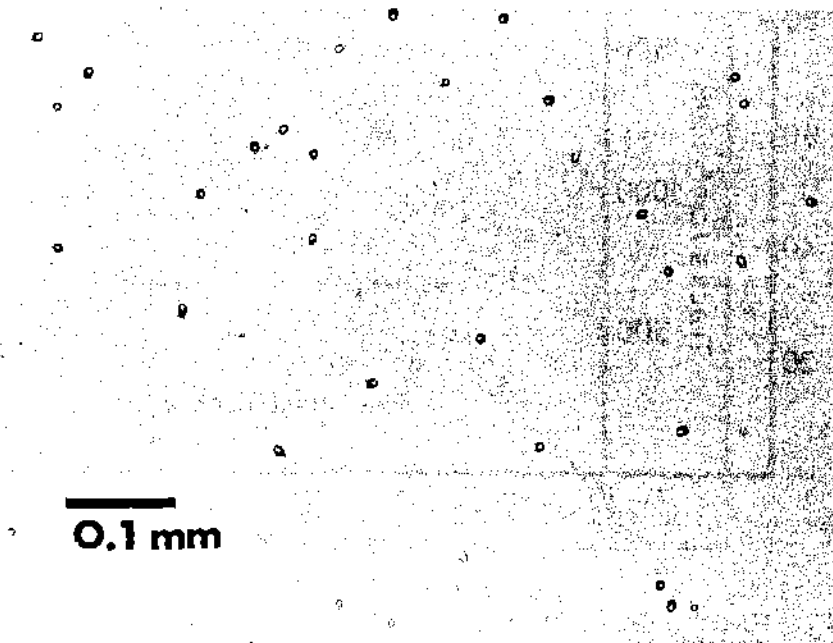


Fig. 2. Fission track etch pit in glass irradiated by X-ray at maximum energy of 29 MV from betatron.

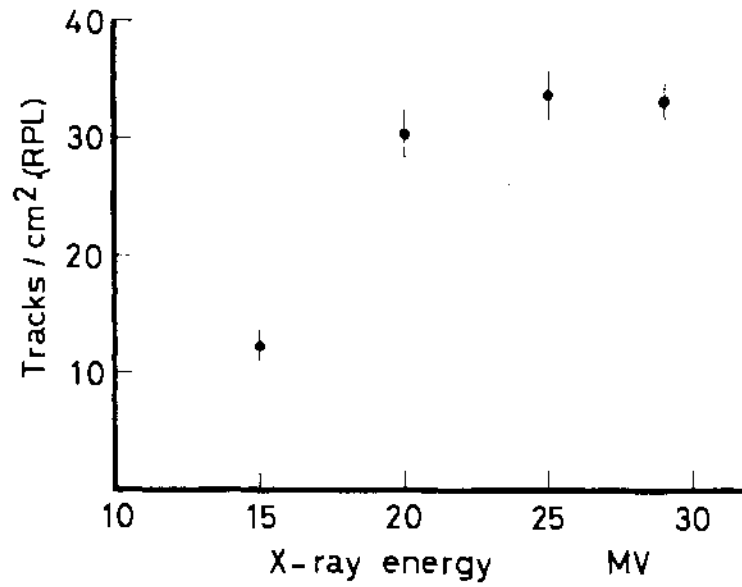


Fig. 3. Track density per 100 R of X-ray at maximum energies of 15 to 29 MV and (RPL).

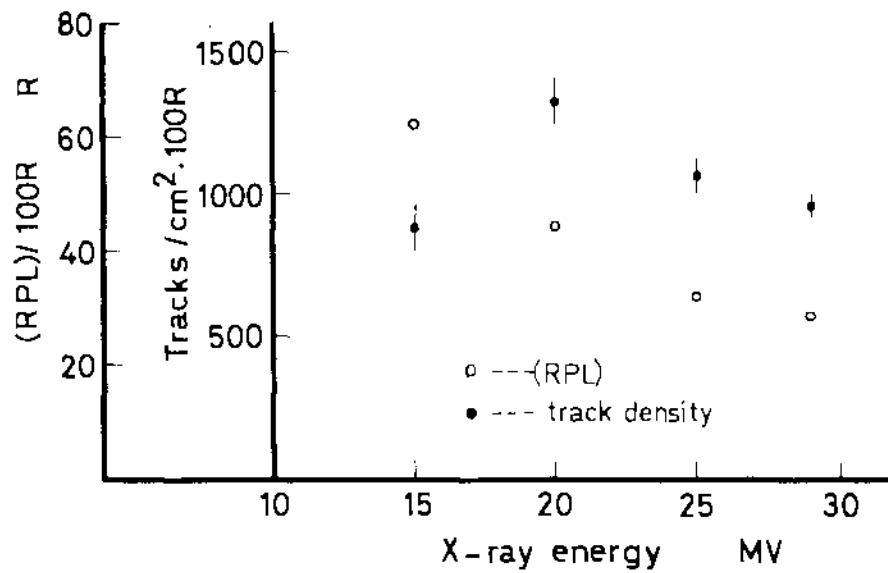


Fig. 4. Track density per (RPL) at maximum energies of 15 to 29 MV.

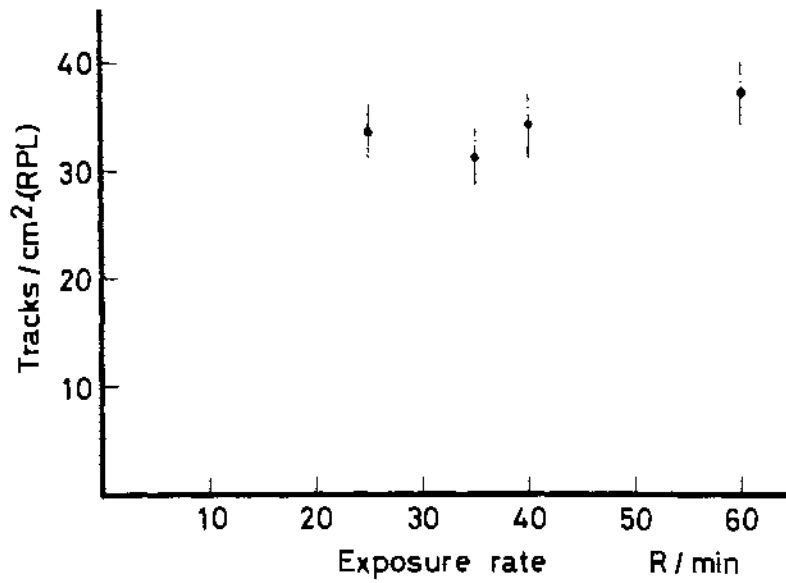


Fig. 5. Effect of exposure rate on track density at maximum energy of 29 MV.

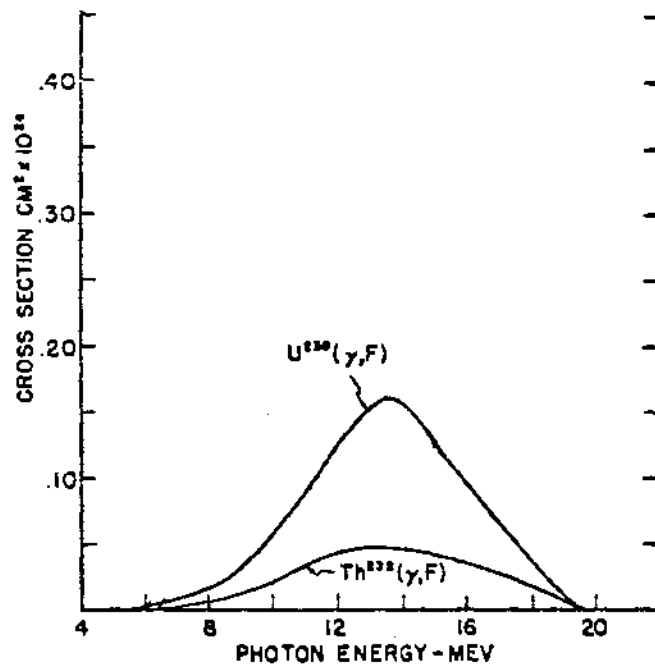


Fig. 6. Photo fission cross section of uranium and thorium.

## AN INEXPENSIVE LIGHTWEIGHT ENVIRONMENTAL SURVEY INSTRUMENT

I. M. G. Thompson\* and H. C. Orchard<sup>†</sup>

\* Central Electricity Generating Board, Berkeley Nuclear Laboratories  
Berkeley, Gloucestershire, U.K.

<sup>†</sup> Central Electricity Generating Board, Nuclear Health & Safety Department,  
London, U.K.

### Abstract

The operation of power producing nuclear reactors in the U.K. involves a statutory requirement to measure radiation levels in the area surrounding each nuclear site. The natural radiation background is measured at specified locations prior to operation of the reactor and afterwards routinely every three months. A survey instrument used for such measurements must therefore measure exposure levels from background (typically a few  $\mu\text{R.h}^{-1}$  in the U.K.) upwards. Other requirements for the instrument are that it should be very portable, be capable of being read accurately at low exposure rates, and should maintain its calibration.

The instrument described in this report weighs only 10 lbs, has a good energy response,  $\pm 15\%$  over the range 50 keV to 6 MeV, and measures exposure levels from a few  $\mu\text{R.h}^{-1}$  to 100  $\text{mR.h}^{-1}$ . Ambiguities resulting from reading a fluctuating meter needle at low exposure rates are overcome by integrating the count over a preselected time, digital readout being provided. The report describes a comprehensive evaluation of the instrument and its comparison with other commercially used instruments. The instrument price is considerably cheaper than that of comparable instruments.

### Introduction

Measurement of gamma radiation in the environment, natural prior to the operation of a power reactor and afterwards natural plus that due to the reactor, requires an instrument that records levels from a few  $\mu\text{R.h}^{-1}$  upwards. As the spectral content of this radiation may be unknown the exposure rate response of the instrument should be as flat as possible over a wide energy range, say 30 keV to 7 MeV.

Portable survey instruments used by the C.E.G.B. for this work indicate the measured exposure rate by means of a moving coil meter display.<sup>1</sup> The reading of such meters at low exposure rates involves problems of interpretation where a needle is fluctuating over a significant range of readings.

Recent advances in electronics have made it possible to produce small portable scalers with digital readout. In addition energy compensating filters are now commonly used to improve the poor energy characteristics of G.M. counters. To keep the total cost low, the instrument described in this report makes use of a commercially available portable scaler, G.M. circuitry and energy compensated G.M. counters.

## Instrument Description

Four energy compensated G.M. counters, 20th Century type B6T's, are mounted radially on a tripod (Fig.1) to give the best directional response and to enable measurements to be made at 1 metre above ground. A short co-axial cable connects the detectors to the input of the electronics.

The electronics consist of the amplifier and E.H.T. unit of a Mini Monitor Mark V, and a Mini Instruments portable scaler type MS6.10. Both these units are housed in a small metal case, 20 cm x 14 cm x 12½ cm. Although the instrument is intended for outdoor use no special seals were fitted to the prototype to make it waterproof. During field trials a large polythene bag was slipped over the detectors and electronics and the measurements on Trawsfynydd Lake during a wet day and on rough water showed this to be adequate protection. Power supplies to the G.M. amplifier and E.H.T. unit are from two 9 volt batteries type PP6. The scaler unit is powered by four 6 ampere-hour Nickel-Cadmium rechargeable batteries and will run up to 10 hours continuously on a single charge.

Each G.M. counter was connected separately and its operating voltage plateau determined. The plateau was also measured with all four counters connected to verify that the same voltage range was obtained and the operating voltage for the counters was set at 680 volts.

## Evaluation of Instrument

### Laboratory Tests

#### Energy Response

The photon energy spectrum of a measured radiation field is frequently unknown and therefore an environmental survey instrument is required to have a 'flat' response over a wide energy range. Many reactors produce significant amounts of 6 MeV gamma radiation, and in gas cooled reactors this arises from the  $^{16}\text{O}$  excited state which is formed by the fast neutron capture in  $^{16}\text{O}$  of the  $\text{CO}_2$  cooling gas and the subsequent beta decay of  $^{16}\text{N}$  to  $^{16}\text{O}^*$ .

Tests were made at 29, 47, 59, 85, 107, 147, 183 and 210 keV using an improved low exposure-rate, filtered, X-ray series whose spectra have resolutions of 20%.

Radionuclide sources were used to measure the response above 200 keV up to 1.33 MeV.

The 335 keV resonance of the  $^{19}\text{F}(p,\alpha)^{16}\text{O}$  reaction was used to determine the 6 MeV response, the radiation field being standardised by associated particle counting of the alpha particles with cross-checks by ionization measurements.

Fig.2 shows the energy response obtained. The response expressed as instrument reading divided by standardised exposure-rate is plotted against photon energy and has been normalised at 0.8 MeV.

#### Linearity of Response

Linearity was tested with standard  $^{226}\text{Ra}$  sources and  $^{60}\text{Co}$  sources. Results are given in Fig.3 and show that the instrument has a linear response up to  $10 \text{ mR.h}^{-1}$ . Although the response is non-linear above this exposure rate no fall-back effects are observed until about  $4 \text{ R.h}^{-1}$  and at approximately  $100 \text{ R.h}^{-1}$  the reading has reduced by 20% compared to that at  $4 \text{ R.h}^{-1}$ . The

meter reading, however, still remains greater than full scale for exposure rates up to  $100 \text{ R.h}^{-1}$ . The apparent non-linearity below  $20 \text{ } \mu\text{R.h}^{-1}$  is discussed later on.

#### Temperature Tests

As the instrument will be used outdoors throughout the year it is important that its response should not change significantly with expected variations in temperature.

The instrument was placed in an environmental cabinet and irradiated in a constant field from a  $^{60}\text{Co}$  source, the temperature was varied over the range  $-20^{\circ}\text{C}$  to  $+50^{\circ}\text{C}$ . Before commencing these tests new 9 volt batteries were fitted and the Ni-Cd batteries were re-charged. The readings remained constant within  $109.0 \text{ cps} \pm 7 \text{ cps}$  over this temperature range except at  $+50^{\circ}\text{C}$  where a reading of 136 cps was obtained.

#### Variations in Instrument Readings with Supply Voltage

Tests were made to measure the variation in instrument reading with change in supply voltage for both battery supplies, the instrument being irradiated in a constant field. The lower limit markings for both battery tests were perfectly adequate.

#### Field Tests

Measurements were made at normal 'district survey' locations at two nuclear power stations.<sup>2</sup> Exposure rates were measured with the instrument and with other commonly used low exposure-rate instruments. The other instruments used in the comparison were the BNL 1 which has a plastic phosphor detector and a bottom range of  $0-30 \text{ } \mu\text{R.h}^{-1}$ , and the A.E.R.E. type 1368A which has 4 G.M. counters, 3 used in parallel for the lower ranges, and a bottom range of  $0-50 \text{ } \mu\text{R.h}^{-1}$ . For two of the locations measurements were also made with the Nuclear Enterprises N2601, which has an energy compensated G.M. counter and a single log range of  $0 - 10 \text{ mR.h}^{-1}$ , and with the General Radiological 1597A which uses a NaI detector and has a bottom range of  $0 - 30 \text{ } \mu\text{R.h}^{-1}$ . All the instruments had been previously calibrated against  $^{226}\text{Ra}$  sources and the readings taken were all corrected for any non-linearities.

The first measurements were made at Trawsfynydd Lake, close to the Nuclear Power Station which was not operating at the time. Four sets of measurements were made above water depths between 20 to 30 feet and the following averaged results were obtained. Prototype background monitor  $4 \text{ } \mu\text{R.h}^{-1}$ , BNL 1 instrument  $5 \text{ } \mu\text{R.h}^{-1}$  and 1368A instrument  $7.5 \text{ } \mu\text{R.h}^{-1}$ .

A second series of measurements were made at another station, with the reactors on load. Two different one mile locations were selected at which the results in Table 1 were obtained. Each reading for the meter display instruments was obtained by observing the needle for approximately one minute and taking the average reading, the figures in brackets show the range of fluctuations of the instantaneous reading.



Table 1

Instrument Type	Corrected Instrument Reading for 1 mile locations in $\mu\text{R.h}^{-1}$	
	Location 1	Location 2
Prototype background monitor	11	10.2
BNL 1	9(7 to 11.3)	8(6.5 to 10.2)
1368A	11.5(10 to 12)	11(10 to 11.7)
1597A	13(10.7 to 15.3)	10(8.5 to 11.5)
2601	12(9.5 to 17.3)	11.5(6.5 to 19.5)

The following conclusions have been made from these comparisons. Accurate assessment of very low exposure rate levels is difficult and differences of only  $1 \mu\text{R.h}^{-1}$  between different instruments must be considered good. Measurements on Trawsfynydd Lake were made at lower exposure rate levels than the background radiation level of the room used for instrument calibration and corrections for non-linearity have therefore been obtained from extrapolation. No standard instrument will measure levels in the  $\mu\text{R.h}^{-1}$  region and hence no accurate measurement can be made of the calibration room background level. The assumed value of  $6.5 \mu\text{R.h}^{-1}$  is an averaged extrapolated result of measurements made in this room with a large number of different instruments of several types. Uncertainty in the absolute value of the room background may contribute to non-linearity of the prototype below  $20 \mu\text{R.h}^{-1}$  (Fig. 3).

For the Lake measurements the radiation field is principally due to cosmic radiation with a small contribution from any radioactive content in the Lake. Levels of about  $3.5 \mu\text{R.h}^{-1}$  are normally reported for cosmic radiation and so the readings of  $4 \mu\text{R.h}^{-1}$  for the prototype and  $5 \mu\text{R.h}^{-1}$  for the BNL 1 instrument appear realistic values.<sup>3</sup> The reading of the 1368A is just over double the cosmic radiation level and may be due in part to the built-in radioactive content of the detectors. This built-in activity would be allowed for in setting the instrument up at  $6.5 \mu\text{R.h}^{-1}$  but would cause it to read high if it were placed in a lower level radiation field.

Looking at the results for the other station it is interesting to see the large variations in readings obtained. For the one mile locations the BNL 1 instrument gave lower results than for all the other types. The 2601 instrument readings were approximately  $1 \mu\text{R.h}^{-1}$  higher than the prototype results, this good agreement is not too surprising since the 2601 uses a single G.M. tube which is identical to the 4 detectors used in the prototype. The large fluctuations in the 2601 readings are due to the poor statistics obtained by using a single small detector. At these 1 mile locations the prototype and the 1368A are in good agreement and apart from location 1, so is the 1597A.

#### Conclusions

The tests on the prototype environmental survey monitor have shown that it compares favourably with other commercial low-level instruments whilst not having the interpretation problems associated with a meter display. The instrument has a good energy response ( $\pm 15\%$  for 34 keV to 6 MeV), is simple and easy to use, and will cost approximately £280 ( $\sim$  \$670).

The environmental monitor described is only a prototype and a number of changes will be introduced in the operational instrument, for example the

controls will be simplified and the meter markings altered to  $\mu\text{R.h}^{-1}$ .

Many of the differences in readings observed when using different types of instruments arise from the problems of calibrating instruments at a few  $\mu\text{R.h}^{-1}$  and further investigations are required on the calibration techniques and standardisation at these radiation levels.

#### Acknowledgements

It is a pleasure to acknowledge the helpful assistance of Dr. P. W. Roberts of Mini Instruments Limited who constructed the initial instrumentation. This paper is published by permission of the Central Electricity Generating Board.

#### References

1. Roberts, W. E., Speight, D. L., "Gamma Scintillation Dose-Rate Meter Type BNL 1", C.E.G.B. Report (1964).
2. Jones, J. K., Lewis, G., Orchard, H. C., Owers, M. J. and Skelcher, B. W., "The Experience of the Central Electricity Generating Board in Monitoring the Environment of its Nuclear Power Stations", Health Physics, Vol. 24, No. 6, 619 (1973).
3. Gibson, J. A. B., Richards, J. E. and Docherty, J., "Nuclear Radiation in the Environment: Beta and Gamma-Ray Dose Rates and Air Ionisation from 1951 to 1968", A.E.R.E. - R5807 (1968).

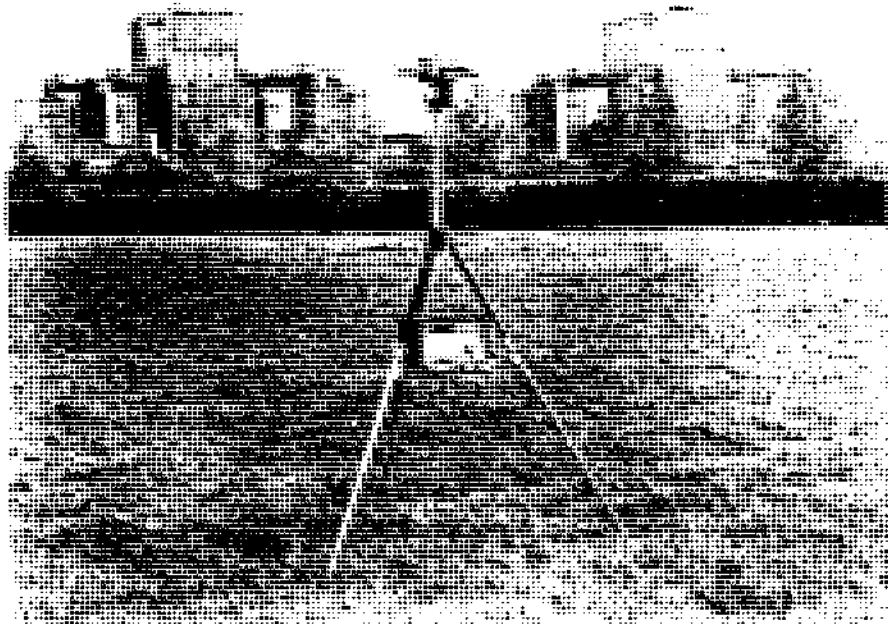


Fig.1 Lightweight Environmental Survey Meter

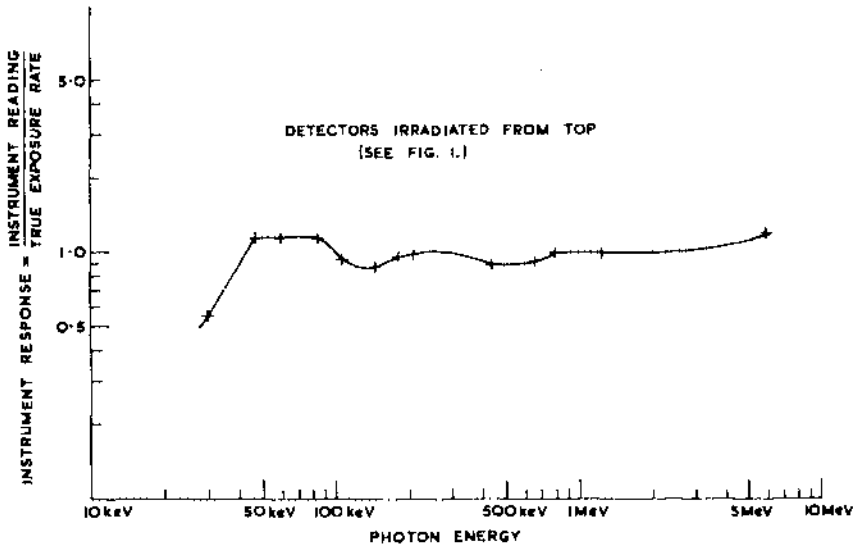


FIG. 2. PHOTON ENERGY RESPONSE.

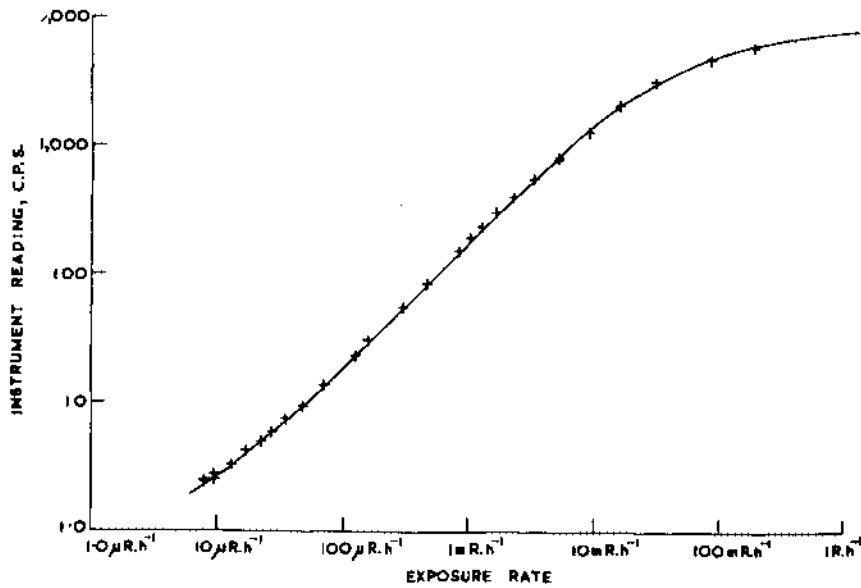


FIG. 3. LINEARITY OF BACKGROUND MONITOR.

A NEW TECHNIQUE IN  
ENVIRONMENTAL NEUTRON SPECTROSCOPY

Miguel Awschalom and Larry Coulson  
National Accelerator Laboratory  
Batavia, Illinois

Abstract

A new phoswich<sup>1</sup> has been developed that permits the measurement of the environmental neutron spectrum using Bonner spheres. The new phoswich consists of an 8 mm diameter, 8 mm long <sup>6</sup>LiI(Eu) crystal surrounded by plastic scintillator. The use of the fast signals from the plastic scintillator in anticoincidence with the slow signals from the <sup>6</sup>LiI(Eu) gives a nearly background free <sup>6</sup>Li(n,α) <sup>3</sup>He peak. Thus, the signal to noise ratio in the vicinity of the peak is very high (20-35 to 1). An adequate number of counts is obtainable in one day for most of the spheres.

Typical counting rates for polyethylene spheres of the diameters given below are:

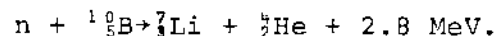
2 inch	2.69 c/hr
3 inch	13.9 c/hr
5 inch	14.0 c/hr
6.87 inch	12.4 c/hr
8 inch	9.07 c/hr
10 inch	6.91 c/hr
12 inch	5.96 c/hr
18 inch	3.77 c/hr

Details of circuitry, phoswich geometry, pulse height analyzer spectra and synthesized spectra will be given.

This paper is an elaboration of the preliminary results presented at an earlier time.<sup>2</sup>

The measurement of the neutron energy spectrum of environmental neutrons is besieged by difficulties caused by the gamma rays, muons, protons and other charged particles which are simultaneously present.

The commonly used neutron detection technique which is also applicable to environmental neutron studies is that of a <sup>10</sup>BF<sub>3</sub> counter moderated with a hydrogenous material such as polyethylene or paraffin. The use of various thicknesses of moderator produces a family of different neutron detection efficiencies which are functions of the incident neutron energies. These responses have been studied by some authors,<sup>3</sup> mostly in a direction perpendicular to the axis of the BF<sub>3</sub> counter. Responses versus energy for other polar angles have not been studied in detail to the best of our knowledge. As it is to be expected, there is a significant polar angle dependence in these counters. The popularity of the BF<sub>3</sub> counter is due to the ease with which charged particle events (either gamma ejected electrons or cosmic ray charged particles) are separated from neutron capture events by the exoergic reaction



The separation is made electronically with a simple integral discriminator. The other environmental events deposit typically a few KeV.

Another type of neutron detector is the Bonner Sphere.<sup>4</sup> This is a spherical hydrogenous (polyethylene) moderator with a point like thermal neutron detector at its center. Hence, to a first approximation it has isotropically uniform energy responses. The energy dependence of its detection efficiency has been rather well studied.<sup>5</sup> The most common point like detector is a <sup>6</sup>Li(Eu) crystal. Here in

diameter spheres, 10 inches or greater, but it is questionable if it is meaningful in smaller ones. A second way to reject the charged particle background is to use a phoswich.

The phoswich used in the experiment described in this paper is shown in Figure 3. The pulses from the  ${}^6\text{LiI}(\text{Eu})$  have a long decay time (1.4  $\mu\text{sec}$ ). The pulses from the plastic scintillator have a very short decay time (2-3 nsec). Due to the monochromaticity of the slow pulses, a very simple passive network was tried. This circuit permitted the separation of the slow and fast components after suitable passive shaping. Figure 4 shows a simplified diagram of the electronics used.

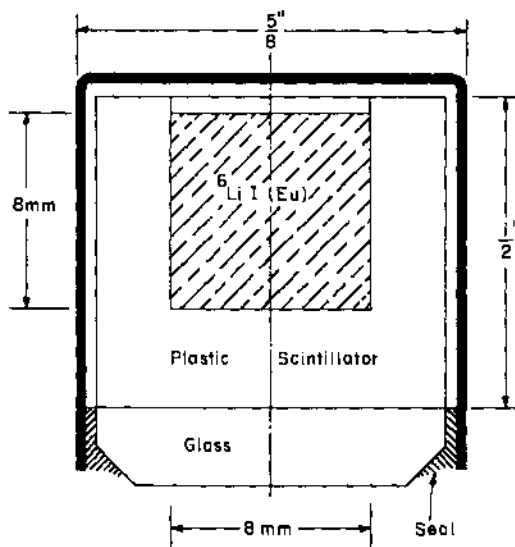


Figure 3. Cross-section of an 8 mm x 8 mm phoswich.

The values first tried and not necessarily optimum are

$$L = 102 \mu\text{H}, C = 33\text{pF}.$$

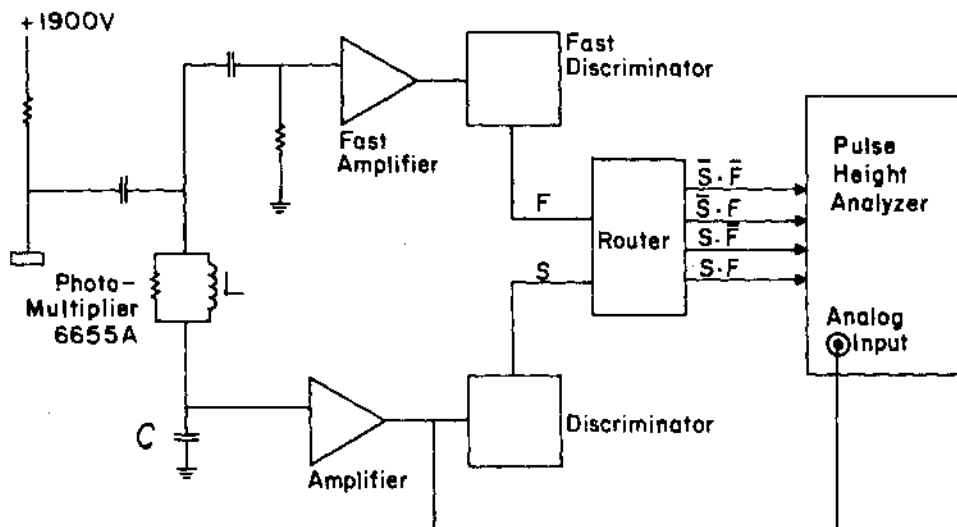
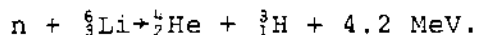


Figure 4. Electronics block diagram.

principle at least, the background charged particles are separated from the neutron capture events by choosing the size of the crystal such that most charged particles traversing it lose less energy than the 4.2 MeV of the reaction



Then again, the two types of events should be simply separable by an integral discriminator. In practice when measuring the extremely low environmental neutron flux the separation between neutron captures and other events is not good.

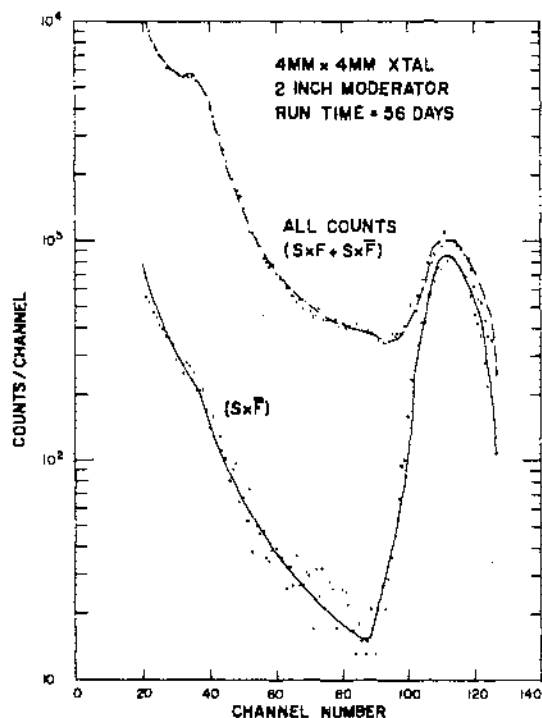


Figure 1. Upper curve is spectrum of all slow pulses. Lower curve is spectrum of slow pulses not accompanied by fast pulses, e.g., "neutrons".

Figure 1, upper curve, is a spectrum of pulses from a 4 mm x 4 mm  ${}^6\text{LiI}(\text{Eu})$  crystal in the center of a 2 inch polyethylene spherical moderator while recording environmental neutrons for 56 days. The signal to noise ratio (peak to valley) is not as good as that obtainable while calibrating the system with a PuBe neutron source as shown in Figure 2, for two sizes of crystals.

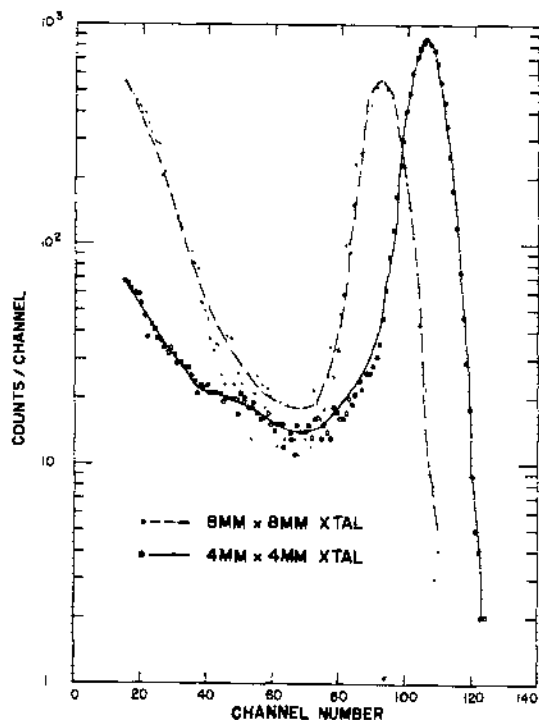


Figure 2. Spectra due to PuBe neutrons from two size  ${}^6\text{LiI}(\text{Eu})$  crystals.

There are two ways to improve the rejection of unwanted counts. One is to use small proportional counters.<sup>6</sup> This method almost works for larger

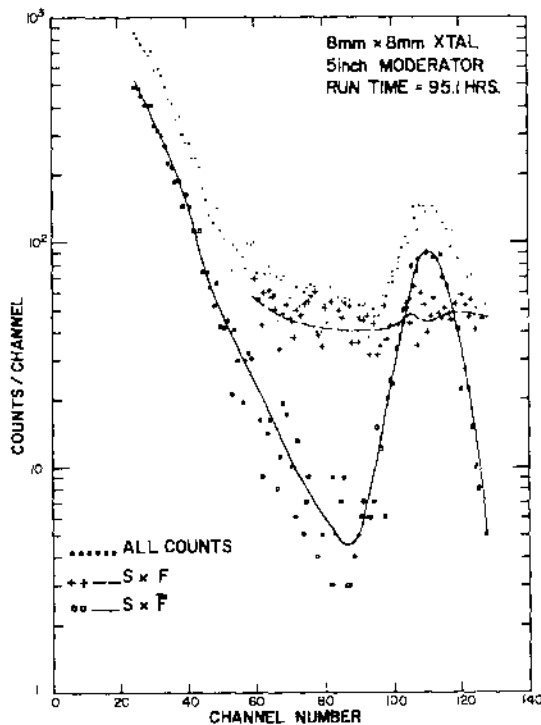


Figure 5. Pulse height spectra from an 8 mm x 8 mm phoswich in a 5 inch polyethylene spherical moderator exposed to environmental radiation for 95.1 hours.

Figure 5 shows the results of an environmental neutron measurement using an 8 mm x 8 mm  $^6\text{LiI}$  crystal in a 5 inch moderator. This run is 95.1 hours long. The solid curve shows the spectrum of slow pulses not accompanied by fast pulses ( $S \times \bar{F}$ ). These event signatures are likely to be due to neutrons. The dashed curve shows the spectrum of slow pulses accompanied by fast pulses ( $S \times F$ ).

These events are very likely "noise" or charged particles from the environment. The dotted curve shows all slow pulses ( $S \times \bar{F} + S \times F$ ). The peak-to-valley ratio for the ( $S \times \bar{F} + S \times F$ ) spectrum is about 2.8, while for the "neutron" pulse only spectrum ( $S \times \bar{F}$ ) it is about 22. Therefore, the phoswich has improved the peak-to-valley ratios for neutron detection in environmental measurements by a factor of about five to six.

Figure 1, lower curve, shows the result of using the 4 mm x 4 mm crystal in a phoswich arrangement with background rejection. Another example of the improvement that is obtainable using this technique may be seen from the curve in Figure 5. Consider the neutron data as being in channels 90 to 125. Then we could derive two figures for the number of "neutron events". Case A, use the curve for "all" events. This gives (Signal + Noise) - Noise = 3157 - 1440 = 1717 counts + 68. Case B, use the curve for ( $\bar{S} \times F$ ) type events. The corresponding difference is 1480 - 52 = 1438 + 39. The fact that two standard deviations do not produce an overlap of the data is a measure of the errors committed in background subtraction in case A.

The dashed line also shows some feed through of neutron events. Hence, improvement in the rejection of the ( $S \times F$ ) events is still possible.

The signal-to-noise ratio is now good enough that a set of Bonner spheres may be used for environmental neutron measure-

ments using a simple integral discriminator for noise rejection. Counting periods of about two days per sphere would be adequate.

Environmental Neutron Flux Measurement

A series of measurements was made inside a light frame house at the NAL site which is at an elevation of 740 feet above mean sea level. The corresponding count rates per hour are given below in Table I.

Table I

Detector Size	Counts Day	Detector Size	Counts Day
2 inch	64	8 inch	218
3 inch	334	10 inch	165
5 inch	336	12 inch	143
6.87 inch	297	18 inch	90

To interpret these results, the spectrum was unfolded using an iterative process.<sup>7</sup>

The absolute detection efficiency of the actual <sup>6</sup>LiI(Eu) crystal used was calculated by measuring the neutron flux from a <sup>238</sup>PuBe which had been calibrated by the National Bureau of Standards. The correction factor to the data in HASL-267, thus found, was 1.8<sub>8</sub>.

Using this value, the cosmic ray neutron flux was calculated as well as the dose and dose equivalent per neutron/cm<sup>2</sup>. These quantities are

cosmic ray  
neutron flux = 63 n cm<sup>-2</sup> hr<sup>-1</sup>

flux-to-dose  
conversion  
factor = 4.9 x 10<sup>-9</sup> rad n<sup>-1</sup> cm<sup>2</sup>

flux-to-D. E. conversion  
factor = 2.7 x 10<sup>-8</sup> rem n<sup>-1</sup> cm<sup>2</sup>

Conclusions

A new technique has been developed which permits rapid low resolution environmental neutron spectroscopy using simple electronics.

References

1. D. H. Wilkinson, The Phoswich a Multiple Phosphor, Rev Sci Gute 23, 414 (1952).
2. M. Awschalom, IAEA, Proc Symp Neutr Monit for Rad Prot, Vienna, Dec. 11-15, 1972 to be published.
3. See for example, references in M. Yamashita, et al., Detection Efficiency of Bare and Moderated BF<sub>3</sub> - Gas Filled Proportional Counters for Isotropic Neutron Fluxes, UCRL - 16103 (1/12/66).
4. R. L. Bramblett, et al., A New Type of Neutron Spectrometer, Nucl Instr Meth 9, 1 (1960).
5. A good review of Bonner sphere work is given by D. Nachtigall and G. Burger, Dose Equivalent Determinations in Neutron Fields by Means of Moderator Techniques, Topics on Rad Dosim, Suppl 1, N. Y. Academic Press, 1972. The latest set of efficiency calculations is from R. S. Sanna, Thirty-One Group Response



Matrices for the Multisphere  
Neutron Spectrometer over  
the Range Thermal to 400 MeV,  
HASL - 267 (3/73).

6. D. E. Hankins, The Substitution of  $\text{BF}_3$  Probe for the  $\text{LiI}$  Crystal in Neutron REM-Meters, Health Phys 14, 518 (1968).
7. M. Awschalom, Use of the Multisphere Neutron Detector for Dosimetry of Mixed Radiation Fields, IAEA, Proc Symp Neutr Monit, Vienna Aug. 29-Sept. 2, 1966.

M. Awschalom, et al., Neutron Spectrum Unfolding in Applied Health Physics, Health Phys Rev 15, 192 (1968), paper 109.

CONDUCTIMETRIC PROPERTIES OF SOME SOLID TRACK DETECTORS  
AND HYPOTHESIS ON THE TRACK FORMATION MECHANISM  
IN THESE IRRADIATED AND ETCHED DETECTORS

by Y. MARTIN-BORRET, M. BOURDEAUX, F. ROUX and C. BRIAND

Laboratoire de Biophysique, Faculté de Pharmacie, 27 Bd Jean Moulin  
MARSEILLE FRANCE.

Abstract

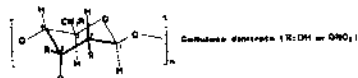
The purpose of this work is to study the conductivity variations of solid track detectors with ester function after etching and irradiation. The different conductimetric behaviour of these detectors allows to assume the chemical etching mechanism. This hypothesis seems to be confirmed by IR and RMN studies.

All the papers about latent track formation in solid track detectors point out the following condition : detectors must have electrical resistivity above 2 000  $\Omega$ /cm to record any track (1). So it is interesting to study the conductimetric properties of these insulators and especially the variation of their resistivity as a function of irradiation dose, etching time and concentration of etching reagent. For this purpose, we have studied cellulosic esters and polycarbonate films.

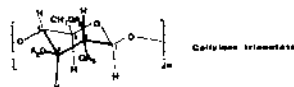
CONDUCTIMETRIC MEASUREMENTS

Experimental procedure

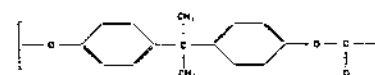
First of all, we studied a 40  $\mu$ m thick film of cellulose dinitrate. Analysis gave a nitrogen quantity of 12 % and a camphre quantity (plasticizer) of about 10 %. This material had been provided by Kodak Pathé Society (France).



Then, we used a 40  $\mu$ m thick film of cellulose triacetate without any plasticizer, purchased from Bayer chimie (France) (Triafol TN).



At last, Makrofol film (4,4' dioxidi-phenyl 2,2 propane polycarbonate) was chosen.



4,4'-dioxidi-phenyl 2,2 propane polycarbonate

These samples were irradiated with the fission fragments from <sup>252</sup>Cf (10<sup>6</sup> fissions/cm<sup>2</sup>/hours).  $\alpha$  particles from <sup>241</sup>Am were also used after being reduced to an energy in the range 0 to 2.27 Mev for a better recording.

Etching reagents were aqueous sodium hydroxyde solutions of 5N, 6,5N and 8N in a constant temperature bath (60° C).

Then etched samples were carefully washed by distilled water. At last they were kept in vacuum. No detectable variation in conductivity measurement of a particular sample was observed after a sufficient drying time (about 10 days). The electrical conductivity was measured with Wayne Kerr impedance bridge.

The measurement cell (fig. n°1) had been made in our laboratory. The electrodes were held by a insulating material in coaxial position inside two brass tubes, one of which was free of movement. This device was mounted in a rigid frame. The movable electrode was connected with the other by the mean of a screw. A torque wrench enabled a constant pression (45 Kg/cm<sup>2</sup>) on the electrodes.

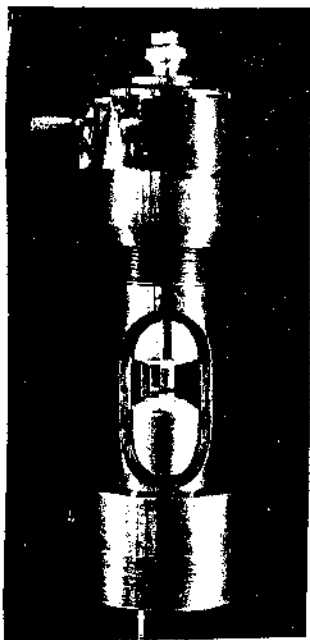


fig. 1

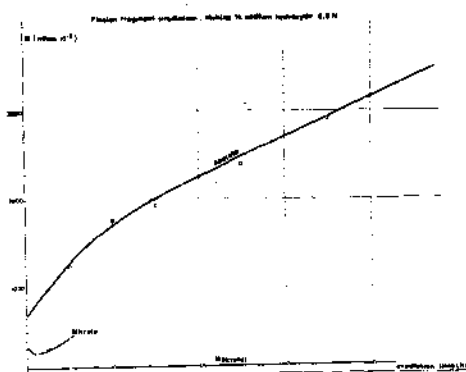


fig. 2

- The following values were found :
- cellulose acetate : 700 mhos  $10^{-9}$
  - cellulose nitrate : 310 mhos "
  - Polycarbonate : 60 mhos  $10^{-9}$

#### RESULTS AND INTERPRETATION

It is well known that ionizing particles produce breaking points in the macromolecular chain of organic polymer detectors.

The experiments were initiated in an effort to display clearly the existence of these defects by a conductimetric method. No conductimetric difference appeared when samples were irradiated but not etched even for large irradiation doses.

The etching process of tracks, making channels more or less wide, more or less deep and so substituting bulk material by air should have decreased the conductivity of the film (because air is a better insulator than cellulosic esters).

Besides, samples measured in vacuum showed a conductivity lower than in atmospheric conditions.

In fact, we observed that conductivity was increasing more or less with irradiating time, this leads to the idea that another phenomenon may be invoked, this phenomenon would increase the heterogeneity of material and consequently its conductivity. Fig. n° 2 summarises the results obtained with three detectors etched with sodium hydroxyde 6,5 N up to one hour. First of all we notice the different conductivity of these three esters etched but non irradiated.

It also appears on fig. n°2 that cellulose acetate and Makrofol show a conductivity augmentation represented by a monotonous increasing function, but this variation is very different according to the detector. In the case of Makrofol indeed, it is nearly negligible.

For cellulose nitrate, the sample destruction etched one hour after 45 minutes irradiation constrained us to reduce the etching time to 35 minutes (fig. n° 3) ; so we obtained for this compound a linear increasing function of the irradiation time (for irradiation times longer than 30 minutes).

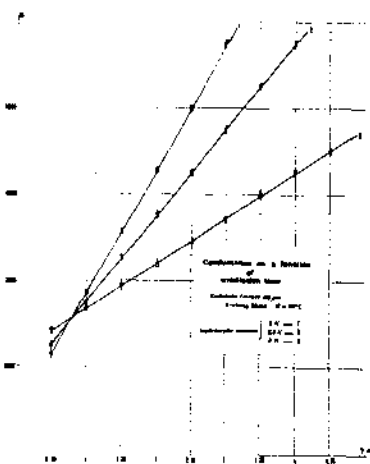


fig. 3

These conductivity augmentations can be naturally used to know the irradiation dose ; in a recent work, (2, 3) we perfected a method based on this observation in order to evaluate the track density. It is of interest to compare the behaviour of the three compounds (i.e. cellulose acetate, cellulose nitrate and polycarbonate) at short irradiation times (below 30 minutes) , the conductivity of cellulose nitrate (and only this polymer) is decreasing with irradiation dose. It is assumed that a different etching mechanism may be invoked for this detector ; this mechanism will be drawn later. Fig. n° 3 also shows the conductivity for a same irradiation time and a same etching time as a function of etching agent concentration. Of course, the augmentation of conductivity with concentration is greater, the irradiation dose is more important. In the case of irradiation by  $\alpha$  particules, similar results have been obtained but the conductivity variations observed are not so important.

We tried to explain these conductivity variations.

It is very important to notice that conductivity increase is not due to sodium hydroxyde itself remaining in tracks because the samples were washed up as long as necessary to have no more change in conductivity measurement. Of course, the thickness of the film fell off after etching but the chemical attack was lasting the same time for a line of samples so that the thickness could be modified in the same manner at the etching step.

However, if track density is very important it is possible the channels become edge to edge ; so, the decrease of thickness would be a function of the irradiation dose. It seems likely this effect would take a part in the conductimetric decrease. But that is not enough to explain the results ; computation show indeed that the observed variations would be correlated with a thickness decrease greater than the undamaged film thickness itself. So, in all probability, another phenomenon would take place.

To be precise, we used a chemical reagent and we assume the conductimetric variation is the result of a chemical damage in polymer. It is therefore possible to invoke a saponifica-

tion reaction. In the case of non irradiated etched samples, the above reaction is limited to a thin layer at the surface of material. When irradiated, the chemical attack would occur deeply along the latent tracks that allow sodium hydroxyde diffusion. Then chemical change, especially formation of new hydroxyl groups should induce a conductivity enhancement.

It would seem this saponification reaction would be slightly efficient in the case of Makrofol. For cellulose nitrate, (4) sodium hydroxyde reacts in two ways : on one hand saponification (with a low rate), on the other hand oxydation degradation (with a fast rate) which would enlarge the tracks. So in case of short irradiation times, oxydation degradation would play a leading part. Perhaps that would explain the specific conductivity decreasing above mentioned for this polymer, because the presence of air, a better insulator, in tracks. When track density was important, reagent diffusion in bulk material became easier, so that hydroxyl group number would be sufficient to increase conductivity.

Furthermore oxydation degradation would be liable for nitrate sample destruction by 45 minutes etching whereas acetate film can suffer that etching time without being broken up.

#### CHEMICAL TRACK FORMATION MECHANISM AFTER ETCHING

The purpose of the following section is to test the above mentioned hypothesis, drawn from observations of conductimetric properties of samples exposed to different irradiation doses and then etched.

First, in order to prove the existence of oxydative degradation reaction, we changed etching condition. For cellulose nitrate we used a sodium hydroxyde solution in reducing condition (saturated by sodium nitrate). As a result (fig. n° 4) a general conductivity decrease was observed for each irradiation dose. This fact could be explained because hydroxyde sodium diffusion was more difficult, oxydative degradation reaction would not be so important, so film thickness is not so

reduced. The inverse phenomenon that is to say general conductivity increase was observed for an nitrate sodium oxydative bath.

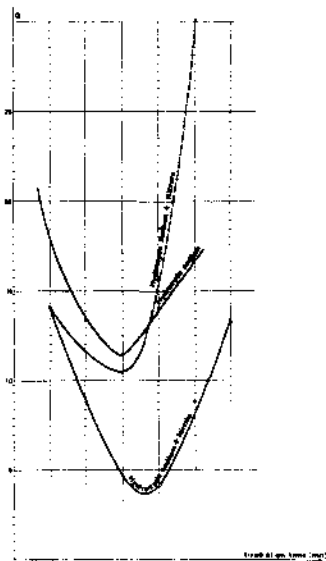


fig. 4

On the other hand, we tried to point out the important enhancement of saponification reaction with the irradiation dose and for a constant etching time. Two powerful tools, i.e. NMR and IR spectroscopies were used for such investigations.

First IR spectra of cellulose acetate are shown on fig. n° 5. It can be seen that the peak corresponding to OH groups ( $3500\text{ cm}^{-1}$ ) is clearly increased.

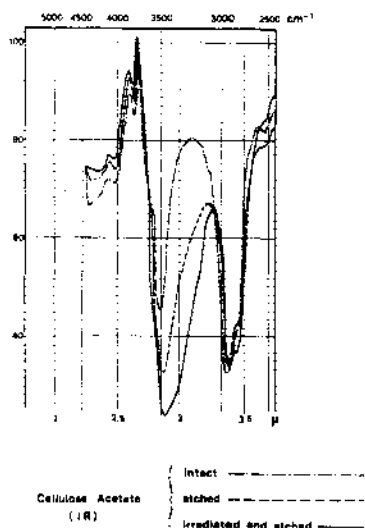


fig. 5

A similar behaviour is observed for cellulose nitrate. In the case of Makrofol fig. n° 6) this peak is very small and practically does not increase with etching and irradiation. Methyl groups and carbonates groups absorb at about  $3000\text{ cm}^{-1}$  and  $1800\text{ cm}^{-1}$  respectively. The absence of OH groups is related first with the chemical formula of this polymer and secondly with the fact that a chain breaking is necessary for this reaction.

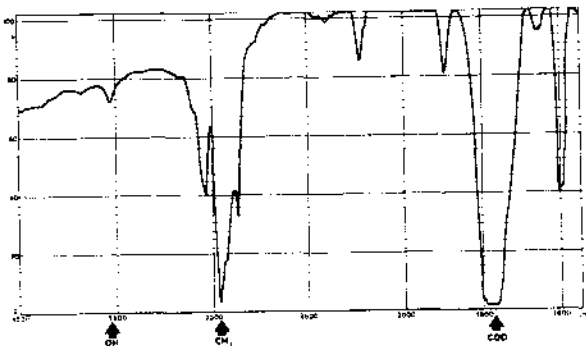


fig. 6

NMR studies confirmed the above observations : unfortunately, solubility problems appeared in the case of cellulose acetate samples. If intact films are soluble in chloroform and methylene chloride (NMR study of this polymer has been done by Gagnaire and Vincendon (5, 6) etched especially when irradiated films are only partly soluble in any solvent. This is an additional argument to support the hypothesis of the important chemical change due to irradiation and etching. One could suggest the following explanation : the result of the saponification is, in fact, to decrease the degree of acetylation. This reaction is only effective in irradiation damaged zones; consequently polymer solubility is affected.

Cellulose nitrate, probably due to the oxydation degradation process, is always soluble in acetone. NMR spectra of various cellulose nitrate films in deuterated acetone were recorded with a Varian HA 100 Spectrometer. On fig. 7, 8, 9, we can see spectra of intact film, of 35 minutes etched with 6,5N sodium hydroxyde film and 35 minutes etched film after irradiation. The

peak corresponding to oxydrile groups appears at  $\delta = 3$  ppm (from TMS internal Standard) it must be noticed that any water trace would give a contribution to this peak. However it is possible to compare different film spectra because all the samples were dried in the same conditions. The area of this peak is slightly increased when the polymer film is etched but a considerable area enhancement is observed when irradiated and etched. At high field, it can be observed camphre peaks (plasticizer). Broad absorption lines in the range 3,5 to 6 p.p.m are connected with the seven cyclic protons.

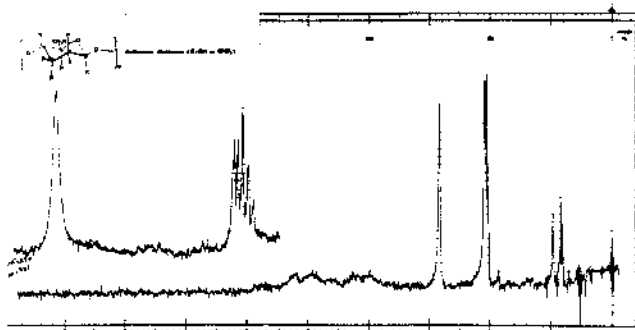


fig. 7

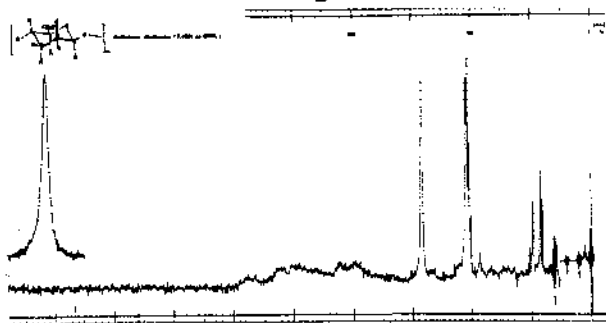


fig. 8

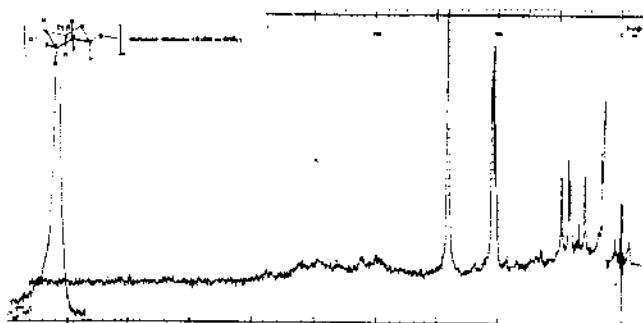


fig. 9

NMR Spectra of Makrofol films in deuterated chloroform were recorded with a 250 MHz Spectrometer from Cameca (Thomp-

son C.S.F.) This high field Spectrometer was needed in order to detect any slight variation that could have occurred. On fig. n° 10, a Makrofol film spectrum is shown. A typical para substituted aromatic signal is founded at about 7 p.p.m (8 phenyl protons). The methyl groups (6 protons) appear at  $\delta = 1,35$  p.p.m. A small peak at  $\delta = 1,25$  p.p.m is attributed to hydroxyl groups (probably the ends of macromolecular chains). This observation fit with the conclusions drawn from IR Spectra. The irradiation and etching effect is not clear because it is difficult to evaluate the hydroxyl peak importance at  $\delta = 1,25$  ppm which is closed by methyl group peak. Nevertheless, in order to obtain a better information we used TFA (trifluoroacetic acid) to shift out the OH group. We calculated the ratio of peak areas at 7 p.p.m and 1,3 p.p.m with and without TFA for each film (intact, etched, and irradiated etched). For instance, for the intact film the above ratio is 1,45 before TFA addition and 1,45 after.

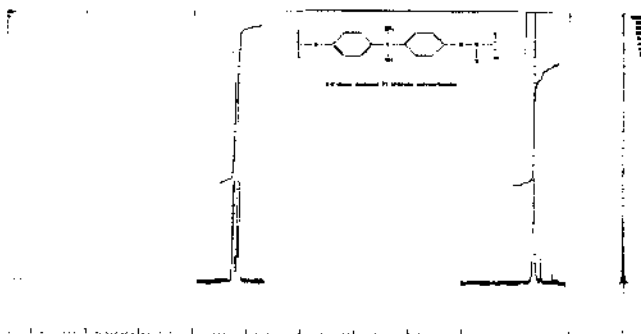


fig. 10

In the case of etched sample as irradiated etched samples the ratio is equal to 1,25 before TFA addition and 1,45 after TFA addition (fig. 11 and 12) (it can be observed on fig. 12 the shifted OH TFA peak at about  $\delta = 10$  p.p.m) So it appears there is a weak augmentation of hydroxyl peak area for an etched sample compared with an intact film ; increase is no more important in the case of an irradiated etched sample. That is in good agreement with IR Spectra for Makrofol. In fact, it would seem likely that reagent diffusion is allowed even in non irradiated sample by flexibility of molecules, so that irradiation would not matter a good deal for saponification reaction.

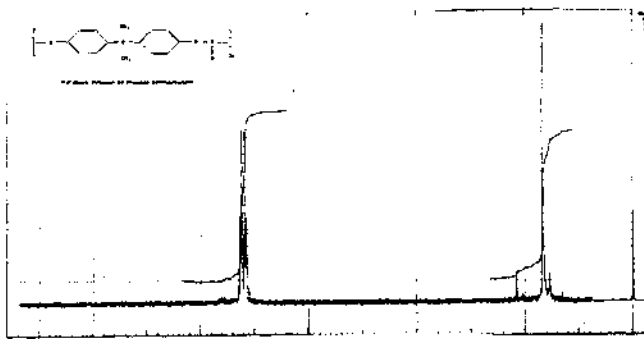


fig. 11

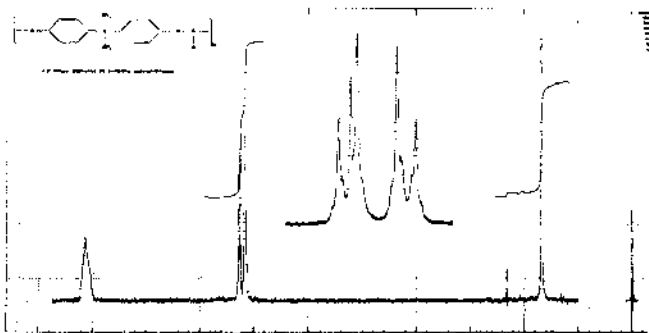


fig. 12

In conclusion, we have noticed correlation between conductimetric properties and saponification reaction for all studied detectors. That reaction is easier for cellulosic esters; in the case of polycarbonate, it is more difficult.

At last it can be noted that the sensitiveness of these detectors, characterized by the critical rate of energy loss  $(\frac{dE}{dx})_c$ , for the lightest recorded particle, rate which is 0,86 and 1,41 for nitrate and acetate (in the case of  $\alpha$  particles) and which is 5,5 for polycarbonate (in the case of  $^{10}B$ ), was also correlated.

Thus we have been able to approach chemical track etching process by the mean of conductivity study i.e. by the mean of physical property study.

#### ACKNOWLEDGEMENT

This work was carried out with the technical assistance of A. DELL'AMICO.

We are grateful to Pr GAGNAIRE and his collaborators who gave us the opportunity to perform NMR experiments.

#### REFERENCES

- (1) R.L. Fleisher, P.B. Price and R.M Walker, J. Appl. Phys., 36, 3645, 1965.
- (2) M. Bourdeaux, F. Roux, P.J. Bernard and C. Briand, Radioprotection, 1972, vol 7, n°3 666-777.
- (3) M. Bourdeaux, F. Roux, J.P. Bisset and C. Briand, Inst. Journal of Appl. Rad. and isotopes (sous presse)
- (4) G. Champetier, Dérivés cellulose Dunod 1954.
- (5) D. Gagnaire and M. Vincendon, Bull. Soc. Chim. 1968 fascicule 8 N° 527, 3413-3415.
- (6) M. Vincendon Thèse es Sciences Physique Grenoble.

## AUTHOR INDEX

- M. S. Abbas, 544, 1150  
 S. Abe, 491  
 J. L. Alexander, 780  
 A. Allisy, 232  
 H. Anderson, 999, 1005  
 I. Aodreuta, 196  
 S. A. Andronov, 610  
 G. M. Angleton, 184  
 Y. Argaman, 356  
 G. V. Arkhangel'skaya, 1358  
 M. F. Audet, 1096  
 M. Awschalom, 1464  
 J. Baarli, 841  
 V. I. Bad'in, 610, 1203  
 V. Balasubrahmanyam, 550  
 M. I. Balonov, 1422  
 J. Barbier, 370  
 R. M. Barkhudarov, 254  
 C. J. Barton, 1060  
 Z. G. Batova, 1203  
 L. Battist, 291, 350  
 P. Beau, 411  
 F. Beaufile, 637  
 K. Becker, 584  
 A. S. Belitskiy, 260  
 V. A. Belyayev, 1022  
 G. Benezech, 370  
 G. Benincasa, 578  
 B. G. Bennett, 1047  
 J. S. Benson, 1096  
 O. Berg, 1121  
 S. R. Bernard, 1400  
 L. Bertron, 411  
 G. F. Besseghini, 1191  
 A. M. Bhagwat, 852  
 D. P. Bhandi, 1226  
 S. Bhati, 683  
 V. Bhushan, 769  
 A. Bianco, 1214  
 R. I. Bikkulov, 85  
 M. Blum, 1169  
 D. A. Bodrova, 830  
 B. B. Boecker, 208, 1243  
 M. Bonleu, 637  
 J. A. Bonnell, 1330  
 V. V. Borisova, 147  
 N. M. Borovikova, 136  
 L. Botter-Jensen, 572  
 M. Bourdeaux, 1470  
 H. A. Boyd, 1237  
 L. Bozoky, 677  
 F. J. Bradley, 79  
 L. Bramati, 1072  
 B. M. Branson, 1180, 1185  
 F. Breuer, 736  
 L. Breuil, 190  
 C. Briand, 1470  
 I. E. Bronstein, 85  
 R. Brown, 999, 1005, 1434  
 H. Brunner, 835, 1287, 1318  
 L. A. Buidakov, 167  
 C. D. Burgess, 749  
 B. Burgkhardt, 394, 1440  
 B. M. Burnett, 1103  
 L. N. Burykina, 178  
 D. Bush, 1347  
 F. Busscher, 563  
 B. Cadet, 370  
 P. Cagnetti, 344  
 A. Cajkova, 538  
 R. D. Caldwell, 448  
 W. M. Carey, 291, 350  
 R. L. Carpenter, 76  
 P. B. Carter, 184  
 P. N. Casbolt, 556  
 D. L. Catt, 719  
 A. Causse, 1418  
 L. Ceravolo, 578  
 K. H. Chadwick, 457  
 R. H. Chamberlain, 791  
 R. Chandra, 1169  
 R. A. Chelysheva, 260  
 A. B. Chilton, 694  
 Ye. D. Chistov, 814  
 P. Christensen, 572  
 A. Ciubotaru-Bordeianu, 201  
 N. Cohen, 279  
 M. Collonge, 637  
 C. L. Comar, 28  
 C. S. Cook, 528  
 L. M. Cook, 272  
 W. A. Cornelius, 112, 783  
 L. Coulson, 1464  
 C. L. Crabtree, 1109  
 D. K. Craig, 719  
 J. F. Crow, 37  
 R. G. Cuddihy, 1243  
 L. N. Curykina, 178  
 G. C. Dale, 1330  
 C. A. de la Rosa, 1066  
 R. M. de Nulman, 1066  
 G. de Tovar, 637  
 N. Degani, 1011  
 R. P. Delahaye, 889  
 A. Delle Site, 532  
 M. Delpla, 1054  
 Ye. A. Denisova, 475  
 L. A. Dewerd, 100  
 A. Dhar, 100  
 C. Diaconescu, 1144  
 A. I. Dobroskok, 1358  
 E. I. Dolguirev, 957, 969, 1422  
 G. W. Dolphin, 760  
 A. Donagi, 356  
 S. K. Dua, 1226  
 R. Ducouso, 1418  
 F. Duprey, 190  
 A. Durakovic, 743  
 P. W. Durbin, 1412  
 P. Durney, 889  
 K. P. Eappen, 945  
 C. F. Eason, 1307  
 F. N. Eichner, 568  
 I. Elöd, 481  
 G. W. R. Endres, 568  
 A. P. Ermilov, 610  
 D. R. E. Ernborg, 1133  
 P. C. Escott, 760  
 R. A. Facey, 688  
 L. Fajita, 469  
 A. Favino, 1191  
 A. V. Fedorova, 147  
 I. Feher, 677  
 Y. Feige, 356, 1396  
 E. S. Ferri, 76



L. G. Filippova, 167  
 M. R. Ford, 619  
 H. Francois, 889  
 Y. Francois, 912  
 F. Fry, 985  
 M. Fujita, 1384  
 H. Fukuda, 1390  
 V. V. Gamper, 485  
 S. P. Garg, 979  
 O. Geszti, 481  
 F. R. Gibb, 1214  
 S. J. Gibbs, 1109  
 E. P. Goldfinch, 364  
 A. Goldin, 376  
 M. I. Goldman, 214  
 G. Gomez-Crespo, 1133  
 J. Gordon, 991  
 R. J. Grant, 1180, 1185  
 I. A. Gribova, 475  
 W. C. Griffith, 1243  
 P. Groer, 950  
 V. A. Grozovskaya, 129  
 W. E. Grummitt, 857  
 G. A. Gulyakov, 1022  
 A. K. Gus'kova, 475  
 N. G. Gusev, 1022  
 G. J. Hagan, 76  
 F. F. Hahn, 208  
 B. Haider, 920  
 L. M. Hairr, 279  
 T. E. Hakonson, 242, 248  
 K. E. Halnan, 760  
 R. N. Hamm, 707  
 D. E. Hankins, 515  
 C. M. Hardin, 1078  
 S. J. Harris, 654  
 N. T. Harrison, 760  
 T. Hattori, 1384, 1390  
 F. F. Haywood, 584  
 E. R. Heinz, 1127  
 L. H. Hempelmann, 713  
 K. R. Hetzel, 1197  
 P. J. Hewitt, 1249  
 J. E. Hickey, 1103  
 G. S. Hill, 631  
 M. J. Hilyer, 631  
 C. H. Hobbs, 208  
 M. Hofert, 841  
 J. G. Hollins, 743  
**R. B. Holtzman, 1406**  
**R. G. Hoops, 1185**  
 S. A. Horne, 882  
 F. Hosain, 1175  
 P. Hosain, 1175  
 M. Houskova, 538  
 C. Y. Huang, 1091  
 R. K. Hukkoo, 683  
 A. P. Hull, 1035  
 M. S. Ibatullin, 266, 1353  
 R. D. Ice, 1197  
 H. Iida, 91, 1452  
 F. H. Ilcewicz, 1406  
 L. A. Ilyin, 1371  
 K. Imai, 382  
 L. Imbert, 637  
 Y. Izumi, 1384  
 W. Jacobi, 920  
 A. C. James, 932  
 H. Jammet, 1261  
 S. Janardhanan, 846  
 J. Jankowski, 506  
 N. Jeung, 1412  
 G. Jha, 945  
 S. M. Jinks, 279  
 H. Joffre, 637  
 L. J. Johnson, 243, 248  
 O. N. Johnson, 1109  
 R. K. Jones, 208  
 P. P. Joshi, 1226  
 H. W. Julius, 563  
 H. Kaiser, 889  
 G. N. Kajdanovsky, 957, 969  
 V. S. Kalistratova, 172  
 G. M. Kanapilly, 1237  
 Yu. T. Kapitanov, 926, 1208  
 D. K. Kapur, 979  
 A. S. Karasik, 1422  
 I. S. Katsapov, 172  
 G. N. Katsnel'son, 485  
 S. V. Kaye, 1060  
 D. J. Keefe, 950  
 W. E. Keene, 291, 350  
 J. G. Kereiakes, 773  
 A. H. Khan, 939  
 A. V. Khortsev, 648  
 S. A. Khukhlayeva, 926  
 H. Kiefer, 1337  
 G. I. Kirsanova, 475  
 P. Kissel, 864  
 Y. Klopfer, 671  
 V. A. Knizhnikov, 129  
 W. Koelzer, 1337  
 J. A. Koen, 1085  
 V. A. Kolotvin, 141  
 L. A. Konig, 400  
 A. L. Kononovich, 824  
 V. V. Kopayev, 810  
 N. F. Korelina, 1358  
 L. N. Korzhov, 810  
 T. Koshijima, 91, 1452  
 P. Kotrappa, 1226, 1232  
 L. Kramer, 1406  
 G. P. Krasnoshchekova, 1358  
 T. N. Krishnamurthi, 846  
 E. M. Krisyuk, 870  
 B. Krych, 506  
 E. Kunz, 538  
 Y. Kurokawa, 491  
 G. A. Kuznetsova, 1022  
 E. N. L'vovskaya, 475  
 J. R. A. Lakey, 1298  
 I. Lancranjan, 501  
 W. H. Langham, 713  
 R. E. Lapp, 14  
 A. V. Larichev, 814  
 L. N. Lavrent'ev, 147  
 W. Leach, 495  
 O. V. Lebedev, 963  
 H. P. Leenhouts, 457  
 I. P. Leggett, Jr., 1078  
 I. S. Leith, 106, 112, 783  
 E. G. Letourneau, 991  
 A. N. Liberman, 85  
 Yu. P. Likhachev, 926  
 I. A. Likhtarev, 1358  
 B. Lindell, 591  
 G. Lindel, 429  
 E. Lindholmer, 1121  
 J. Liniacki, 506  
 E. P. Lisachenko, 870  
 B. Ya. Litver, 266, 1353  
 E. G. Litvinova, 648  
 R. Ye. Livshits, 141  
 R. D. Lloyd, 417  
 N. V. Loseva, 485  
 I. Lovanyi, 481  
 J. O. Lubenau, 1078  
 E. Lubin, 1396  
 E. N. L'vovskaya, 475  
 P. P. Lyarskiy, 830, 926, 976, 1208  
 G. Madelaine, 912  
 K. Maeda, 491  
 B. Majborn, 572  
 L. G. Makeyeva, 1208  
 V. M. Malykhin, 963  
 Y. Mamane, 356  
 V. A. Manchuk, 1446  
 A. N. Marey, 254, 1022  
 M. E. Marks, 1096  
 C. H. Marshall, 1169  
 J. H. Marshall, 417  
 T. O. Marshall, 556  
 J. J. Martin, 411  
 Y. Martin-Borret, 1470  
 T. Marzullo, 1072  
 J. J. Mauro, 285  
 C. W. Mays, 417  
 R. O. McClellan, 208

D. McCannon, 799  
 W. P. McDowell, 950  
 R. C. McMillan, 882  
 N. N. Mel'nikova, 178  
 H. E. Meyer, 1434  
 S. M. Michaelson, 51  
 A. I. Milovidova, 153  
 C. G. Milu, 1144  
 Y. Minagawa, 491  
 J. Miribel, 370  
 N. T. Mitchell, 301, 319  
 I. Miyanaga, 382  
 S. Mizushita, 1390  
 G. Modoran, 1139, 1144  
 A. A. Moiseyev, 266  
 P. I. Moiseyev, 830  
 R. E. Moore, 1060  
 K. Z. Morgan, 589  
 S. Moriuchi, 382  
 F. Morley, 313  
 P. E. Morrow, 1214  
 Yu. I. Moskatov, 117, 153, 167  
 I. Ye. Mukhin, 136  
 M. Murata, 388  
 R. Nader, 8  
 A. Nagaratnam, 1163  
 E. Nagel, 308  
 L. I. Nagovitsyna, 136  
 K. K. Nagpal, 1156  
 A. Naharin, 1396  
 L. Namestrek, 538  
 M. Naritomi, 388  
 M. A. Nevstruyeva, 141  
 L. L. Nichols, 568  
 A. P. Nifatov, 167  
 A. I. Nizhnikov, 266, 1353  
 N. Ya. Novikova, 254  
 B. L. O'Neal, 700  
 R. J. Olson, 719  
 H. C. Orchard, 1458  
 Ye. I. Orlova, 260  
 R. L. Orlyanskaya, 178  
 V. V. Orobei, 85  
 R. V. Osborne, 1428  
 E. E. Oscarson, 643  
 M. Pagliari, 344  
 Yu. D. Parfenov, 1364  
 J. F. Park, 719  
 G. M. Parkhomenko, 1203  
 M. W. Parrott, 1412  
 C. Pasquier, 1418  
 F. H. Passant, 326  
 E. D. Pavlitskaya, 147  
 N. A. Pavlovskaya, 1208  
 G. Pepin, 190  
 V. Ye. Pestrikov, 963  
 I. K. Petrovich, 117  
 A. A. Petushkov, 976, 1446  
 E. Piesch, 394, 1440  
 P. Plavosin, 1144  
 C. Pomroy, 991  
 V. L. Ponomareva, 178  
 H. I. Popescu, 501  
 N. V. Porozov, 957, 969  
 G. Portal, 889  
 J. W. Poston, 1115  
 J. Pradel, 912  
 T. Predmerszky, 481  
 M. Price, 1  
 O. N. Prokofiev, 1422  
 R. J. Purrott, 760  
 W. D. Purtynum, 248  
 O. G. Raabe, 1237  
 N. T. Racoveanu, 196, 1139, 1144  
 M. Raghavayya, 939  
 B. Raghunath, 1232  
 P. V. Ramzayev, 266, 1353  
 A. R. Reddy, 1156, 1163  
 C. Renard, 864  
 V. S. Repin, 1358  
 C. R. Richmond, 713  
 E. Righi, 736  
 E. Riklis, 1011  
 P. J. Robbins, 1180  
 S. N. Roberto, 79  
 C. D. Robinette, 184  
 B. Robinson, 999, 1005, 1434  
 P. S. Rohwer, 1060  
 I. Rosa, 1072  
 H. H. Rossi, 229  
 F. Roux, 1470  
 V. P. Rublevskiy, 296  
 E. I. Rudnitskaya, 167  
 R. Rugh, 495  
 J. Rundo, 731  
 T. Sadeh, 1396  
 E. L. Saenger, 773  
 L. A. Sagan, 429  
 N. Y. St. Denis, 1307  
 M. S. Sakovskaia, 85  
 G. Santori, 532  
 V. A. Sarychev, 810  
 D. Sashin, 1127  
 S. Ya. Satarina, 926  
 G. Scarpa, 578  
 R. Schaeffer, 659  
 T. Schlesinger, 671  
 J. Schwammberger, 835  
 L. Seabron, 1078  
 J. Sedlet, 731  
 V. K. Sel'tser, 485  
 R. F. Selman, 950  
 A. I. Semenov, 117  
 N. I. Shalak, 870  
 V. P. Shamov, 870, 957, 969  
 R. C. Sharma, 979  
 J. W. Shaver, 1078  
 K. B. Shaw, 556  
 W. E. Sheehan, 1434  
 V. I. Shetesnova, 153  
 V. M. Shubik, 85, 141  
 V. L. Shvedov, 159  
 P. Sigli, 370  
 P. Simon, 889  
 V. A. Smirennaya, 260  
 L. N. Smirenniy, 648  
 V. H. Smith, 719, 725  
 J. O. Snihs, 900  
 M. B. Snipes, 208  
 W. S. Snyder, 603, 619, 625  
 Yu. Ja. Sokolov, 1377  
 S. D. Soman, 846, 852, 1232  
 S. Somasundaram, 683, 979  
 H. Spencer, 1406  
 F. W. Spiers, 439  
 R. Spiess, 1287  
 G. Staeblein, 1337  
 V. Stanculescu, 201  
 J. R. Stanton, 1103  
 A. I. Stepanov, 485  
 E. J. Sternglass, 1127  
 T. G. Stoebe, 100  
 V. P. Stolyarov, 1377  
 M. C. Storr, 743  
 V. N. Strel'tsova, 117  
 J. C. Strong, 932  
 M. C. Subba Ramu, 1220  
 M. J. Suess, 64  
 S. Suma, 1139  
 T. Surendran, 979  
 L. Szegedi, 550  
 Y. Takahashi, 1390  
 S. I. Tarasov, 870  
 A. P. Tchesnokova, 85  
 L. A. Teplykh, 266  
 A. V. Terman, 814  
 C. Testa, 532  
 T. D. Tham, 584  
 J. F. Thiel, 96  
 G. E. Thoma, 773  
 R. G. Thomas, 1255  
 I. M. G. Thompson, 1458  
 F. K. Tomlinson, 999, 1005  
 K. L. Travis, 1103  
 H. C. Tresise, 338  
 R. Trial, 637  
 V. I. Trifonov, 117, 172  
 H. A. Trindade, 376

M. N. Troitskaya, 266, 1353  
A. D. Turkin, 296, 1377  
J. E. Turner, 707  
G. Tyler, 985  
D. Underhill, 376  
K. Unnikrishnan, 683  
A. C. Upton, 43  
A. P. Ushakova, 135B  
G. Valle, 190  
L. A. Vasil'yeva, 178  
C. Vasilescu-Fulea, 1144  
C. W. Verhoef, 563  
N. V. Vershinin, 830  
K. A. Veselovskaya, 178  
G. L. Voelz, 713  
V. V. Volkovitskaya, 830  
L. F. Wailly, 184  
D. A. Waite, 406  
N. Wald, 773

G. G. Warner, 631, 1115  
M. M. Wasson, 319  
E. G. Weatherley, 749  
G. A. M. Webb, 313  
J. Weber, 1085  
P. S. Weng, 1091  
B. M. Wheatley, 332  
G. H. Whipple, 1017  
M. H. Williams, 1412  
M. Winter, 394, 400  
M. E. Wrenn, 279, 285  
H. A. Wright, 707  
M. C. Wukasch, 96, 272  
H. O. Wyckoff, 225  
J. M. Wyckoff, 694  
A. Yabe, 1384, 1390  
N. G. Yakovieva, 147  
H. Yamagishi, 1390

M. S. Yegorova, 810  
L. T. Yelovskaya, 926, 1208  
A. V. Yorke, 1298  
Y. Yoshida, 388  
G. A. Zalikin, 117  
N. A. Zapol'skaya, 147  
G. Zara, 1072  
O. M. Zaraeyev, 814  
A. L. Zel'din, 1353  
M. R. Zel'tser, 976  
J. C. Zerbib, 637  
P. Zettwoog, 912  
Yu. A. Zhakov, 260, 1022  
Sh. Zhanadilov, 178  
L. Ya. Zhorno, 147  
Ye. S. Zhorova, 117  
I. A. Zvonova, 1358  
A. S. Zykova, 296, 1022

