INHALATION CASES OF ENRICHED INSOLUBLE URANIUM OXIDES*

NEIL B. SCHULTZ
Union Carbide Corporation,
Nuclear Division,
Oak Ridge Gaseous Diffusion Plant
Oak Ridge, Tennessee

Abstract—The retention and excretion of uranium by about 80 employees routinely assigned to calcining and fluorinating uranium-bearing materials enriched in the ^{235}U isotope have been closely followed for more than a year. *In vivo* measurements of body radioactivity were made by means of gamma spectrometry with two NaI scintillation detectors in an "iron-room" type, low background counter. Initial chest cavity burdens ranged from less than 0.006 μCi to about 0.050 μCi of enriched uranium, with only two cases having annual averages greater than 0.017 μCi. The effective half-lives for chest cavity retention of the three employees who were temporarily removed from uranium processing areas ranged from about 120 to 250 days. Urine samples collected over 24 hr were analyzed for uranium mass, alpha activity, and ^{235}U enrichment. Average ratios of chest cavity burden to urine excretion rate increased from about 550 to 1100 between the initial and final determinations. Fecal excretion rates averaged 40% to 180% of the uranium urinary excretion rates. Material balance calculations showed agreement ranging from 50% to 75% between total urinary–fecal excretion and reduction in chest cavity burden as shown by *in vivo* measurements. Pulmonary function tests of the employees revealed normal respiratory performance. Medical data, including chest X-rays, clinical urinalysis for albumin, and microscopic examination of urine for pathological cells and organisms, are normal in all cases. There is no evidence of personnel injury from these transient internal uranium depositions.

INTRODUCTION

During more than 20 years of operation of a uranium gaseous diffusion plant, appropriate environmental monitoring and operating controls have been maintained to limit exposure of employees to acceptable levels. For the past 5 years, *in vivo* gamma spectrometry measurements of the 186 keV gamma radiation associated with ^{235}U have been performed to supplement the urinalysis program for monitoring personnel involved in uranium processing. Over 90% of the personnel monitoring data reflect either less-than-detectable or negligible results, and present no problem in evaluating exposures. The retention and excretion of uranium by about 80 male employees routinely assigned to calcining and fluorinating uranium-bearing materials enriched in the ^{235}U isotope have been closely followed for the period of November 1964 through June 1966. The purpose of this paper is to record the results of this study, with data for 3 employees reported in detail.

SUMMARY

The operation of industrial-scale uranium recovery facilities at the Oak Ridge Gaseous Diffusion Plant by the Union Carbide Corporation, Nuclear Division, for the U.S. Atomic Energy Commission involves the calcining and fluorinating of uranium-bearing materials enriched in the ^{235}U and ^{234}U isotopes. Environmental air sampling and work surface

* This document is based on work performed at the Oak Ridge Gaseous Diffusion Plant, operated by Union Carbide Corporation, Nuclear Division, for the U.S. Atomic Energy Commission.

radio-activity surveys are conducted on a routine basis in all processing areas to evaluate internal exposure potential via inhalation. Personnel monitoring practices include urine analysis, fecal analysis, and *in vivo* gamma spectrum analysis programs. Urine and fecal samples, either single voiding or excreta collected over 24 hr, are analyzed for uranium mass alpha activity, and ^{235}U enrichment. *In vivo* measurements of body radioactivity were made by means of gamma spectrometry with 2 NaI scintillation detectors in an "iron-room" type, low background counter. The retention and excretion of uranium by about 80 male chemical operators and maintenance mechanics routinely assigned to calcining and fluorinating uranium-bearing materials enriched in the ^{235}U isotope have been studied during the period of November 1964 through June 1966. Only 8 detectable chest cavity burdens were recorded; of these, 3 had an annual average value less than 0.008 μCi of uranium; 3 averaged between 0.008 μCi and 0.017 μCi, and 2 averaged 0.028 μCi of uranium. The subjects of this report are the 2 latter cases and a third case in the 0.008 μCi to 0.017 μCi range, all of whom are male chemical operators and were temporarily transferred from uranium processing areas following the initial elevated urinalysis and *in vivo* measurements. The effective half-lives for chest cavity retention of these 3 employees ranged from about 120 to 250 days. Average ratios of chest cavity burden to urine excretion rates increased from about 550 to 1100 between the initial and final determinations. Fecal excretion rates averaged 40% to 180% of the uranium urinary excretion rates. Material balance calculations indicated that 50% to 75% of the reduction in chest cavity burden as shown by *in vivo* measurements are reflected in the total urinary-fecal excretion. Pulmonary function tests revealed normal respiratory performance, and medical data are within normal limits in all cases. There is no evidence of personnel injury from these transient internal uranium depositions.

BACKGROUND INFORMATION

Operations

Personnel in the study group were 80 employees assigned to the operations and maintenance activities in the enriched uranium recovery facility, involving the conversion of uranyl nitrate, $UO_2(NO_3)_2$, by calcining at about 750°C to urano-uranic oxide, U_3O_8, and the direct fluorination of the oxide to uranium hexafluoride, UF_6. These operations, conducted in closed systems, are located in relatively open areas of approximately 12,000 ft^2 and 3000 ft^2, respectively.

Environmental Monitoring

Established practices limit internal exposure to acceptable levels by appropriate environmental controls. To evaluate both the effectiveness of these controls and the internal exposure potential via inhalation, air sampling is conducted on a routine basis in all processing areas. In general, environmental air levels are well within the limits prescribed by the National Committee on Radiation Protection [1] and the International Commission on Radiological Protection. [2] Long-term or 8-hr shift-length samples are automatically collected, counted for alpha radioactivity concentration, and recorded by locally developed continuous air monitors. [3] During this study, 954 shift-length samples were collected, with an average concentration of 3.3×10^{-11} μCi/cc, and a maximum 8-hr value of 1.5×10^{-9} μCi/cc. Spot-air samples, taken over periods of 10 min or less in close proximity to equipment during maintenance or operation activities requiring momentary opening of the system to atmosphere, resulted in maximum air levels of 4.9×10^{-7} μCi/cc. Where the probability of relatively high air activity exists, the use of appropriate respiratory equipment is mandatory. Measurement of transferable uranium alpha activity on work surfaces provides a broad sampling base for estimating the potential personnel exposure. [4] Uranium alpha levels on floors and work surfaces averaged 1.1×10^{-5} μCi/cm^2, with a maximum value of only 4.5×10^{-5} μCi/cm^2, reflecting effective environmental control techniques.

Aerosol Characteristics

Evaluation of the potential hazard from inhaling radioactive airborne particulates requires not only monitoring of the concentration levels, but also careful consideration of the

physico-chemical characteristics of the particles, and the physiological characteristics of the human respiratory system. The two compounds of most likely exposure in these cases are urano-uranic oxide, U_3O_8, and uranium hexafluoride, UF_6. The oxides of uranium, generally listed as being insoluble in water, actually have widely varying solubilities (0–100%) in synthetic lung fluids,[5] with the U_3O_8 of interest in this study being about 16% soluble in test tube studies over a 4-month period. The uranium content of the powder is 84%, approximately 2/3 being hexavalent and 1/3 being tetravalent. This oxide powder has a surface area of 3.05 m^2/g and a density of 8.38 g/cc. A count median diameter of 1.0 $\pm$ 3.0 μ was determined from the photomicrograph shown in Fig. 1. The other potential exposure material, gaseous uranium hexafluoride, hydrolyzes if released to the atmosphere, with reaction products of hydrogen fluoride, HF, and uranyl fluoride, UO_2F_2. The particle size range of the highly soluble, short biological half-life UO_2F_2 aerosols varies widely from submicron to several microns in diameter, depending upon several factors, such as ambient temperature, relative humidity,

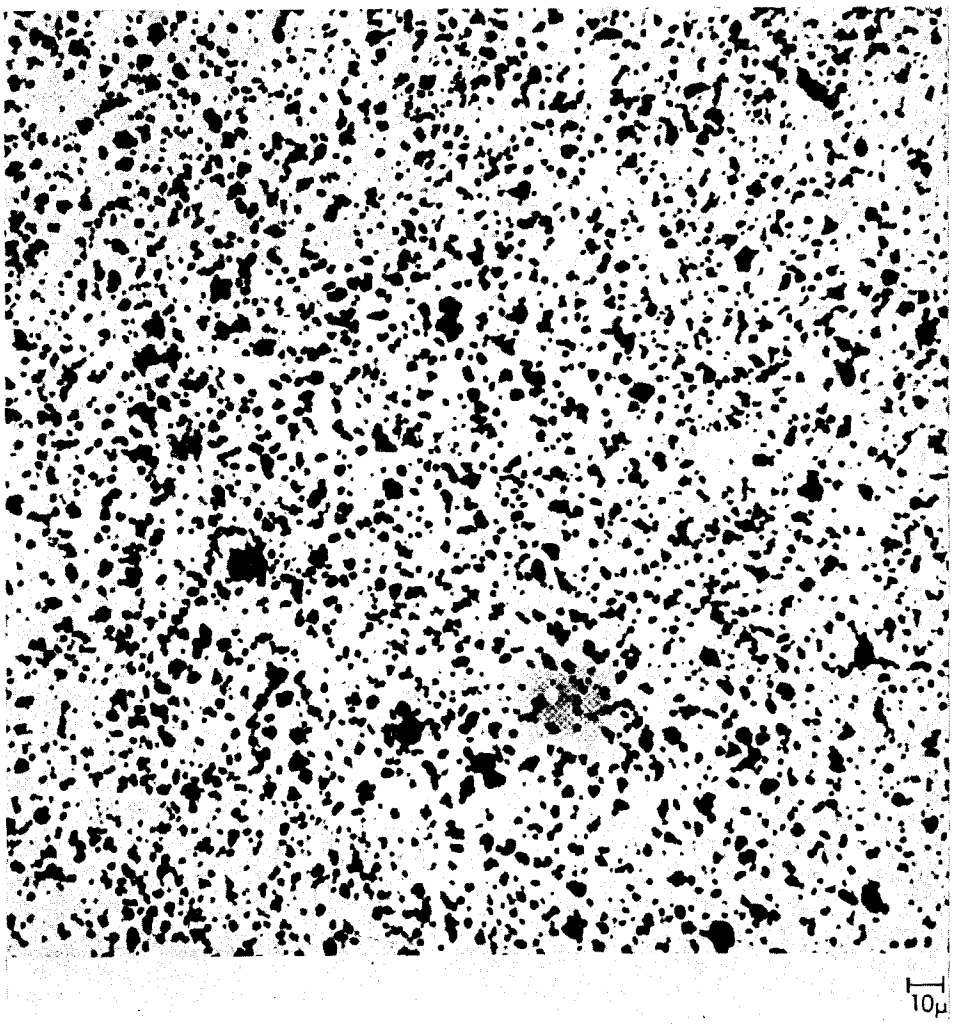

FIG. 1. Photomicrograph of U_3O_8 × 500 reduced to $\frac{8}{10}$.

and the concentration of the UF_6 at the time of the release.[4] During the study period, enrichments in the ^{235}U isotope ranged from about 10% to approximately 90%; for the 3 cases reported, the most likely enrichment of the exposure material was about 25% ^{235}U.

Personnel Monitoring

In order to evaluate the effectiveness of engineering design and operational practices, and to estimate the degree of internal exposure, personnel monitoring programs have been established. Monitoring of all personnel who work in uranium areas is accomplished by uranium urinalysis and *in vivo* gamma spectrum analysis. If either method points up potential exposure cases, fecal sampling as well as more frequent urinalysis and *in vivo* gamma spectrum analyses are used to more precisely evaluate the degree of exposure. The scheduled urinalysis sampling frequency is monthly for both the chemical operators and the maintenance mechanics, as well as the supervisory staff. These routine samples are obtained on a spot basis and are submitted after a 2- or 3-day work break, allowing time for the highly soluble uranium to be eliminated. When more frequent sampling is indicated, 24 hr urine and fecal voidings are submitted. In this study, when the employees were requested to provide such timed excretion samples, the importance was stressed to secure knowledgeable co-operation. Volumes of urine and masses of feces reported to be 24-hr samples averaged 1263 ml/day and 135 g/day, respectively, and are summarized in Table A5. Urine analyses are routinely made for both uranium mass and alpha activity, using the familiar laboratory procedures of (a) the fluorophotometric method for determining uranium mass, and (b) electroplating and counting for alpha activity. The ^{235}U enrichment is determined by surface ionization mass spectrometry.

The *in vivo* gamma spectrometry measurements of the 186 keV gamma radiation associated with ^{235}U reported in this study were made at the nearby Y-12 Plant, also operated by the Union Carbide Corporation, Nuclear Division, for the U.S. Atomic Energy Commission. Figure 2 is a photograph showing the positioning of the subject and the detectors.

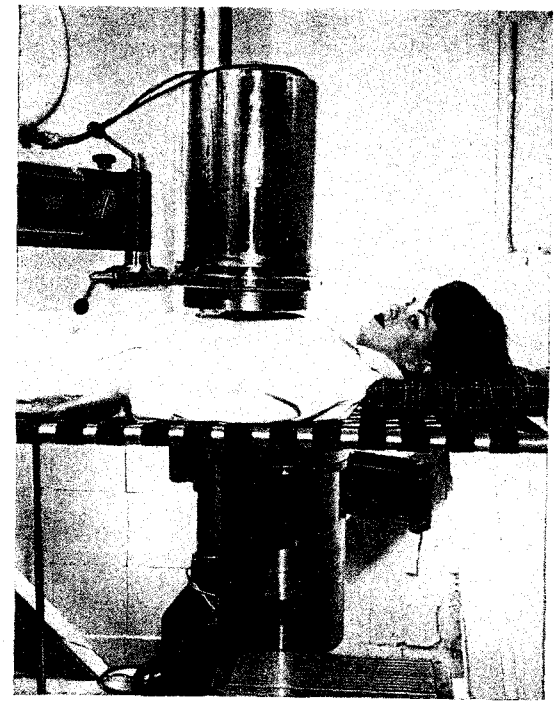

FIG. 2. *In vivo* gamma measurement—positioning of subject and detectors.

This gamma radioactivity measurement facility consists of the familiar low-background "iron-room" counting chamber, two 9-in. diameter by 4-in. thick thallium-activated sodium iodide crystals, and a 400-channel pulse height analyzer.[6] Quantities as low as 30% of the maximum permissible body burden of any of the isotopic enrichments investigated can be measured. For a wide sampling of employees representing the many crafts and skills working in a large gaseous diffusion plant, all chest cavity burdens, except as indicated in this report, averaged well below 0.008 μCi of uranium.

EVALUATION OF DATA

Medical Findings

Deposition, retention, and translocation of inhaled radioactive particles are related not only to their physical and chemical properties, but also to the physiologic and anatomic characteristics of the host. For example, cigarette smoke, a ciliostatic substance, might reduce the rate of mucus transport by over one-half,

and thus influence the respiratory system clearance, particularly during and shortly following the dust exposure.[7] Pulmonary function tests of the employees, two of whom are relatively heavy cigarette smokers, revealed normal respiratory performance. Medical data, including chest X-rays, clinical urinalysis for albumin, and microscopic examination of urine for pathological cells and organisms, are within normal limits in all cases.

In Vivo Gamma Measurements

Of the 80 employees considered in this study, 8 had detectable chest cavity burdens, 3 had an annual average value less than 0.008 μCi of uranium, 3 averaged between 0.008 μCi and 0.017 μCi of uranium, and 2 averaged 0.028 μCi. The subjects of this report are the 2 latter cases and a third case in the 0.008 μCi to 0.017 μCi range, all of whom are male chemical operators and were temporarily transferred from uranium processing areas following the initial elevated *in vivo* measurement. These data are detailed in Tables A1, A2, and A3 of the Appendix.

Excretion Data

The least square fit for excreta measurements on these employees predicted initial levels as high as 71 pCi/day in the urine and 593 pCi/day in the feces. The excretion pattern changes at a rate different from that of the chest burden, as shown in Figs. 3, 4, and 5.

Data Summary

For ease of interpretation, the tabulated data were fed to a computer program to determine the least square fit for the log of the measurement as a function of the number of days since the initial elevated *in vivo* measurement. The dates of the first elevated *in vivo* measurements are the time datum ($t = 0$) for the derived curves of uranium excretion and chest burden data for the 3 cases, shown, along with the exponential function-of-time formulae, in Figs. 3, 4,

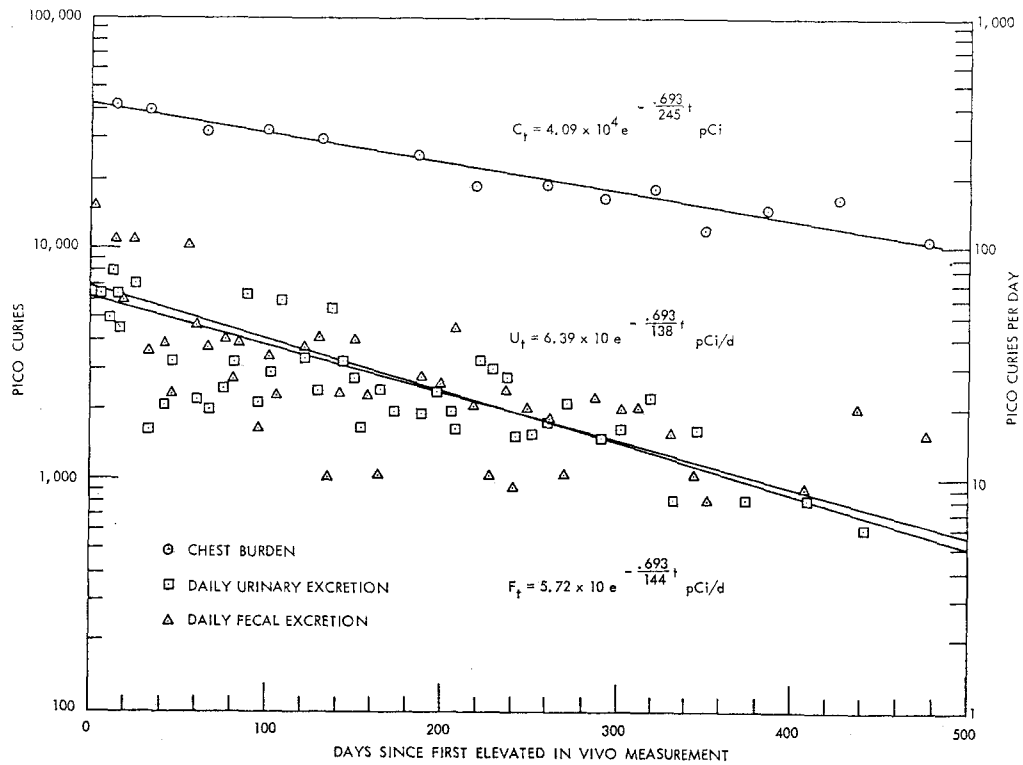

FIG. 3. Excretion and *in vivo* curves, case K-1.

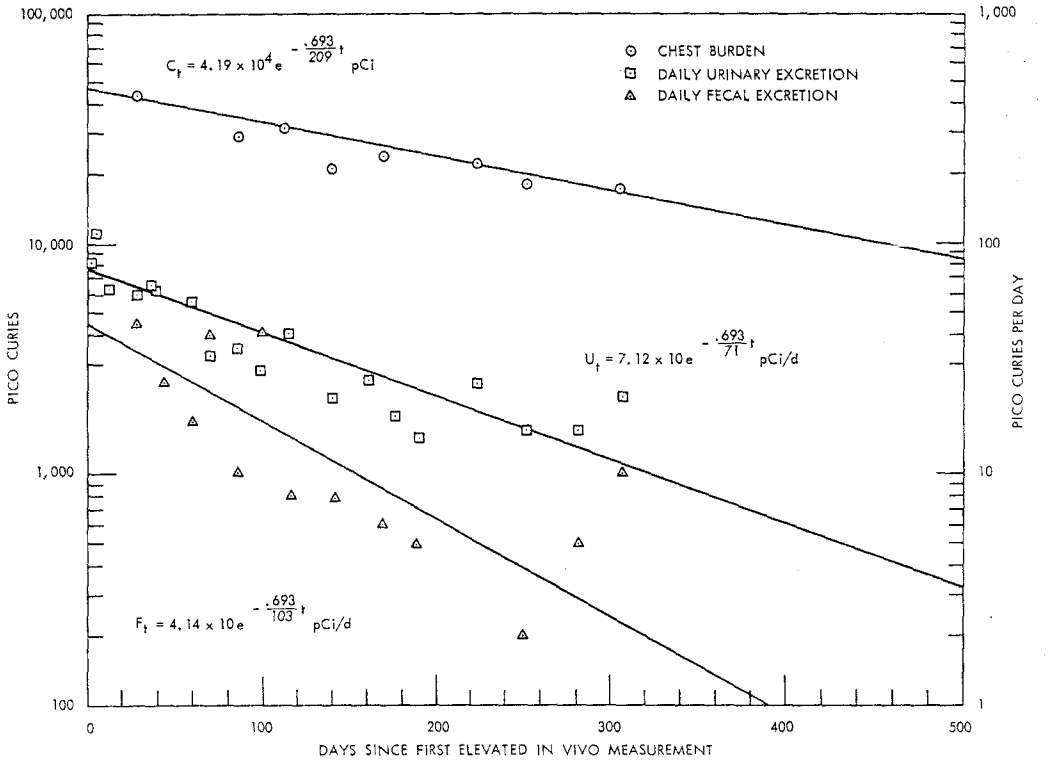

FIG. 4. Excretion and *in vivo* curves, case K-2.

and 5. Overall bioassay data averages for the 3 cases are summarized in Table 1. For the convenience of the reader, comparisons of excretion and *in vivo* data are listed for these and other cases [8–10] in Table A4.

The data suggest that the half-lives of insoluble or slightly soluble inhaled uranium in the chest cavity have long-lived components in these 3 cases ranging from about 120 to 250 days. The initial shorter half-life component of uranium excretion found in cases F-1[8] and UR-2[9] was seen in case K-3, but not in cases K-1 and K-2. Ratios of monthly averages of *in vivo* chest cavity burden and the quantity of uranium excreted daily in the urine varied from about 120 to 4130, averaging 1032, 904, and 357, respectively, for the 3 cases, suggesting the multiplication factor for estimating the quantity of uranium retained in the lung from the quantity of uranium excreted per day in the urine. For the 2 more similar cases, urinary excretion levels of 23 $\pm$ 17 pCi/day and 24 $\pm$ 15 pCi/day at 95% confidence levels would predict 0.017 μCi chest cavity burden of uranium. Fecal excretion was a significant mode of clearance in the 3 cases, averaging 92%, 44%, and 183%, respectively, of the urinary uranium elimination. These variances may be explained by such factors as differences in particle size, effective solubility, physiology of respiration, individual physical characteristics of the human anatomy governing dust deposition, and the elapsed time after exposure that fecal sampling started.

Material balances were calculated on the 3 cases for total excretion, and compared with the decrease shown by *in vivo* measurements. These data suggest that 50% to 75% of the chest burden decrease is accounted for through urinary and fecal excretion. A possible explanation is that a portion of the uranium retained in the lung is being transferred to another component,

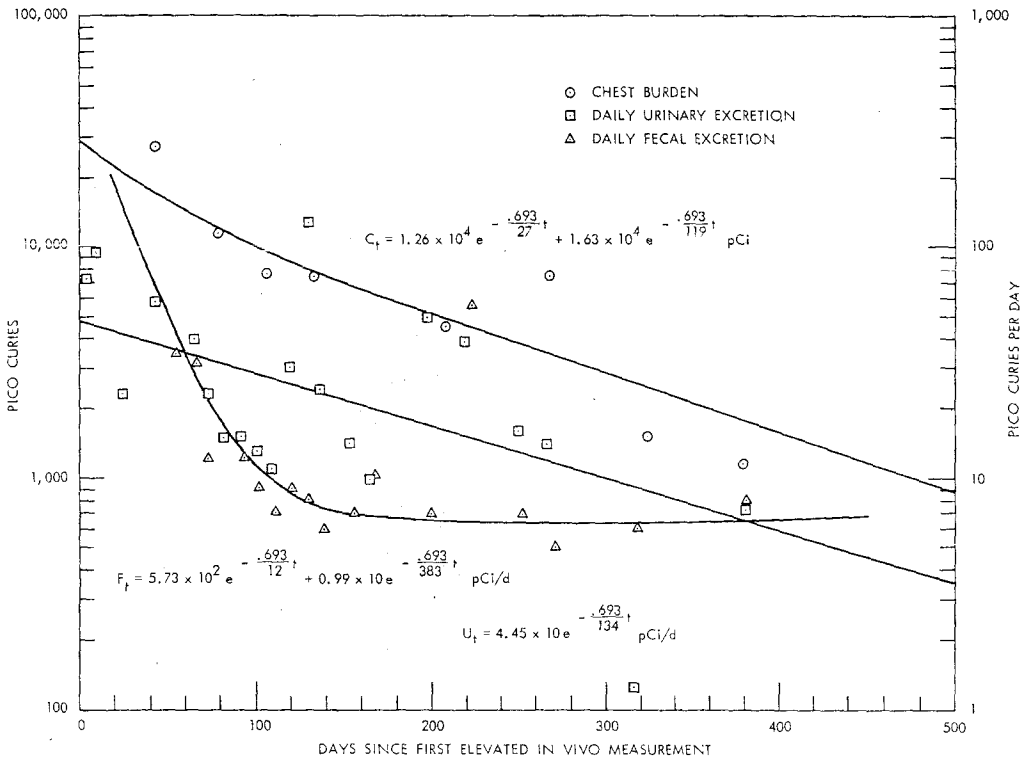

FIG. 5. Excretion and *in vivo* curves, case K-3.

Table 1. *Data Summary*

	Case K-1	Case K-2	Case K-3
Urinalysis			
Early excretion level (pCi/day)	64	71	45
Biological half-life (days)	138	103	134
Number of measurements	48	21	22
Fecal Analysis			
Early excretion level (pCi/day)	57	41	583
Biological half-life (days)	144	71	12,383
Number of measurements	43	17	18
Total Excretion			
Urine–fecal excretion (pCi)	22,000	13,000	20,000
In Vivo			
Early burden (pCi)	40,900	41,900	28,900
Biological half-life (days)	245	209	27,119
In vivo decrease (pCi)	29,000	26,000	29,000
Number of measurements	17	12	10
Ratio			
Fecal/urinary excretion	0.92	0.44	1.83
Total excretion/*in vivo* decrease	0.76	0.50	0.69

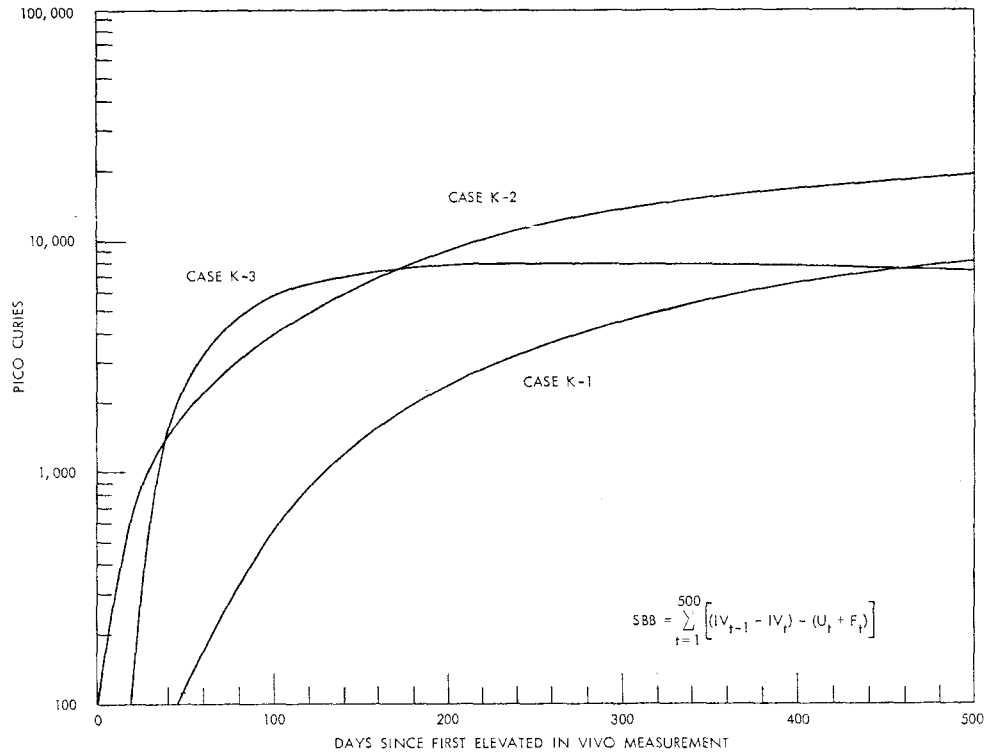

FIG. 6. Systemic burden build-up.

such as the bone or lymph nodes, where it is retained but is not detectable by *in vivo* gamma spectrometry. An estimate of the buildup of uranium in other body components was derived from a computer program which sums the differences between excretion and apparent elimination from the chest cavity. Plots of this buildup are shown in Fig. 6. As a point of reference, 50,000 pCi is the maximum permissible body burden (bone) for soluble, enriched uranium.

CONCLUSIONS

Three individuals with detectable quantities of enriched uranium in their chest cavities and excreting measurable quantities in their urine and feces were monitored for a period of over a year. The data are still being collected. For these cases, the following general conclusions are made:

1. Biological half-lives for insoluble uranium dust retained in the lung ranged from 120 to 250 days.
2. In the absence of current soluble uranium exposures, factors of about 1000 were reflected by the ratios of the chest burden and the daily urinary uranium excretion levels.
3. Fecal excretion is a significant mode of uranium elimination.
4. The apparent decrease of uranium in the chest is greater than the amount of uranium excreted, suggesting transfer of uranium from the chest cavity to another organ.
5. Systemic buildup for these lung burdens represents about 20 per cent of the maximum permissible body burden (bone) for soluble enriched uranium.
6. Urinalysis is of value in highlighting potential uranium exposure cases.
7. Urinalysis, fecal analysis, and *in vivo* gamma spectrometer measurements are

suggested for thorough evaluation of a significant uranium exposure.

8. Medical findings for these employees are within normal limits and there is no evidence of personnel injury from the transient internal uranium depositions.

ACKNOWLEDGEMENTS

The author gratefully acknowledges the valuable contributions of the following: the Y-12 Plant Radiation Safety Department, for providing the *in vivo* gamma radioactivity measurements; Dr. L. M. Scott, for his advice and assistance with data interpretation; ORGDP Production Division personnel for their cooperation in this study; and the subjects, for their good-natured acceptance of the sampling and counting procedures.

REFERENCES

1. Maximum Permissible Body Burdens and Maximum Permissible Concentrations in Radionuclides in Air and Water for Occupational Exposure. *National Bureau of Standards Handbook* 69 (1959).
2. Report of ICRP Committee II on Permissible Dose for Internal Radiation (1959) with Bibliography for Biological, Mathematical, and Physical Data. *Health Physics* **3,** 1 (1960).
3. W. O. GENTRY, R. SCHEDE and R. C. SMITH. Monitors for Alpha Air Contamination. Sixth Tripartite Instrumentation Conference at Chalk River. AECL-802, 94 (1959).
4. N. B. SCHULTZ and A. F. BECHER. Correlation of uranium alpha surface contamination, air-borne concentrations, and urinary excretion rates. *Health Physics* **9,** 901 (1963).
5. L. M. STECKEL and C. M. WEST. Characterization of Y-12 uranium process material correlated with *in vivo* experience. Y-1544 (1966).
6. P. C. MCREE, C. M. WEST and J. D. MCLENDON. Y-12 Radiation Safety Manual. Y-1401-Revised, 35 (1965).
7. Report of Task Group on Lung Dynamics, ICRP Committee II. *Health Physics* **12,** 173 (1966).
8. B. R. FISH. Inhalation of uranium aerosols by mouse, rat, dog and man. *Inhaled Particles and Vapors,* 151 (1961).
9. W. N. SAXBY, N. A. TAYLOR, J. GARLAND, J. DUNDO and D. NEWTON. A case of inhalation of enriched uranium dust. *Assessment of Radioactivity in Man* **2,** 535 (1964).
10. C. M. WEST and L. M. SCOTT. An evaluation of cases showing uranium chest burdens with long biological half-lives. *Health Physics,* in press (1966).

(*Appendix, Tables A1–A5, follow on pages 1214–17*).

APPENDIX

Table A1. *Excretion and* In Vivo *Data. Case K-1*

Date	*In vivo* (pCi)	Urine (pCi/day)	Feces (pCi/day)	Date	*In vivo* (pCi)	Urine (pCi/day)	Feces (pCi/day)
1/12/65	34,489	209	—	7/20/65	24,895	19	27
1/15/65	50,276	67	—	7/31/65	—	24	25
1/17/65	—	65	155				
1/18/65	—	25	—	8/9/65	—	16	42
1/24/65	—	49	—	8/20/65	17,730	—	—
1/27/65	41,209	82	—	8/22/65	—	32	20
1/28/65	—	64	109	8/29/65	—	30	10
1/31/65	—	45	59				
				9/5/65	—	27	24
2/7/65	—	73	109	9/12/65	—	15	9
2/15/65	—	16	35	9/19/65	—	15	20
2/16/65	39,549	—	—	9/28/65	18,337	—	—
2/23/65	—	21	37				
2/28/65	—	32	23	10/3/65	—	17	18
				10/10/65	—	21	20
3/7/65	—	8	94	10/29/65	—	16	22
3/14/65	—	22	45				
3/21/65	—	20	36	11/1/65	16,192	—	—
3/22/65	31,858	—	—	11/13/65	—	16	20
3/28/65	—	24	39	11/28/65	—	22	20
				11/30/65	17,609	—	—
4/4/65	—	32	26				
4/12/65	—	63	38	12/12/65	—	8	15
4/18/65	—	21	16	12/26/65	—	16	10
4/25/65	—	29	33	12/29/65	11,415	—	—
4/26/65	32,465	—	—				
				1/23/66	—	8	8
5/2/65	—	59	23				
5/16/65	—	33	37	2/4/66	14,370	—	—
5/23/65	—	24	40	2/27/66	—	8	9
5/25/65	28,700	—	—				
5/31/65	—	54	10	3/17/66	16,556	—	—
6/6/65	—	32	23	4/3/66	—	6	20
6/13/65	—	27	40				
6/20/65	—	16	22	5/8/66	10,160	10	15
6/27/65	—	24	10				
				6/15/66	5667	—	—
				6/19/66	—	8	17

Table A2. *Excretion and* In Vivo *Data. Case K-2*

Date	*In vivo* (pCi)	Urine (pCi/day)	Feces (pCi/day)
5/25/65	42,673	81	—
5/27/65	42,356	111	—
6/7/65	—	62	—
6/20/65	—	60	45
6/23/65	42,582	—	—
6/28/65	—	64	—
7/2/65	—	62	25
7/21/65	37,055	55	17
8/4/65	—	32	40
8/18/65	—	34	10
8/20/65	28,403	—	—
9/1/65	—	28	41
9/15/65	30,849	—	—
9/19/65	—	40	8
10/12/65	—	21	8
10/13/65	20,929	—	—
10/31/65	—	25	15
11/11/65	23,375	18	6
11/30/65	—	14	5
1/6/66	21,925	24	1
2/3/66	—	15	2
2/4/66	18,029	—	—
3/3/66	—	15	5
3/30/66	18,392	21	10
5/25/66	—	14	2
6/22/66	18,256	11	12

Table A3. *Excretion and* In Vivo *Data. Case K*-3

Date	*In vivo* (pCi)	Urine (pCi/day)	Feces (pCi/day)
1/11/65	24,868	74	—
1/19/65	—	94	—
2/4/65	—	23	—
2/23/65	27,610	59	—
3/4/65	—	33	34
3/14/65	—	40	31
3/23/65	—	23	12
3/29/65	11,227	—	—
4/1/65	—	15	15
4/11/65	—	15	12
4/19/65	—	13	9
4/29/65	7680	11	7
5/9/65	—	30	9
5/19/65	—	130	8
5/24/65	7424	—	—
5/27/65	—	24	6
6/14/65	—	14	7
6/24/65	—	10	10
7/28/65	—	52	7
8/10/65	4425	—	—
8/19/65	—	39	56
9/19/65	—	16	7
10/7/65	7460	—	—
10/10/65	—	14	5
11/25/65	—	3	6
12/2/65	1499	—	—
1/27/66	—	8	8
1/28/66	1134	—	—
6/16/66	4132	—	—

Table A4. *Comparison of Excretion and* In Vivo *Data.*

Case	Chest burden: Urine excretion (pCi/pCi per day)		Chest burden*: Total excretion (pCi/pCi per day)		Fecal excretion*: Urine excretion (pCi/pCi)	Chest burden drop*: Total excretion (pCi/pCi)
	Initial	Final	Initial	Final		
1	270	610	—	—	0.1	0.9
UR-2	2200	3100	200	1200	1.1	2.6
Y-1	1700	2200	1200	1400	0.5	1.6
Y-2	1600	3800	900	1800	0.9	1.1
Y-3	1100	1600	520	540	1.6	0.9
Y-4	2100	4900	1300	1800	1.0	0.7
Y-5	3200	5700	920	1300	3.3	0.7
K-1	780	980	370	560	0.9	0.8
K-2	660	1100	440	840	0.4	0.5
K-3	550	160†	370	110†	1.8	0.7

* Over the period covered by fecal analyses.
† Chest burden less than 6000 pCi enriched uranium.

Table A5. *Excretion Quantities*

Case Number	Urine (volume)				Feces (mass)			
	No. samples	(ml/day)			No. samples	(g/day)		
		Min.	Avg.	Max.		Min.	Avg.	Max.
K-1	39	610	1325	1900	39	51	181	362
K-2	14	860	1369	1700	14	18	54	126
K-3	18	440	1023	1540	18	48	92	186
Total	71	440	1263	1900	71	18	135	362

DISCUSSION

W. N. SAXBY (*U.K.*):

1. Were measurements carried out in other parts of the trunk?

2. Were any further D.T.P.A. treatments carried out after the first series?

3. Is there any later data on the faecal and urinary output and in particular of their ratio?

A. BRODSKY:

1. Yes, as also mentioned in the printed version of this paper, measurements were taken near many body positions to determine the locations of internal and external contamination. However, the detailed data are voluminous, so only a sample of the more important chest measurements are to be published here.

2. Of course, it would have been of scientific interest to give further administration and follow subsequent excretions, even after the lung burden was below detectable limits. However, the medical members of this team felt that DTPA was not without its risks and thus further administrations could not be justified after the lung burden was well within safe limits.

3. Faecal excretion dropped to very low values within a few weeks after the DTPA administration. About 5 months after the accident, the employee changed jobs and we no longer have access to this data. However, the lungs and other parts of the body were counted through May, 1966, and no appreciable indication of contamination reappeared after the period covered by Table 2 of this paper.

J. R. HORAN (*U.S.A.*):

In the calibration of your phantom did you employ radioactive material from the incident itself, such as the radioactive particle recovered from the operator's undershirt?

A. BRODSKY:

Not directly, since insufficient material was available. However, samples of the contamination were examined with respect to the relative attenuation of the 17 and 60 keV peaks in water, and the information was utilized in analyzing the results obtained with the more intense 1 liter standard. Since ultimately only the 60 keV peak was used to obtain Am^{241} burdens, this procedure was satisfactory.

F. BERTHOLD (*Germany*):

For the assessment of plutonium incorporated in lungs using external detectors, Kiefer, Maushart and co-workers have described a system using a triple large area flow-counter allowing both energy and range discrimination. Their results were better than with any practical scintillation counter system.

With an improved version of this instrument it is possible to resolve the different Pu-X-ray lines. Using the relative line-intensities of the *in vivo* spectrum, a calculation of the depth from which the radiation originated and the absolute activity incorporated is possible. The detection limits are about 0.01 μCi incorporated Pu.

B. A. J. LISTER:

I am, of course, aware of the work of Maushart and the parallel work of Rundo at Harwell. The present limit of detection by direct measurement is about three-quarters of the maximum permissible lung burden. Probably with even greater sophistication of instrumentation this will be reduced—but I am not sure that the reduction will ever be considerable. We still have a great interest in assuming lung burdens well below this level and we shall still need the alternative approaches.

D. K. CRAIG (*South Africa*):

Dr. Lister, you several times mentioned that analyses of particle size were carried out. Could you please give us some idea as to the range of the observed particle size distributions involved?

B. A. J. LISTER:

Yes, I can certainly give you the range of particle sizes found in practice. The largest particles are several hundred microns and the smallest are much smaller than a tenth of a micron. We can find almost any size you care to mention.

Hub. WIJKER (*Netherlands*):

Showing his second slide, the speaker remarked that 15% of the material was soluble. Can the speaker give his opinion on how to define soluble? Soluble in body fluids or an equivalent of it? If so, what equivalent? Have other parameters to be included in the definition, e.g. the speed with which the

material dissolves, which in itself may be a function of particle shape and size?

N. B. SCHULTZ:

The solubility studies are described in detail in ref. 5 of our paper: L. M. STECKEL and C. M. WEST, Characterization of Y-12 uranium process material correlated with *in vivo* experience. Y-1544 (1966).

W. N. SAXBY (*U.K.*):

In the U.K. we have had experiences similar to those reported by Mr. Schultz. The cases he quotes are the result of incidents resulting in largish single exposures. The case he attributes to the study by my co-workers and myself (Saxby *et al.*, IAEA Symposium, Heidelberg 1964, IAEA, Vienna 1964) was also probably from a single incident. Routine monitoring of enriched uranium workers is carried out, for reasons of practicability by urinalysis—it is convenient to have a quickly useable factor to convert daily urinary output of uranium to uranium in the chest—not as an absolute measure but as a guide to further investigation. In my earlier paper (see above) a factor of 500 was suggested. Recent work with chronically exposed workers and the data quoted by Schultz to-day suggest that this figure should be reconsidered, and replaced by a factor of 1000. Mr. Wijker who preceded me asked for further information on the difficulty of deciding on the solubility of the uranium. This subject is discussed at some length by the ICRP Task Group on Lung Dynamics and their report is in the February 1966 *Health Physics* (ICRP Task Group on Lung Dynamics, *Health Physics*, Feb. 1966, Pergamon Press, Oxford, 1966). The use of the terms "soluble" and "insoluble" is perhaps unfortunate, as the phenomena observed are not solely dependent on chemical solubility as it is normally defined—chemical form and solubility, valence states, physical form and dimensions, previous heat treatment, and metabolic processes, and biological factors in the body all affect the material's subsequent movements. It would be more readily understandable to talk, not of a material being "soluble" or "insoluble", but of it being "readily transferable" within the body, "mildly transferable" or "easily transferable".

N. B. SCHULTZ:

Thank you for your comments, with which we concur.

J. CHALABREYSSE (*France*):

Sur la base de son expérience personnelle et dans le cas particulier de l'uranium, je voudrais demander à l'orateur si d'une part il est d'accord avec les conclusions du Groupe de Travail de l'ICRP concernant le schéma pulmonaire de rétention et d'épuration des particules inhalées et si d'autre part il peut préciser les différentes fractions s'épurant par voie sanguine, donc s'excrétant dans les urines, puis par voie digestive, donc s'éliminant dans les fèces, et enfin par voie lymphatique, donc susceptibles de rester fixées définitivement dans les ganglions.

N. B. SCHULTZ:

The recent Task Group report appears to incorporate the latest research and carefully weighed opinions of our scientific community.