LONGTERM EVALUATION OF AN ACCIDENTAL PLUTONIUM INHALATION

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INTRODUCTION

In the course of routine urine analysis of an EIR worker, an alpha-activity of 0,3 pCi/l was found. The isotopes identified by alpha-spectrometry were Pu-239 or Pu-240 and Am-241 or Pu-238. Further analysis confirmed an unobserved incorporation of plutonium. The only acceptable explanation was an inhalation of aerosols 2 months earlier during welding on a plutonium contaminated glove box.

MEASUREMENTS AND METHODS

To assess the activity inhaled, low energy X-ray measurements of the lung were made with phoswich detectors and excreta were collected. The first lung measurement, made in April 1980, was not clearly interpretable. An additional incorporation of 50 nCi I-125 was detected, which disturbed severely the measurement in the plutonium X-ray region. The EIR-plutonium contains always known amounts of Am-241 which emits an easily detectable γ-radiation of 60 keV. After correction for the I-125 contribution in the 60 keV energy region there remained no significant higher counting rate compared with similar persons. With a calculated detection limit of 1 nCi Am-241 in a measuring time of 40 minutes and a Pu-239/Am-241 activity ratio of 5 - 7 an upper limit for the lung activity of around 6 nCi Pu-239 at that time can be given. The activity ratio was estimated from the fuel handled in the box. Further lung measurements gave no other results.

URINE AND FECES ANALYSIS AND MEASUREMENTS

In the first year, one day every week, later on on three consecutive days every month and finally on five concecutive days every three months all excretions were collected and analysed. This programm was stopped in October 1983 because the detection limit for plutonium was reached.
From every urine sample plutonium and americium were extracted with cupferron. The samples for proportional counting and alpha-spectrometry were prepared by evaporation.
The fecal samples were ashed at 600°C. Aliquots of 0,5 g or 1 g were analysed by the cupferron-method.
Additional uranium analyses from feces were made using the aliquat-method. The activity was constant (6±3) pCi/24h.

DISCUSSION OF THE EXCRETION PATTERN

The results from the excretion measurements show large variations in the amount of alpha-activity. Statistical evaluations of the data in Fig. 1, Fig. 2 and Fig. 3 were made using linear regression methods.

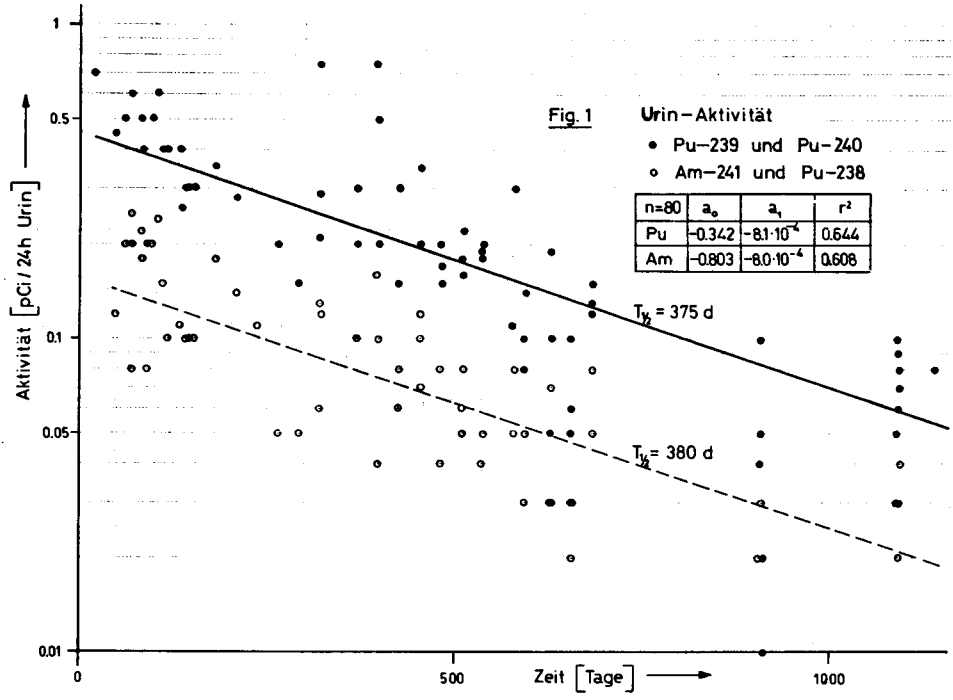

Fig. 1: Daily urinary excretion

The decrease of plutonium and americium activity in urine and feces has a half live of 370 days and 310 days, respectively. There are constant but different Pu/Am activity ratios in urine and feces. In urine the ratio is 3 and in feces it is around 5. The higher solubility of americium may explain this effect.

As compared with other publications the wide variation of the daily activity excretion is normal. One reason for this effect may be in our case a strongly changing Po-210 activity up to 0,3 pCi/day in urine and up to 3 pCi/day in feces. For low level plutonium analysis the Po-210 must be removed.

Nevertheless, we had expected a better homogeneity of the urine values, because the person under investigation had a steady drinking pattern. The daily drinking volume is 1 l mineral water, 1 l tea and some cups of coffee.

DOSE CALCULATIONS FROM EXCRETION DATA

The urine and feces results were interpreted using the ICRP 30 lung model for class Y plutonium, but with the experimentally determined biological halflife, a particle size of 1 µm AMAD, a urine volume of 1,8 l/d and a fecal ash weight of 9 g/d.
Using these parameters, we found a dose difference about a factor of

10 in interpreting the urine and feces results. This discrepancy led to the engagement of specialists from other institutions to calculate the dose from the excretion data [1][2][3].
In Tab. 1 all results are compared. In general, our findings were confirmed.

Tab. 1 Intake and committed effektive dose equivalent calculated from different institutions

INSTITUTION	eff. $t_{1/2}$ (d)	INTAKE (nCi) based on urine	INTAKE (nCi) based on feces	H_{50} (rem) from urine	H_{50} (rem) from feces
EIR-Switzerland	370	170	19	55	6
Chalk-River Canada	374	175	21	55	7
BGA-Germany	365	86	19	25	6
KFA-Germany	273	250	20	75	8
ICRP 30	500				

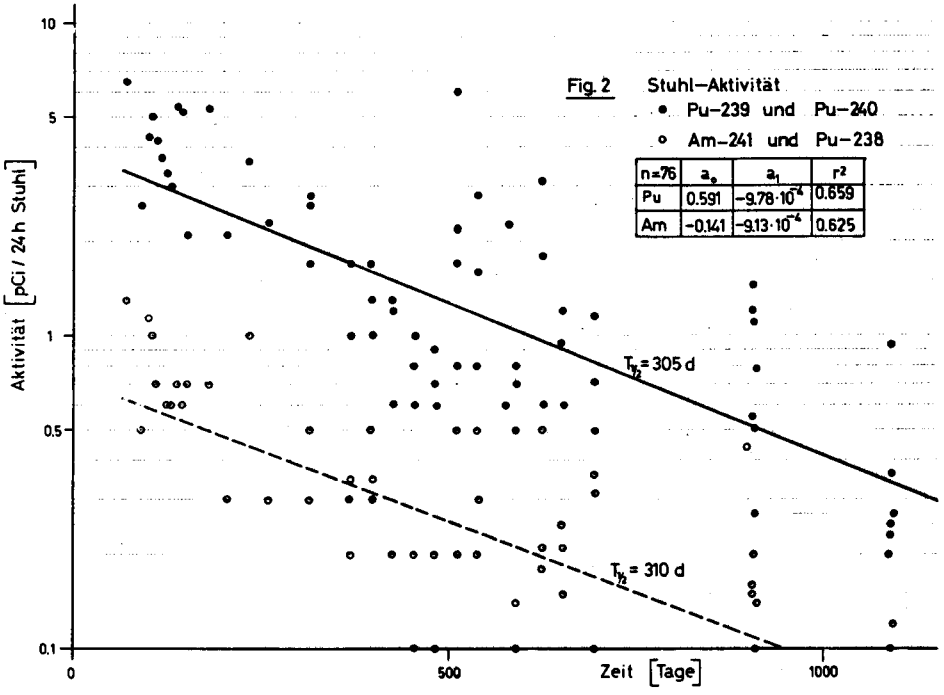

Fig. 2: Daily fecal excretion

Attempts made by J. Johnson [1] and K. Hendrichs [2] to find a dose agreement by varying single ICRP-parameters failed.

Urine samples of the worker were distributed for an international intercomparison for the determination of metabolized plutonium. It proved the absence of systematic measurement errors. The comment of all persons involved in the dose calculation was, that in this and other cases [4] the ICRP parameters are not directly applicable.

If we compare the excretion data and the lung measurements, there is no disagreement between fecal excretion and lung counting, but there is one between urinary excretion and lung counting.

Obviously the urine analysis overestimates the dose by a factor of nearly 10. This may produce serious problems with the control authorities. It is suggested that the ICRP 30 parameters of the lung model should be reviewed.

[1] J. Johnson, Chalk River Nuclear Laboratories, Canada
Private Communication
[2] K. Hendrichs, Bundesgesundheitsamt, Neuherberg, Germany
Private Communication
[3] D. Beyer, Kernforschungsanstalt Jülich, Germany, Private Com.
[4] G. L. Voelz, Health Phys., Vol. 29(4), 551-561, Oct. 75