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R. Caldwell, The Detection of Insoluble Alpha Emitters in the Lung, AEC Bioassay and Analytical Chemistry Conference, Gatlinburg, 1966.

Introduction

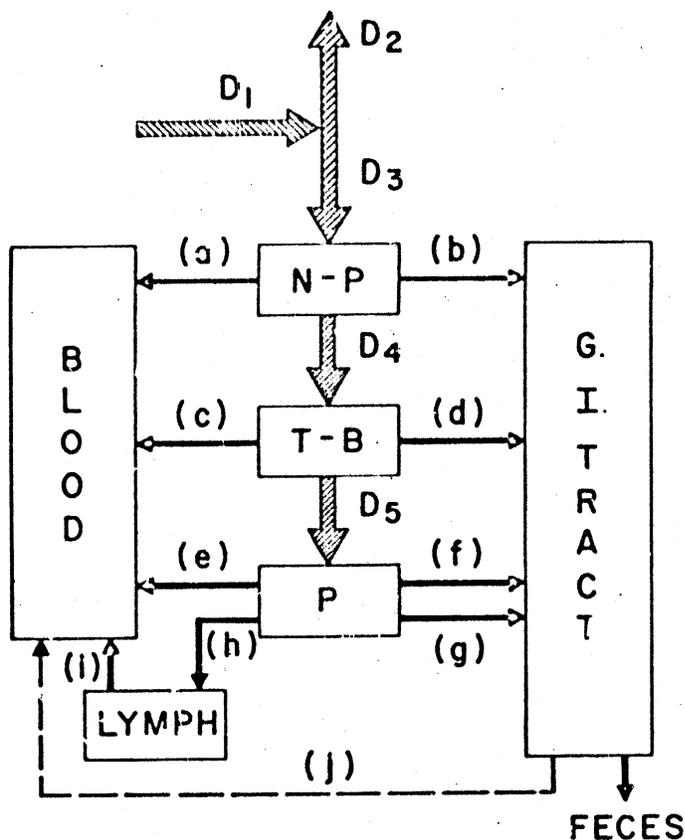
The pulmonary lung is the critical organ for many industrial exposures to radioaerosols. This is because the oxides of the actinide elements are cleared very slowly from the pulmonary compartment⁽¹⁾. As little as 0.5 microcurie of $^{239}\text{PuO}_2$ has produced a high percentage of lung cancers in beagles⁽²⁾. And studies on uranium miners from the Colorado Plateau⁽³⁾ show no human immunity to radiation induced lung disease.

If we designate the lung as a critical organ, then it is necessary to estimate accumulated lung burden in occupationally exposed workers. This paper will show that fecal sampling is the only satisfactory method for estimating lung burdens of insoluble alpha emitters. These insoluble alpha emitters are those actinide compounds classified as Class Y in the new lung model. The most important are $^{239}\text{PuO}_2$, $^{241}\text{AmO}_2$, $^{234}\text{UO}_2$ and $^{232}\text{ThO}_2$.

I. The Deficiencies of Urinalysis and In-vivo Counting for Class Y Actinides

In 1964 Sill⁽⁴⁾ pointed out the errors in using urinalysis as a routine monitoring method for internal radioactive contaminants. In his experience, radioactivity measured by whole body counting could not be detected in the urine. He found however that in all cases fecal samples showed measureable quantities of the radio nuclide.

This can be easily understood if we consider the new ICRP deposition and retention model shown in Figure 1. Inhaled particles are deposited in three regions of the respiratory tract, the nasal-pharynx, tracheo-bronchial



ICRP DEPOSITION AND RETENTION MODEL

Figure 1.

and pulmonary compartments. Almost 100 per cent of the insoluble alpha emitters deposited in the N-P and T-B regions are removed by ciliary mucous transport to the G.I. Tract in a matter of minutes. This rapidly eliminated fraction, represented by (b) and (d), together with a similarly rapidly removed pulmonary fraction (f) make up an early clearance phase (Phase I). All Phase I insoluble alpha activity is eliminated in the feces. A second clearance phase with a half time of one year or greater from the pulmonary part of the lung is represented by (e), (g) and (h). Only about 5 per cent of the Class Y material originally deposited in the pulmonary compartment is absorbed (e) into the circulating blood. Another 15 per cent is removed to the lymph system (about 10% of which is later transferred to the blood).

The remainder is eliminated by endocytosis and the ciliary escalator through the G.I. Tract to the feces.

Dolphin and other UKAEA workers have suggested that lung burdens of PuO_2 can be estimated by urine sampling. The disadvantage of this technique is obvious in light of this new lung model. Only a twelfth of the long term lung burden is absorbed into the blood and subsequently deposited on the bone and other organs. The plutonium thus deposited will be excreted very slowly. The maximum urinary excretion from a maximum permissible lung burden (16 nc) would be less than 0.2 d/m/day. This maximum would occur several months after the deposition.

On the other hand easily measured quantities of plutonium are excreted in feces. This is demonstrated in Figure 2. A fecal excretion reference level is derived. Elimination from the pulmonary lung is by way of the blood, lymph and G.I. Tract. Three basic assumptions are made:

1) the half time of elimination from the pulmonary lung is 500 days.

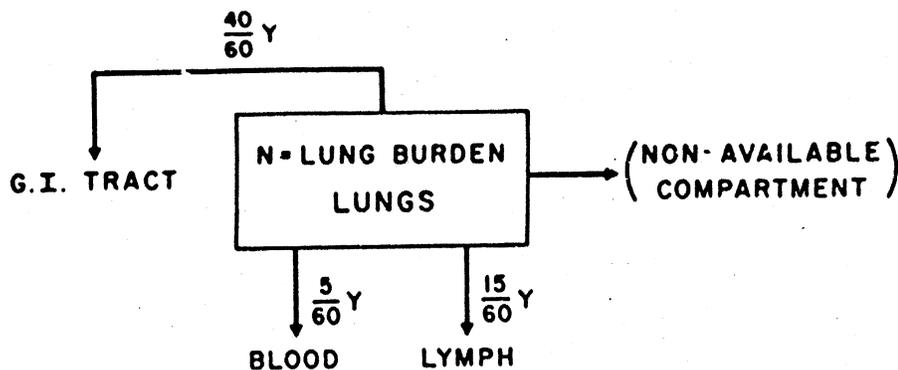
This is the recommended value in Appendix I of the Lung Dynamic Task Group's report.

2) The daily elimination from the Lung (Y) is proportional to the current burden N. This is a collary of the first assumption.

3) The original burden N_0 is equal to the sum of the daily elimination from $t = 0$ to infinity.

The calculation shows that 49 d/m/day is eliminated from a 16 nc lung burden, 32 d/m by way of the feces. One tenth this value, or 3.2 d/m/day would be a suitable reference level. Persons excreting safely below this level could be safely assumed to have non-hazardous lung burdens.

PHASE II FECAL EXCRETION REFERENCE LEVEL



I. TASK GROUP DEFINITION, PuO_2

$$N = N_0 e^{-\frac{.693}{500} t} \text{ (days)}$$

DAILY LUNG ELIMINATION (Y) \propto N

$$Y = Y_0 e^{-\frac{.693}{500} t}$$

$$\begin{aligned} \text{II. } N_0 &= \int_0^{\infty} Y dt = Y_0 \int_0^{\infty} e^{-\frac{.693}{500} t} dt \\ &= Y_0 \left[\frac{e^{-\frac{.693}{500} t}}{-\frac{.693}{500}} \right]_{t=0}^{t=\infty} \\ &= Y_0 \left[0 + \frac{500}{.693} \right] \\ N_0 &= \frac{Y_0 500}{.693} \end{aligned}$$

$$\text{III. } MPN_0 = 16 \text{ nc} = 3.5 \times 10^4 \text{ d/m}$$

$$\begin{aligned} Y_0 &= \frac{.693 (3.5 \times 10^4 \text{ d/m})}{500 \text{ days}} \\ &= 49 \text{ d/m/day} \end{aligned}$$

$$\text{IV. DAILY FECAL EXCRETION} = \frac{40}{60} Y$$

$$= 32 \text{ d/m/day}$$

Figure 2.

The non-available compartment represents possible retention of activity in the lung which is not available for elimination. This could occur by inclusion in scar tissue, etc. and would invalidate burden estimation by fecal sampling unless the fraction retained were known. This phenomenon can be seen in Sill's data⁽⁴⁾ and a case by Saxby⁽¹⁰⁾ where fecal excretion half times were shorter than chest burden elimination half times.

In-vivo or whole body gamma counting has demonstrated excellent results for the accurate assay of many radionuclides in the human body. The only requirement is the presence of an energetic gamma emitted with reasonable abundance from the radionuclide of interest.

Unfortunately, the actinide elements are not blessed with abundant energetic gamma radiation. ^{235}U is detected to levels as low as 7 NC.(6). ^{232}Th can be detected by gammas from its daughters, but redistribution of these daughters make evaluation difficult(7). Recent advances(8)(9) by using thin NaI crystals have improved the detection of ^{241}Am and ^{239}Pu . 2 nc of ^{241}Am and 16 nc of ^{239}Pu can be detected in the lung. The detection of ^{239}Pu depends on the ^{241}Am present.

There are two basic shortcomings of present In-vivo counting methods for actinide elements. Adequately shielded and sensitive whole body counters are expensive (>\$250,000) and suitably inexpensive counters cannot usually detect lung burdens smaller than permissible.

I. Fecal Sampling Experience at the NUMEC Plutonium Laboratory

Fecal sampling has been carried out at NUMEC's Plutonium facilities since January, 1966. It is used to accomplish three goals: (1) the early detection of acute inhalation exposures, (2) the estimation of detected lung burdens, and (3) the screening of potential chronic exposures.

The Early Detection of Acute Inhalation Exposures

The value of fecal sampling for the early detection of inhalation exposures is illustrated in Figure 3. This shows the excretion data for an acute inhalation exposure following a glove box explosion. The glove box, filled with propane gas from a leaking Bern-z-matic torch, exploded when

EXCRETION DATA

ACUTE INHALATION EXPOSURE, January 17, 1966

$^{241}\text{AmO}_2$

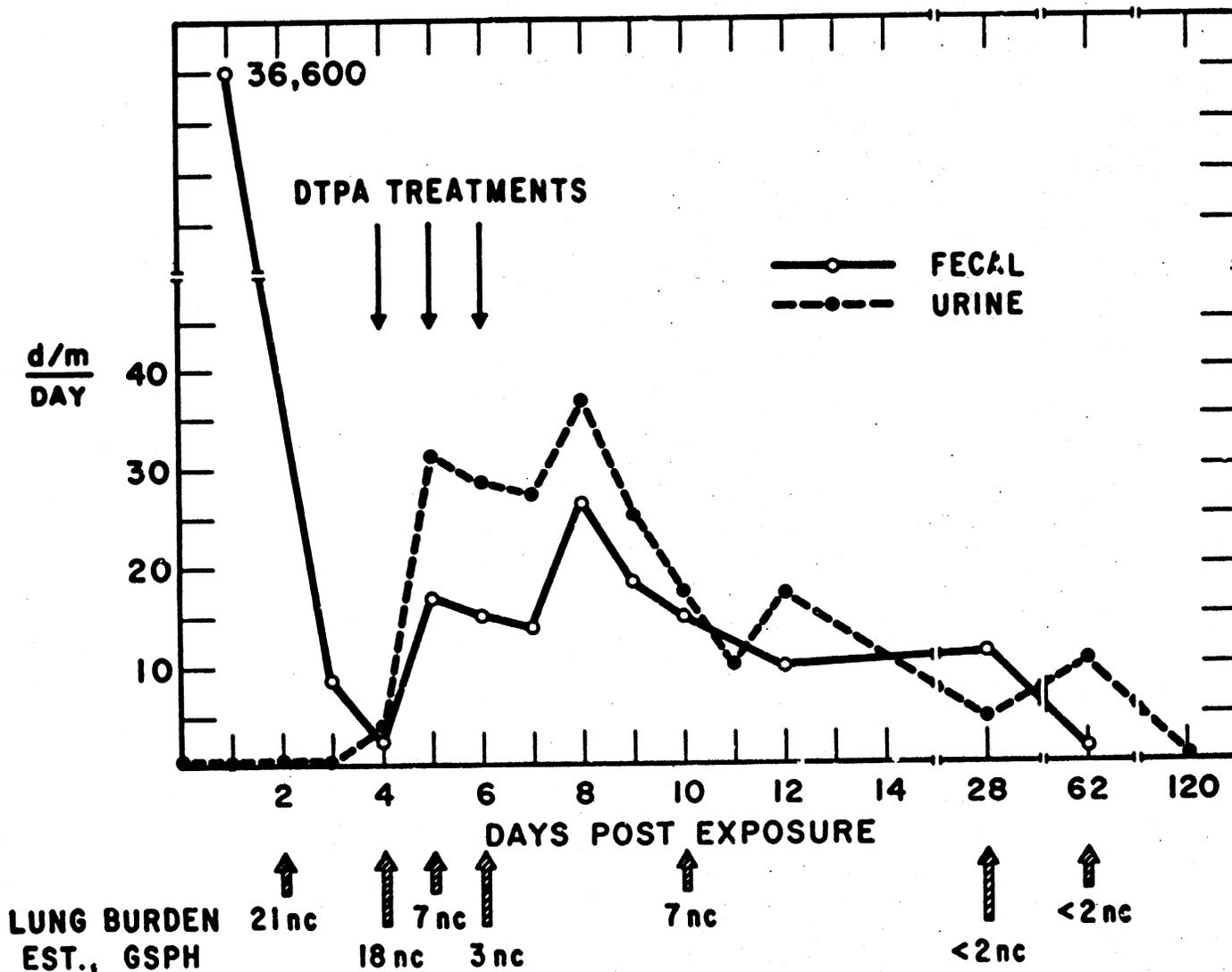


Figure 3.

the technician struck the lighter. $^{241}\text{AmO}_2$ was the main contaminant. Urine samples collected from the time of the accident through the third day showed negligible quantities of ^{241}Am . However, the first fecal sample, comprising the bulk of the Phase I clearance, contained more than 16 nc. This fact plus positive indications from whole body counts on the second day prompted medical consultants to perform chelation treatments the fourth, fifth and sixth days. The urine excretion level rose to more than 30 d/m/day and fell off over

the next 100 days to normal levels. The fecal level also rose, repeating the pattern of urine excretion. More than 2 nc were eliminated.

Lung burden estimates were made by thin crystal counting at the Presbyterian Hospital Whole Body Counter operated by Pitt GSPH personnel. These counts were performed on the second, fourth, fifth, sixth, tenth, twenty-eighth and sixty-second days past exposure. The indicated elevated counts on the second and fourth day are suspect, since residual external contamination was found on the patient's chest. When this was removed, the count dropped off by a factor of three. This decrease cannot be attributed to DTPA treatment, since less than 100 d/m were excreted in the interval. If it were true, then the value of the DTPA administration could be in doubt, since only 2 nc were ultimately excreted and several nanocuries should have been transferred from the lung to the bone.

The important point is that only the initial positive fecal sample gave a clear indication that an inhalation exposure had occurred. If only urinalysis had been used, it would have been concluded that no exposure had occurred. No DTPA treatment would have been prescribed and the technician would have unknowingly retained a lung burden of insoluble alpha emitters.

It is gratifying to learn that DTPA can effectively remove AmO_2 and PuO_2 from the lung. This possibility has been suggested by Tombropoulos⁽¹¹⁾. He also indicated that the heat treatment history of the aerosol particles might alter the effectiveness of DTPA for removing PuO_2 from the lung. Perhaps this is the reason, Rocky Flats has reported a lack of success with similar chelation treatment⁽⁸⁾.

Estimating Lung Burdens

The potential of fecal sampling for estimating plutonium lung burdens is illustrated in Figure 4. When routine fecal sampling was commenced in

EXCRETION DATA, ^{239}Pu "NITRATE" SCRAP RECOVERY OPERATOR

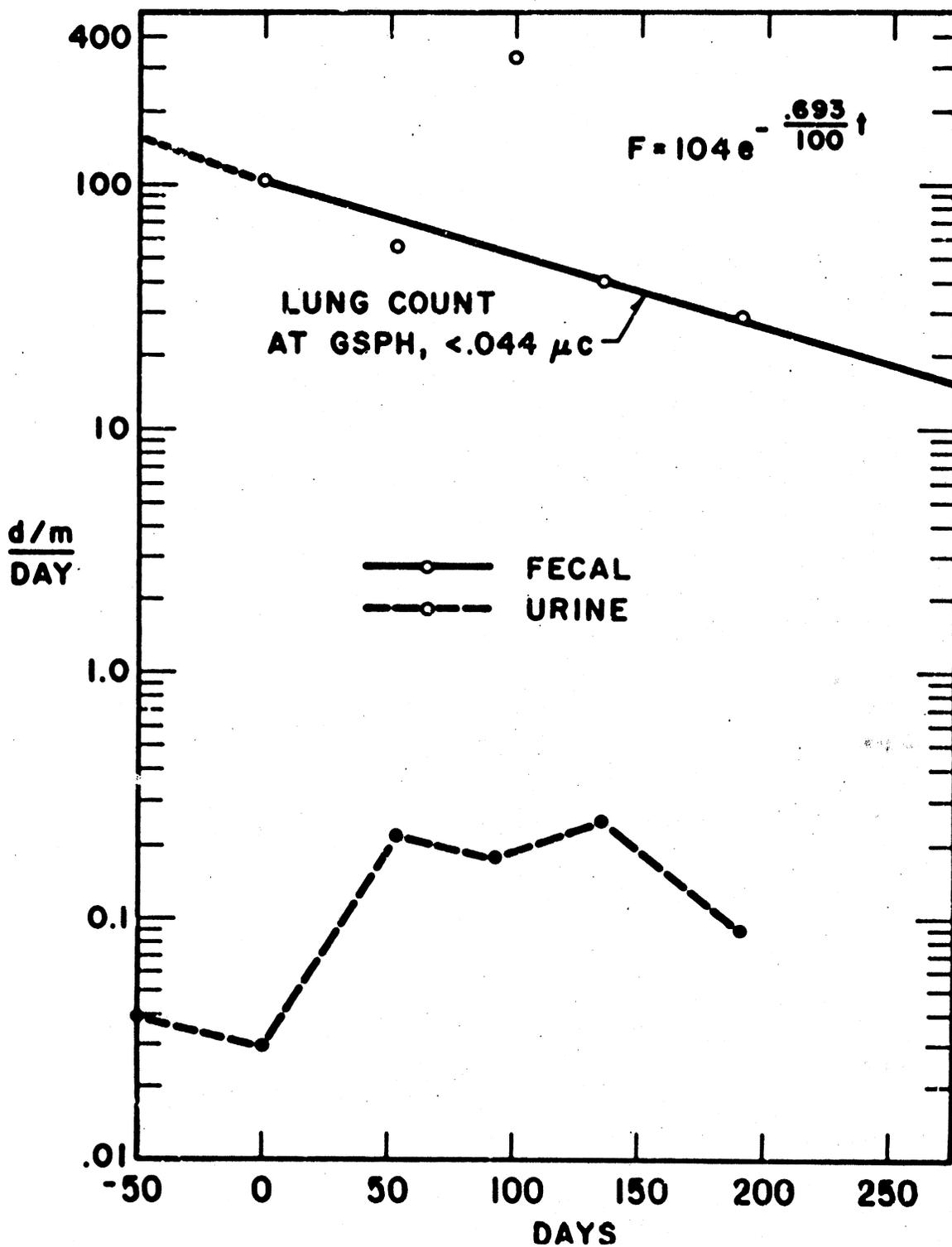


Figure 4.

early 1966, we discovered that some individuals were excreting ^{239}Pu in their feces from previously undetected exposures. These excretion data show that the fecal to urine excretion ratio may be large even for other chemical forms of plutonium. An investigation into this operator's exposure revealed that his principal exposures occurred during clean up of plutonium nitrate solution leakage from external piping. The actual chemical form of the aerosol in such cases is in doubt. However, the lung clearance seems to have a shorter half time, 100 days. The corresponding urine excretion rate seems to be about what would be expected for transferral from lung to bone.

The estimated lung burden at the time of the first fecal measurement is 7 nc.

I would like to discuss the third fecal datum point. Ordinarily, one takes stray bioassay data, records them and marks the deviation off to the vagaries of human metabolism. However, we discovered that the operator had been freshly exposed the day prior to submitting the sample. The high result probably is due to Phase I clearance from a low level exposure. We have learned that fecal sampling must be done after a person has been away from exposure for at least two days.

Routine Monitoring of Chronic Plutonium Exposures

Figure 5 shows the importance of routine fecal sampling in a plutonium bioassay program. The excretion rates via feces and urine are plotted against each other for all cases where samples were collected the same day. The UKAEA reference level is set at 0.2 pc. Exposures below this are given no further consideration. The derived fecal reference level was given earlier. It is at 1.5 pc.

URINE vs FECAL EXCRETION RATES
CHRONIC $^{239}\text{PuO}_2$ EXPOSURES

January - July, 1966

NUMEC PLUTONIUM LABORATORY

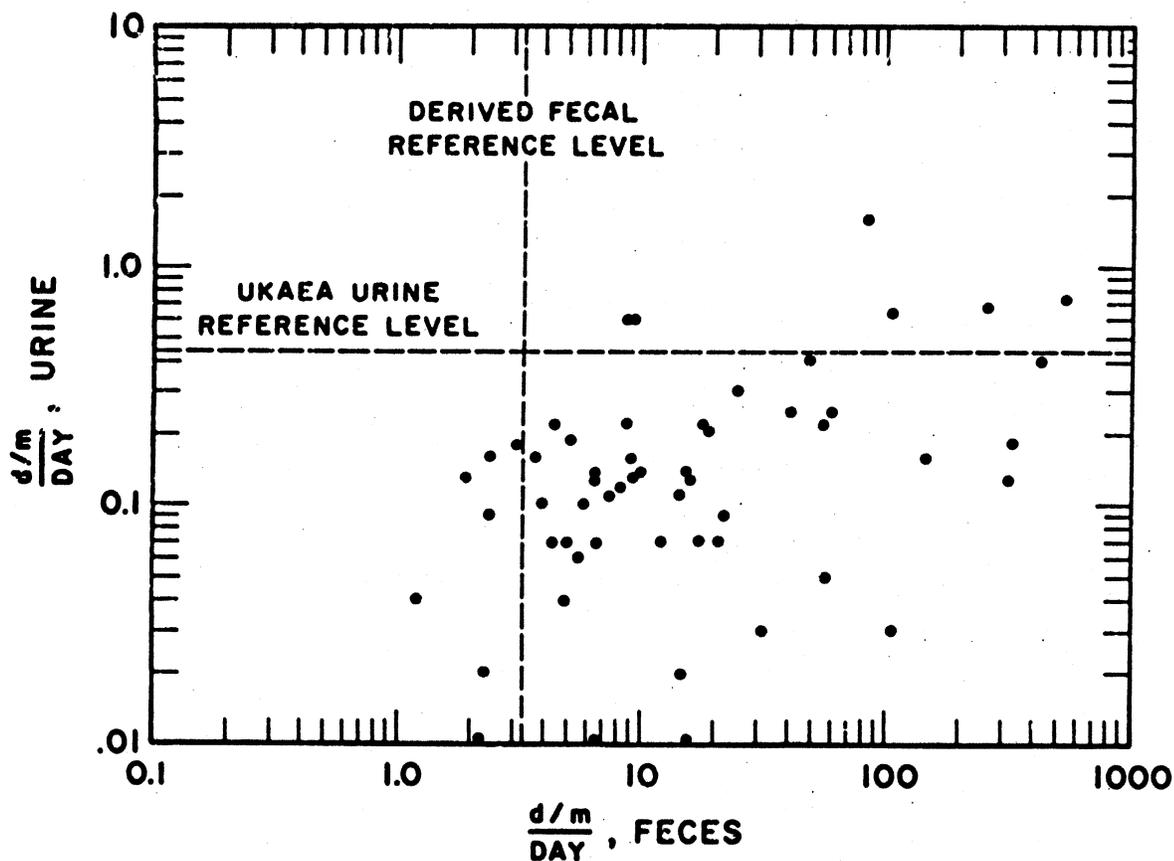


Figure 5.

Many of the fecal data undoubtedly represent Phase I clearance and are not necessarily important. However, it is obvious that urinalysis gives false assurance on the adequacy of environmental control. The information presented here caused us to investigate and correct conditions of which we had been unaware. Breathing zone sampling and fecal analysis will jolt any established plutonium facility out of its comfortable routine.

The Mechanics of Fecal Sampling

Fecal sampling is generally considered objectionable. We have found, given proper indoctrination and sampling technique, that our employees have

been as cooperative with the fecal sampling programs as they are with urine sampling.

We give the employee a quart plastic refrigerator carton, a small roll of tape, a paper bag and a written set of instructions. He takes this home to submit the sample. We have found it necessary to do home-only sampling to avoid low level contamination of samples. After depositing the sample in the carton, he replaces the lid and seals it with the tape. The carton is placed in the paper bag and brought back to the laboratory to await shipment to the Bioassay Vendor. No fuss, no smell, no messy handling problems. We even add formaldehyde, as requested by one vendor, by injecting the carton with a large hypodermic syringe. The resulting hole is sealed with plastic adhesive.

I. Preliminary Results from Uranium Fecal Sampling

Fecal sampling was begun on a large scale at NUMEC's Uranium plant in June, 1966. Generally we have found that uranium is easily detected in the urine, but that the feces is the principal route of excretion.

One interesting set of early bioassay data following exposure to a small release of enriched UF₆ is shown in Figure 6. Here the fecal excretion rate continues to increase until the fifth day before falling off. UO₂F₂ is the inhaled product during "hex" releases. This is generally considered to be highly absorbed into the blood from the lungs. Perhaps the delayed fecal excretion rate peak is an indication of elimination of systemic uranium via the feces. The urine curve ends the second day since the sequence of succeeding urine data is in doubt.

The last figure (No. 7) shows three groups of selected fecal to urine ratios. The ratios were selected from 126 cases to eliminate data where

EARLY BIOASSAY DATA
ENRICHED UF₆ EXPOSURE
 August 10, 1966

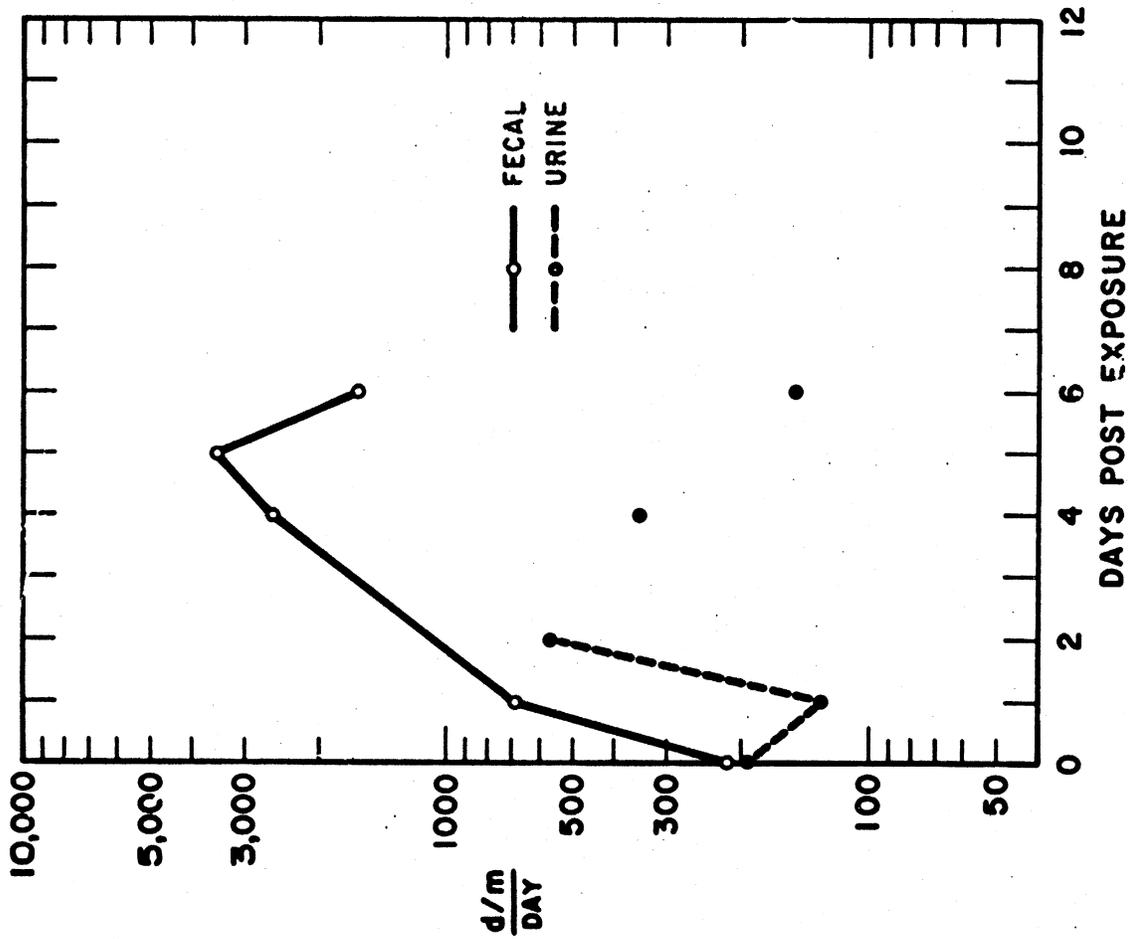


Figure 6.

FECAL / URINE RATIOS
CHRONIC ²³⁴UO₂ EXPOSURE
 July, 1966

<u>GROUP 1</u>	<u>GROUP 2</u>	<u>GROUP 3</u>
2.6	13.7	58.7
1.8	16.2	76.7
1.2	18.6	45.0
2.7	16.5	
0.6	11.1	
3.1	7.1	
4.1		
3.5		
1.0		

DATA SELECTION RULES:

1. BOTH SAMPLES COLLECTED SAME DAY
 2. OFF WORK AT LEAST TWO DAYS
 3. EXPOSURE ONLY TO UO₂
- DATA SELECTED FROM 126 CASES

Figure 7.

fecal and urine samples were not collected simultaneously, where Phase I clearance will mask the long term retained excretion and where exposures to compounds other than UO₂ are possible.

The resulting 18 ratios fall almost magically into three well defined groups: Group I has nine ratios whose mean is about 2.3, Group II's 6 cases average 14 and Group 3, 60 for 3 cases. The study is continuing and whether this log-normal distribution will remain is problematical. However, it is obvious that at least some UO₂ exposures are poorly detected by urinalysis. Whole body counting is effective for enriched uranium lung burdens greater than 7 nc. But fecal sampling is necessary for the estimation of smaller fractions of the permissible lung burden.

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Acknowledgments

I wish to express my appreciation for the past and continuing help I have received from the faculty of the Graduate School of Public Health, University of Pittsburgh, in particular Drs. N. Wald, A. Brodsky and J. Sayeg. The whole body counting data in this paper was received from them.

Eberline, Inc. performs NUMEC Plutonium urine and fecal analysis. They also do our uranium urinalysis. The quality of their work and the cooperation of Radiological Sciences Manager, Eric Geiger is greatly appreciated.

The preparation and analysis of the uranium bioassay data was done by Mr. Edward Schnell of the Uranium Plant Health and Safety Staff.