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EXCRETION DATA WITH OBSERVED LUNG
BURDENS AT AUTOPSY

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A COMPARISON OF LONG-TERM RETENTION OF PLUTONIUM
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OBSERVED LUNG BURDENS AT AUTOPSY

by

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ABSTRACT

Experience in the nuclear industry has shown that the most frequent route for significant plutonium deposition in man is by inhalation. After an accidental inhalation exposure to plutonium oxide has occurred, an attempt is usually made to determine the lung burden using in-vivo measurement techniques. However, this will not be possible if the amount deposited is less than the detection capability of the counter. Likewise, it may not be possible for even larger intakes if they are first discovered as a result of a routine examination after much of the plutonium has transferred out of the lungs. Under these conditions, the initial lung burden and dose assessment may, of necessity, be based entirely on excretion data.

This paper discusses three plutonium inhalation cases for which autopsy data are available to demonstrate the difficulties and uncertainties of such assessments and to emphasize the need for enhanced routine surveillance of the worker and work place to assure that intakes are detected at the time they occur.

An in-depth review of the findings for these three cases suggests that the use of urine excretion data applied to ICRP lung model assumptions will result in large uncertainties in estimates of the pulmonary burden and lung dose. These estimates will be improved if the material inhaled can be better characterized and by using clearance half times developed for the individual rather than the default values for Class Y material recommended by the ICRP.

INTRODUCTION

Experience in the nuclear industry has shown that the most frequent routes for intake of plutonium are by inhalation or by wounds (ROSS 1968). When the mode of intake is via a wound the individual is nearly always aware of the fact at the time it occurs and the collection of in-vivo and excreta data can be started promptly to assess the intake and resulting uptake. After an accidental intake via inhalation it is important that the initial lung burden be measured using in-vivo counting equipment. However, this is not always possible. Many intakes result in an initial lung burden of plutonium that is below the lower limit of detection in-vivo. Also, not all intakes are detected at the time they occur and the first indication that an intake has occurred may be a positive urine result collected as part of the routine monitoring program. In this case, much of the initial lung burden may have already transferred out of the lung and the residual in the lung and pulmonary lymph nodes may be less than can be measured using in-vivo techniques. Even if measurable ^{241}Am is present, it may still be difficult to determine the Pu/Am alpha activity ratio. For these cases, assessment of initial lung burden and resulting dose may, of necessity, be based entirely on excreta data and assumed mathematical models. This conclusion has been previously published (WATTS 1975) but comparisons with human autopsy data were not included for comparison with the calculated lung burdens based on urine excretion data.

SUMMARY OF CASES

Estimates of the initial pulmonary burden and the residual burden in lung and pulmonary lymph nodes of three former plutonium workers at the time of death were calculated based on urine excretion data and, when available, in-vivo chest measurement data applied to the ICRP Publication 30 (ICRP 79) lung models. The initial burden was calculated by assuming a time after intake that would result in 95% transfer of the inhaled plutonium that would ultimately be transferring from pulmonary compartment to body fluids. For non-transportable material this is about 10 clearance half times of material leaving the pulmonary compartment. The calculated residual lung and pulmonary lymph node burden at time of death was then compared to the activity in the lungs and pulmonary lymph nodes determined from post mortem radiochemical analysis. The radiochemical method to determine the residual lung and lymph burden post mortem has been described (MCINROY 1979). Post mortem data were made available through the courtesy of the United States Transuranium Registry.

Case 1

Summary

Occupation	Maintenance Worker
Year of Intake	1956
Year of Death	1971

The subject identified as Case 1 incurred an intake of plutonium via inhalation in 1956. Since in-vivo capabilities were not available at that time all calculations are made using urine excretion data. Based on application of these data to the Healy Model (HEALY 1957) it was estimated

that the subject incurred an intake that would result in a systemic deposition of 22 nCi of nontransportable plutonium with a clearance half time of 140 days for the fraction transferring out of the pulmonary compartment to the lymph or body fluids. This suggests a clearance half time of 280 days for the material leaving the lymph since ICRP Publication 30 suggests that for this pathway the clearance half time is twice as long as the clearance half time for the material leaving the pulmonary compartment. This also implies that 21 nCi (95% of the systemic burden) would have been transferred to body fluids by 1300 days post intake. The above data yield an initial pulmonary burden of 114 nCi and a residual lung and pulmonary lymph node burden of 1.7 nCi at time of death 16 years post intake, whereas only 0.3 nCi were found by post-mortem radiochemical analysis.

Case 2

Summary

Occupation	Process Operator
Year of Intake	1962
Year of Death	1971

The subject identified as Case 2 was involved in several potential accidental intakes but the primary intake is assumed to have occurred in 1962 nine years before death in 1971 and resulted in a systemic deposition of 1.7 nCi of nontransportable plutonium. The urinary excretion pattern suggests that the ICRP recommended clearance half-times of 500 days for the fraction leaving the pulmonary and 1000 days for that leaving the lymph are appropriate for this subject. Using the ICRP clearance rate constants implies that 95% of the ultimate systemic deposition would have been transferred to body fluids by day

5000 post intake. These data yield an initial pulmonary deposition of 8.8 nCi and a residual lung and pulmonary lymph burden at time of death nine years later of 0.2 nCi.

In-vivo measurements obtained in 1968, six years post intake, indicated a current lung and pulmonary lymph burden of 34 nCi of plutonium, an initial pulmonary burden of 183 nCi and a residual lung and pulmonary lymph burden of 3 nCi at time of death in 1971. These estimates of the residual lung and pulmonary lymph burden at time of death of 0.2 nCi based on urine excretion data and 3 nCi based on in-vivo measurement data can be compared to the 20 nCi measured in lung and pulmonary lymph tissue by post-mortem radiochemical analysis.

Case 3

Summary

Occupation	Millwright
Year of Intake	1963
Year of Death	1980

The subject identified as Case 3 was exposed to similar working conditions as the subject in Case 2; however, neither the incident data nor the early urine data suggested any significant intake had occurred. Evaluations based on urine excretion data and ICRP recommended clearance rate constants since the urine excretion data were not useful in predicting clearance half-time, suggest a systemic deposition of 2.6 nCi of nontransportable material, an initial pulmonary burden of 14 nCi with the intake occurring in 1963 and a residual lung and pulmonary lymph burden at time of death in 1980 of about 1 nCi. This estimate of 1 nCi burden compares favorably with the 1.7 nCi found in lung and pulmonary lymph by post-mortem radiochemical analysis.

DISCUSSION

The lung model published by the International Commission on Radiological Protection in Publication 30 (ICRP 1979) specifies that clearance from the pulmonary lung compartment to body fluid will be as follows:

Class Y Compounds

- a. Five percent of that initially deposited in the pulmonary compartment (P_0) will transfer directly to body fluids with a clearance half-time of 500 days, and that
- b. fifteen percent of P_0 will transfer to lymph nodes with a clearance half-time of 500 days. It further specifies that 10% of the 15% transferring will remain in the lymph indefinitely and that the other 90% will translocate from lymph node to body fluids with a clearance half-time of 1000 days.

Class Y Compounds

FIGURE 1. Clearance Pathways from the Pulmonary Compartment of Lung to Body Fluids Including the Fractions Transferring by Each Pathway and the Assumed Clearance Half-Times for Each Pathway for Class Y Materials

METHODS

A schematic of these clearance pathways is shown in Figure 1.

The following equations from ICRP 30 are used to calculate the residual content of the pulmonary, lymph node and fluid compartments with no clearance from the body fluids being considered for this paper.

$$(1) \quad P_t = P_0(f_1 e^{-\lambda_1 t} + f_2 e^{-\lambda_2 t})$$

$$(2) \quad L_t = f_2 P_0 \left(\frac{\lambda_2}{\lambda_3 - \lambda_2} \right) (e^{-\lambda_2 t} - e^{-\lambda_3 t})$$

$$(3) \quad F_t = f_2 f_3 P_0 \left(1 + \frac{\lambda_2}{\lambda_3 - \lambda_2} \right) e^{-\lambda_3 t} - \frac{\lambda_3}{\lambda_3 - \lambda_2} e^{-\lambda_2 t} + f_1 P_0 (1 - e^{-\lambda_1 t})$$

$$(4) \quad P_0 = \frac{F_t}{\left[f_2 f_3 \left(1 + \frac{\lambda_2}{\lambda_3 - \lambda_2} e^{-\lambda_3 t} - \frac{\lambda_3}{\lambda_3 - \lambda_2} e^{-\lambda_2 t} \right) + f_1 (1 - e^{-\lambda_1 t}) \right]}$$

where

P_0 = initial pulmonary deposition

P_t = pulmonary deposition at time t post intake

L_t = lymph node burden at time t post intake

F_t = amount transferred to body fluids by time t post intake

f and λ = as shown in Figure 1

t = time in days required for 95% of the material that will transfer to body fluids to have completed the transfer.

Similarly the chest burden over which in-vivo measurements are made (excluding the possible contribution from ribs) may be expressed as:

$$(5) \quad C_t = P_t + L_t$$

where

C_t = residual lung and pulmonary lymph burden at time t

P_t = pulmonary deposition at time t

L_t = lymph node burden at time t

t = time in days post intake

C_t for Class Y compound may then be expressed as:

$$(6) \quad C_t = 0.05 P_0 e^{-\lambda_1 t} + 0.15 P_0 e^{-\lambda_2 t} + 0.15 P_0 \left(\frac{\lambda_2}{\lambda_3 - \lambda_2} \right) (e^{-\lambda_2 t} - e^{-\lambda_3 t})$$

which can be reduced to:

$$(7) \quad C_t = P_0 [0.05 e^{-\lambda_1 t} + 0.15 e^{-\lambda_2 t} + 0.15 \left(\frac{\lambda_2}{\lambda_3 - \lambda_2} \right) (e^{-\lambda_2 t} - e^{-\lambda_3 t})]$$

The health physics data for three cases involving plutonium inhalation for which both urine excretion data and tissue analysis data at autopsy are available were reviewed. The intakes all occurred prior to the time in-vivo measurement capability was developed thus no initial in-vivo measurement data are available. Limited in-vivo measurement data at later times are available for Case 2. For each case, the initial pulmonary deposition (P_0), is

estimated from ~~on~~ urine excretion data and is used to estimate the lung and lymph node burden at time of death. For Case 2 an estimate of lung and lymph node burden at time of death was also made using in-vivo chest measurement data and these estimates were compared to lung and lymph node burdens found by post-mortem radiochemical analysis of tissues.

DATA ANALYSIS

Case 1

This case involves the inhalation of a mixture of transportable and nontransportable plutonium by a maintenance worker. The intake occurred in 1956 during maintenance work on a centrifuge used in the plutonium purification process. The centrifuge had been used to co-precipitate plutonium with LaF_3 which was subsequently metathesized with KOH to a mixture soluble in HNO_3 . However, no attempt was made to determine the solubility of the material inhaled. Since the cell was grossly contaminated with process material, resolution of the exact chemical form at this time is not feasible.

Application of the Langham (LANGHAM 1956) and Healy (HEALY 1957) models to the urine excretion data for this individual indicated a systemic burden of 30 nCi of plutonium, of which 22 nCi was due to nontransportable material. The fit of the Healy model suggested a clearance half-time for the less transportable portion of 140 days. No in-vivo examinations were obtained for the individual as the intake occurred in 1956, prior to the time in-vivo counters were available. Thus, estimating the initial pulmonary burden, P_0 and the residual in the pulmonary and lymph nodes at death is dependent upon urine excretion data only.

Assuming clearance half-time of 140 days for f_2 pathway (pulmonary to lymph) and 280 days for the f_3 pathway (lymph to fluids) are appropriate, and substituting the following values for Class Y materials in Equation 4:

$F_t = 22$ nCi of nontransportable plutonium

$$f_1 = 0.05$$

$$f_2 = 0.15$$

$$f_3 = 0.9$$

$$\lambda_1 = 0.005$$

$$\lambda_2 = 0.005$$

$$\lambda_3 = 0.0025$$

$$t_1 = 1300 \text{ days}$$

$$t_2 \text{ and } t_3 = 5000 \text{ days}$$

yields a value for P_0 for a Class Y compound as follows:

$$P_0 = \frac{(22\text{nCi})(0.95)}{(f_2 f_3) \left(1 + \frac{\lambda_2}{\lambda_3 - \lambda_2} e^{-\lambda_3 t} - \frac{\lambda_3}{\lambda_3 - \lambda_2} e^{-\lambda_2 t} \right) + f_1 (1 - e^{-\lambda_1 t})} = 114 \text{ nCi}$$

Applying Equation 7 to these initial pulmonary estimates yields residual burdens in lung and lymph at death approximately 16 years post intake of:

$$C_t = P_0 [0.05 e^{-\lambda_1 t} + 0.15 e^{-\lambda_2 t} + 0.015 (1 - e^{-\lambda_2 t}) + 0.135 \left(\frac{\lambda_2}{\lambda_3 - \lambda_2} \right) (e^{-\lambda_2 t} - e^{-\lambda_3 t})] = 1.7 \text{ nCi}$$

Based upon tissue analysis at autopsy, the residual burden in lung and pulmonary lymph was 0.3 nCi. This is much less than the 1.7 nCi expected based on urine excretion data. The faster clearance suggests that the deposition may have behaved more like Class W than Class Y material. Perhaps this is appropriate since the urine excretion data applied to the Healy Model suggested a clearance half time of 140 days (between Class W and Class Y) rather than the standard 500 days proposed for Class Y materials.

Case 2

The subject identified in Case 2 was employed as a process operator and worked in a plutonium purification facility for over 20 years with plutonium in the form of oxides, nitrates, fluorides, and metals. During this period, he was involved in 18 reported incidents in which he was potentially exposed to airborne plutonium. For most of this period, in-vivo counting equipment was not available and evaluation of any uptake was dependent upon urine excretion data.

Special and routine urine results were normal for his first 12 years of employment. Slightly elevated results were obtained beginning in 1963 indicating that an intake had been incurred. Evaluation of the excretion data suggested an intake of 0.6 nCi of transportable material plus 1.7 nCi of nontransportable material occurred in May 1962. Results of urine samples processed for Pu over the next eight years, until retirement in 1971, were low with only an occasional positive (0.05 to 0.1 d/m/day) result being observed. It is possible that the May 1962 intake date assigned was in error and that the individual had incurred an intake several years earlier which was not detected until translocation from the lung was sufficient to be seen in the

urine. This would suggest that the material inhaled was extremely insoluble. A later review of reported incidents did not reveal any such potential intake except for a chemical explosion in a process glove box in 1951. The details for this incident did not suggest that any substantial intake was likely. For the purposes of this paper, it is assumed that an intake in May 1962 resulted in a systemic burden estimated at 2.3 nCi based on urine excretion data including 1.7 nCi, nontransportable. Applying Equation 4 to the nontransportable portion of the deposition implies that the initial pulmonary burden was:

$$P_o = \frac{(1.7 \text{ nCi}) (0.95)}{0.135 (1 - 2 e^{-3.25} + 2 e^{-6.5})} = 13.3 \text{ nCi}$$

And by applying Equation 7 to this estimate, the residual quantity in pulmonary/lymph at death was:

$$C_t = (0.015 (13.3)) = 0.2 \text{ nCi}$$

In 1967, in-vivo counting for plutonium was begun, using ^{241}Am as a tracer and estimating the plutonium burden based on the Pu/Am alpha ratio. In June 1968, a chest measurement for the individual was made following his involvement in another incident. The measurement indicated 2 nCi of ^{241}Am and this remained constant through his final chest measurement in 1970. The Pu/Am alpha ratio was not known, but was estimated, assuming that the plutonium had been recently separated (<1 yr) and that the ^{241}Am measured beginning in 1968 was due to ingrowth from the ^{241}Pu present in the plutonium mixture, and

that the plutonium and americium clearance patterns were the same. Using these assumptions, a ratio of 17 was estimated for the Pu/Am alpha in 1968.

This suggested a plutonium thorax burden of 34 nCi at six years post intake.

Applying this burden to Equation 7 yields an initial pulmonary burden estimate of 183 nCi and a residual lung/lymph burden at time of death of 3 nCi.

The amounts estimated to be in lung/lymph at time of death based on

bioassay data (0.2 nCi based on urine excretion data and 3 nCi based on

in-vivo measurements) do not compare well with the 20 nCi measured in

lung/lymph at autopsy. It suggests that clearance half-times of 500 days from the pulmonary and 1000 days from the lymph nodes were in appropriate for this case.

Case 3

The work history of this subject was very similar to that of the subject in Case 2 except that he was employed as a millwright for a period of 20 years (1947 to 1967) and was involved in maintenance work during this period. He was reportedly involved in eight incidents having a potential for intake of plutonium, primarily in the oxide form; however, neither the incident data nor the urine excretion data suggested any significant deposition occurred as a result of these reported incidents.

In 1964, urine results were slightly positive (0.05 to 0.1 d/m per day) and continued to be positive until the time of his retirement in 1967. An evaluation of these data using the Healy model suggested a systemic deposition of 2.6 nCi of nontransportable plutonium with the intake occurring in 1963. The intake date was assumed to be one day after the last prior negative

sample. No in-vivo measurements were made since the subject retired before this capability existed.

The estimated systemic burden of 2.6 nCi nontransportable plutonium applied to Equation 4 suggests an initial pulmonary burden of 14 nCi if the intake occurred in 1963. This value applied to Equation 7 suggests a residual lung/lymph burden at time of death in 1980 of about 1 nCi. Based on tissue analysis at time of autopsy a total of 1.7 nCi was found in the lung/lymph.

CONCLUSION

Estimates of the initial pulmonary burden, based on urine excretion data alone does not appear to be consistent with findings based on tissue analysis at autopsy and can be very misleading, especially if any of the data needed to make the calculations are assumed. In-vivo measurements were not generally available for these early cases, but if available undoubtedly would have provided a better means of calculating the initial pulmonary burden. More evaluations comparing estimates of residual lung and pulmonary burdens based on urine excretion data to burdens found by post mortem radiochemical analysis of tissue are needed; and should be made as more autopsy data becomes available from the transuranic registries being conducted.

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