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Richland, Washington 99352

ROUTINE AND EMERGENCY
EVALUATIONS OF INTERNAL EXPOSURE
AT HANFORD
JULY 1971

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U. S. ATOMIC ENERGY COMMISSION
Under Contract AT(45-1) 1830

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EVALUATIONS OF INTERNAL EXPOSURE AT HANFORD

K. R. Heid, J. J. Jech, and
P. E. Bramson

Pacific Northwest Laboratories
BATTELLE MEMORIAL INSTITUTE
Richland, Washington

July 1971

This paper was prepared for oral presentation at the 16th Annual Meeting, Health Physics Society, New York, New York - July 12-16, 1971.

Based on work performed under United States Atomic Energy Commission Contract AT(45-1)-1830.

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INTRODUCTION

In recent years transplutonium nuclides have been used more and more extensively in many facilities. They will undoubtedly find many important applications in the fields of nuclear, chemical, space, and biomedical in the near future. The transplutonium production program of the AEC will make available increasing amounts of very heavy elements. Safe handling of these elements will depend not only on the radiation characteristics of each, but also on their use. In a production facility, for example, it is possible to standardize handling procedures to a much greater extent than is possible in a research facility. Design engineering methods should be used to develop better equipment which will result in safer working conditions. Radiation protection controls which have over the years proven satisfactory for work with plutonium, must be applied and enforced even more vigorously for transplutonium materials since on a mass basis, contamination and radiation control problems are much greater than for plutonium. The transition from plutonium to transplutonium can be facilitated by providing extra training to involved workers.

CONTROLS

The design of a facility to handle transplutonium material should be concerned with two major areas: prevention of unnecessary exposure, both internal and external; and area monitoring to facilitate early detection of non-standard conditions.

Because of the higher degree of radiation hazard associated with these materials, administrative control assumes much greater importance. Contamination spreads which were in the nuisance category when plutonium was involved will be serious problems when transplutonium material is involved. Several papers on this subject are available in the literature.^{(1) (2) (3) (4)} Despite the fact that complete containment is the order of the day, occasional releases to the work areas are inevitable. To assess the effect on workers of such releases, the radiation protection program must be able to quickly detect the release, identify the workers at risk, evaluate the internal and external exposure to the workers and make recommendations as appropriate for improvements in equipment and techniques.

INTERNAL DOSIMETRY

Evaluation of internal plutonium contamination has advanced a great deal in the last few years, especially as the result of improved in-vivo measurement techniques and the new ICRP Lung Model.⁽⁵⁾ In-vivo measurements for plutonium are usually based on the detection of the 17 KeV photons from the plutonium itself or the 60 KeV photons from the americium associated with the plutonium. To be effective, utilization of this last technique requires supporting data such as the knowledge of isotopic composition of the mixture in addition to knowledge of the particle size of the aerosol inhaled, and certain questionable assumptions regarding the metabolism of the material in the body and the application of lung model concepts. These will be discussed in more detail later in this paper.

ROUTINE SURVEILLANCE PROGRAMS

Because of the many problems inherent in interpreting internal contamination, an effective routine surveillance program for workers is difficult. Probably the most effective

routine surveillance program for workers handling plutonium has been one combining bioassay sampling with routine in-vivo chest examinations. The frequency of these examinations will depend upon the intake potential for each worker. Such a program provides reasonable assurance that a plutonium intake of concern will not go undetected.

Up to the present time, we have had relatively little experience at Hanford with unsealed transplutonium sources, but we would anticipate establishing a similar surveillance program for transplutonium should such work increase to the extent it was needed. The need for such a program is obvious when the maximum permissible body and organ burdens are reviewed. The extremely small masses that constitute maximum permissible lung and systemic burdens are summarized for selected elements in Table I.

Detection capabilities, using in-vivo counting techniques are adequate for most of these elements. Existing capabilities for detecting selected transplutonium elements utilizing in-vivo examination techniques at Hanford are summarized in Table II. The obvious exception is berkelium. Surveillance for workers handling this element is totally dependent upon bioassay sampling. As can be seen from Table II, the minimum detectable amount is significantly lower if the element can be positively identified and the low energy x-ray emissions utilized to interpret the measurement. Ideally, an employee will be working with only one transplutonium element during a given period of time. In actual work situations such as separations processes, this will usually not be possible. When the latter situation exists and positive in-vivo measurements are obtained, it may be necessary to identify the material through other means such as chemical analysis of bioassay samples collected.

MAXIMUM PERMISSIBLE BURDENS - LUNG AND SYSTEMIC
FOR SELECTED TRANSPLUTONIUM ELEMENTS

<u>Isotope</u>	<u>Maximum Permissible Lung Burden</u>		<u>Maximum Permissible Systemic Burden</u>	
	<u>μCi</u>	<u>μg</u>	<u>μCi</u>	<u>μg</u>
Plutonium-239	1.6×10^{-2}	2.6×10^{-1}	4×10^{-2}	6.4×10^{-1}
Americium-241	1.5×10^{-2}	4.6×10^{-3}	5×10^{-2}	1.5×10^{-2}
-243	1.6×10^{-2}	8.4×10^{-2}	5×10^{-2}	2.7×10^{-2}
Curium-244	1.4×10^{-2}	1.8×10^{-4}	1×10^{-1}	1.2×10^{-3}
Berkelium-249	12.	6.7×10^{-3}	7×10^{-1}	4.2×10^{-4}
Californium-252	4.0×10^{-3}	6.2×10^{-6}	1×10^{-2}	1.9×10^{-5}
Einsteinium-254	1.2×10^{-2}	1.1×10^{-5}	2×10^{-2}	1.9×10^{-5}

TABLE I

IN-VIVO COUNTING CAPABILITIES
FOR SELECTED TRANSPLUTONIUM ELEMENTS

<u>Isotope</u>	<u>X-rays (keV)</u>	<u>MDA* (μCi)</u>	<u>γ Photons (keV)</u>	<u>MDA* μCi</u>
Plutonium-239	17	1×10^{-2}	39 & 384	4×10^{-1}
Americium-241	17	5×10^{-4}	60	2×10^{-4}
Americium-243	17	3×10^{-3}	75	2×10^{-4}
Curium-244	17	3×10^{-3}	43	4×10^{-1}
Berkelium-249	~ 18	1×10^5	320	1×10^2
Californium-252	~ 19	3×10^{-3}	100	5×10^{-1}
Einsteinium-254	~ 20	2×10^{-4}	63	4×10^{-3}

TABLE II

* Minimum detectable amount in the chest area.

EVALUATION OF INTAKES

As part of a long-range study, various data associated with Pu incidents have been collected in an effort to develop guides useful in making early initial assessments. Guides based on these data have been useful at Hanford for estimating the intake of plutonium utilizing activity on nasal smears and initial in-vivo measurements. These guides are based on 30 oxide inhalation cases in which extreme care was utilized in obtaining data, especially that involving nasal smears, to preclude cross contamination from the lip or nearby skin surfaces. The relationships are derived from the nasal smear data, in-vivo lung measurements and the new ICRP Lung Model concepts.

Based on the first guide shown in Table III, 1 nCi nasal smear indicates an initial pulmonary burden of 1 nCi and a systemic burden of 0.1 nCi. However, judgment must be used in making an early estimate based on nasal smear data since incident details such as particle size, breathing habits of the exposed person and cross contamination may cause large variance in the correlation.

Based on the second guide shown in Table III, an initial in-vivo measurement of 1 nCi of ^{241}Am as measured using the lung counter indicates a plutonium lung burden of 15 nCi since we assume a standard ratio of $\text{Pu}\alpha$ to $\text{Am}\alpha$ of 15 until a more precise ratio can be established. This also relates to 5 nCi Pu initial pulmonary burden and a resultant systemic Pu burden of 0.5 nCi. These guides have been published⁽⁶⁾ along with a few words of caution on limitations for use of the data at Hanford.

In the event of an intake of transplutonium material at Hanford, data similar to that obtained for an intake of plutonium would be collected. Techniques used to evaluate

NASAL SMEAR ACTIVITY GUIDE

<u>Nasal Smear</u>	<u>Initial Pulmonary Burden</u>	<u>Initial Lung Burden</u>	<u>Ultimate Systemic Burden</u>
1 nCi	1 nCi	3 nCi	0.1 nCi

The above data are based on 30 plutonium oxide inhalation cases at Hanford. Relationships are derived from initial nasal smear data that were obtained using extreme care to prevent cross contamination from lip or facial contamination and from evaluations of depositions using early fecal data, lung counter examination data and the new ICRP Lung Model Concepts. The correlations made appear to be valid within a factor of ± 5 .

IN-VIVO DATA GUIDE

<u>Initial Lung Burden-²⁴¹Am</u>	<u>Initial Lung Burden - Pu</u>	<u>Initial Pu Pulmonary Burden</u>	<u>Resultant Systemic Burden-Pu</u>
1 nCi	15 nCi	5 nCi	0.5 nCi

These guides are based on the new ICRP Lung Model Concepts for plutonium oxide inhalation. A $Pu\alpha/Am\alpha$ ratio of 15 is assumed until the ratio is determined. The guides appear to provide reasonable estimates when applied to several Hanford cases.

Note: Guides should be used only for initial estimates primarily as an aid in determining prompt mitigatory action.

TABLE III

Guide for Estimating Internal Deposition of Plutonium
from Inhalation of Plutonium Oxide

plutonium intakes at Hanford, which have been previously reported, (7) (8) (9) provide sufficient information to make prompt, interim and formal evaluations of known or suspected intakes. The techniques utilized for cases involving intake of plutonium via inhalation are reviewed below.

TYPES OF DATA COLLECTED - PU CASES

The detection of plutonium on protective clothing, skin, nasal smears, nose blows, sputum samples, work area surfaces, etc. are usually the first indicators of the severity of an inhalation that has or may have occurred. Other supporting data that will assist in evaluation of the intake and which are collected as soon as practical after the incident are shown in Table IV.

Isotopic composition data are necessary to determine the Pu alpha to Am alpha ratio and to establish the ^{241}Pu content. This ratio is vital for evaluating lung counter data at Hanford since the technique used involves the measurement of the 60 keV photon from the ^{241}Am . Knowledge of the ^{241}Pu content is essential at Hanford to predict the ^{241}Am growth which will affect subsequent lung counter examinations. The importance of this factor has been discussed in detail in several publications. The necessary isotopic analysis is obtained by a combined mass and alpha spectrometry analysis.

PROMPT ASSESSMENT

The first assessment of the severity of the intake is based on extremely limited data. The nasal smears offer the first clue using the guide described earlier. An early in-vivo chest measurement as well as spot samples of urine and feces provide supporting information.

Rush results of early urine samples are applied to the Langham model as an additional aid in making prompt

- ✓1. URINE ANALYSIS
- ✓2. FECES ANALYSIS
3. IN-VIVO EXAMINATION
- / 4. ISOTOPIC COMPOSITION AND PU ALPHA/AM-241 ALPHA RATIO
5. CHEMICAL FORM OF THE AEROSOL
6. SOLUBILITY OF THE AEROSOL
- ✓7. PARTICLE SIZE (AIR SAMPLES - NASAL SMEARS)
- ↓ 8. NASAL AND SKIN CONTAMINATION ACTIVITY
9. CONCENTRATION OF PLUTONIUM AEROSOL
10. DURATION OF EXPOSURE
11. OTHER DETAILS OF INCIDENT

TABLE IV

Types of Data Which may be Collected for
Plutonium Inhalation Incidents

assessments. This early assessment of the probable severity of the intake is most useful if available within the first few hours to facilitate decisions regarding therapeutic treatment. Treatment may include administration of DTPA, either intravenous or nebulized, plus the inhalation of a lung irritant to induce coughing and hopefully speed up clearance from the upper respiratory tract. However, it should be kept in mind that urine data will need to be corrected when DTPA has been administered as will usually be the case for the more serious intakes.

INTERIM EVALUATION - INHALATION CASE

An interim evaluation is more complicated and utilizes concepts presented in the new ICRP lung model.⁽⁹⁾ The new ICRP Lung Model depends upon dust sampling data and describes the deposition in three lung compartments: the nasopharynx (N-P), the tracheobronchial tree (T-B) and the pulmonary (P). The clearance rates from the T-B and N-P are rapid with the GI tract being the primary pathway. The clearance pathways from the pulmonary are more involved. Forty percent of the original pulmonary deposition clears to the GI tract with a one-day half life clearance rate. The slower clearance includes 40% of the GI tract, 15% of the lymph nodes, and 5% of the blood, each with a 500-day half life clearance rate. These clearance rates and pathways suggest a number of methods for calculating depositions incurred by inhalation which are used in making our estimates from fecal and lung counter data.

For all incidents, at least one fecal sample is scheduled, preferably on the second day post intake, since fecal excretion data of inhaled materials are more likely to peak at that time. This single result is used primarily as an indication of the need for additional followup. For more serious intakes, a series of fecal samples are scheduled. In addition to plutonium

analysis, the ^{241}Am content in feces is determined by gamma ray spectrometry, since these results are available shortly after submission of the sample. The Pu content is then estimated based on the $\text{Pu}\alpha/\text{Am}\alpha$ ratio. Particle size data are obtained using techniques described by Andersen⁽¹⁰⁾. Usually nasal smears, air samples and smears of skin contamination or nearly flat surfaces are used for determining median particle sizes. If the median particle size is unknown, a standard size of $4\mu\text{m}$ is used for evaluation purposes. This $4\mu\text{m}$ size is based on the average median of some 60 inhalation cases at Hanford.

An in-vivo chest examination is scheduled within 4 hours of intake. If this is positive, then additional lung examinations are made during the first week to check the early clearance rates; however, more meaningful data are obtained at day 7 and later when the rate of clearance is much slower.

These data are used either directly or indirectly in making interim and formal evaluations of depositions. Interim evaluations are made primarily using early fecal data (excreted during the first week), in-vivo examination data combined with the new lung model concepts, and urine excretion data using a fit to the Langham model.

Figure 1 shows the relationship between systemic or pulmonary burdens and early fecal data for varying particle sizes. Estimates may be made at one week post intake using this relationship, the ^{241}Am data obtained from gamma analysis, and the appropriate $\text{Pu}\alpha/\text{Am}\alpha$ ratio. The same relation is used with final Pu data. Delayed fecal data may also be used as an aid in evaluations. Based on lung model concepts for plutonium oxide, a daily excretion rate of 1 pCi per day at times > 10 days post intake is indicative of a pulmonary burden of 1 nCi.⁽¹¹⁾ An estimate based on delayed fecal data can be

FIGURE 1

Burden (Initial Pulmonary or Systemic) for one nCi Excreted in the Feces during the first Week Following Intake as a Function of Particle Size - Based on the ICRP Task Group II Lung Model - for Plutonium Oxide Inhalation Only

greatly biased by a very small intake just prior to sample collection; therefore, it is advisable to collect samples following absences from plutonium exposures of at least two days. Excretion from systemic pathways, particularly for cases involving prior depositions may also bias estimates by this method.

Estimates of the current lung deposition, the initial pulmonary deposition and the systemic burden are made from a series of in-vivo examination data starting on about day 7 following intake. Extrapolation of these in-vivo data to time of intake yields an estimate which is 0.6 of the initial pulmonary burden as shown in Figure 2. This estimate is relatively independent of the particle size of the contaminant; however, in our case it is for ^{241}Am only and must be corrected using the appropriate $\text{Pu}\alpha/\text{Am}\alpha$ ratio. Using the lung model concepts, the systemic deposition is estimated from the evaluated initial pulmonary burden. This relationship is shown on Figure 3. Sufficient urinary excretion data are normally available at this time for making a more reasonable estimate of the deposition of any initially soluble plutonium using the Langham model. These interim evaluation techniques normally provide reasonable estimates of the deposition to be expected and may be used to assist in decision making concerning future actions for the case (i.e. work restrictions, continue treatment, advise management, etc.)

FORMAL EVALUATION - INHALATION CASES

Although early survey data, lung counter examination data, fecal data, nasal smear data, etc. are used for making early and interim evaluations, the final, formal evaluation of the systemic deposition of plutonium is based on urine data. The systemic deposition of initially soluble plutonium is evaluated using early urine data and the Langham model⁽¹²⁾

FIGURE 2

FIGURE 3

Systemic Burden for an Initial Pulmonary Deposition of 1 nCi as a Function of Particle Size - Based on the ICRP Task Group II Lung Model - for Plutonium Oxide Inhalation Only

while the deposition of the initially insoluble plutonium is evaluated from delayed urine data and the Healy model.⁽¹³⁾ Sample curves are shown on Figure 4. Evaluation following treatment with DTPA is always more difficult. The rate of excretion per day decreases more rapidly than expected by the Langham model for 100 days or more post treatment but will eventually stabilize to follow the expected rate of decrease. Our normal practice, for cases involving DTPA treatment, is to apply urine data at ~100 days or more to the Langham model. When the urine excretion stabilizes, estimates of deposition at different time periods are similar. When this point in time is reached, we apply the best fit of the Langham curve to these data to estimate the soluble systemic deposition.

The deposition in the lung is evaluated using techniques previously described for interim evaluations. Appropriate corrections for ²⁴¹Am growth must be applied to delayed lung count data.

It is anticipated that these same evaluation techniques, applied to cases involving intakes of transplutonium elements, will provide reasonable estimates of lung burdens and some idea of the systemic burden as well. Adequate models for the formal evaluation of systemic burdens, using bioassay data, are not generally available for many of the transplutonium elements. One of the members of this panel will be discussing the status of such models later on this evening.

Urinary Excretion Curves for Systemic
 Deposition of 1 nCi of Plutonium

Figure 4

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