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PROCEEDINGS SERIES

DIAGNOSIS AND TREATMENT OF INCORPORATED RADIONUCLIDES

PROCEEDINGS OF AN INTERNATIONAL SEMINAR ON
DIAGNOSIS AND TREATMENT OF INCORPORATED RADIONUCLIDES
ORGANIZED BY
THE INTERNATIONAL ATOMIC ENERGY AGENCY AND
THE WORLD HEALTH ORGANIZATION
AND HELD IN VIENNA, 8 - 12 DECEMBER 1975

IAEA-SR-6/30

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INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 1976

FILE BARCODE
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Abstract

CONSIDERATIONS IN THE ASSESSMENT OF PLUTONIUM DEPOSITION IN MAN.

Data from human cases of plutonium inhalation are used to illustrate several important problems in the current methods of estimating plutonium body burdens. Individuals exposed to $^{239}PuO_2$ particles in a highly insoluble matrix showed an unusually slow rising urinary excretion curve over 300 - 400 days. In-vivo chest counts during the first 6 months estimated lung burdens to be 10 - 30 nCi, but urinary excretion methods calculate residual systemic body burdens of 50 - 100 nCi at 1200 days after exposure. Current assumptions used in the in-vivo calibration do not consider possible lung distribution of particulates soon after exposure that could alter the interpretation significantly. Tissue analysis of a lung from another case after recent inhalation exposure shows a significantly lesser concentration of plutonium in the subpleural region – the principal region of plutonium measurement by in-vivo chest counting – as compared to distributions found years after exposure. Tissue analyses indicate that urinary excretion estimates of body burden over the long term tend to err on the high side up to a factor of 5 or more. This procedure serves well for the purpose of protection of workers, but high estimates can lead to unnecessary job reassignments for the individual worker and can cause misleading conclusions when such data are used uncritically for reference in health effects studies. Additional research is needed to improve urinary excretion data analysis to reflect newer information derived from tissue data and to seek further understanding of the sources of variation in in-vivo counting technology so it can be used with greater confidence.

INTRODUCTION

Measurement of the quantity of plutonium in exposed personnel is an important tool for those concerned with the safety of plutonium workers. In addition to gauging the effectiveness of health physics controls, the data of persons exposed years ago can also help reassess the validity of the basic occupational exposure limits. These were derived 25-30 years ago by a mixture of human experience with external radiation, internal radium exposures, and comparative animal experiments. It is important that our continuing experiences be measured and recorded accurately to help confirm or deny these early conclusions.

In this paper we examine three of the methods in use at the Los Alamos Scientific Laboratory to determine such body burdens. These are: (1) the measurement of plutonium in the urine; (2) direct measurement of the low energy photon emissions from the plutonium deposited in the lung; and (3) the determination of tissue burdens by autopsy (or in some cases biopsy). Several recent cases which provide specific data relating to such measurements are used as illustration.

URINARY EXCRETION

The measurement of plutonium in urine samples has been used as an index of exposure and potential accumulation in the body since the days of the Manhattan District. The classical study of

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Langham, [1,2] determining urinary excretion rates following ^{239}Pu citrate injections of 16 individuals, judged to have near-term fatal disease, provided a quantitative basis for such interpretations and has served as a foundation for many programs.

Although many of the uncertainties in the use of this method are recognized, there are no methods for determining the influence of these uncertainties in any one case. Some of the more important problems are:

1. The plutonium was administered intravenously as the citrate. In using Langham's data, one must accept that the plutonium absorption, deposition, and excretion in the occupational case will be like that of the injectees.

2. The excretion pattern was followed for only limited periods of time in these individuals and was extended to about 1600 days by use of data from occupationally exposed individuals. While this time period is adequate for following new exposures, extrapolations to long-term excretion patterns become uncertain when one is considering the body burdens of individuals exposed 10-30 years ago.

3. Examination of the initial data used by Langham indicates that there are significant individual variations even among people exposed in essentially the same manner. Use of an average excretion function does not reduce this uncertainty in the individual case.

4. The urinary excretion reflects only the plutonium that circulates through the kidneys and is not influenced by deposits unavailable to the bloodstream. A deposit in a region with slow transfer to the blood will result in a gradual increase in the urinary excretion over some period of time until the available plutonium is redistributed. Attempts to use the shape of the curve to infer the rate of transfer and a measure of total deposition have been made by Healy [3] and Beach and Dolphin [4] but such approximations must be regarded as somewhat speculative and uncertain.

In spite of these uncertainties, the method is used worldwide and, in nearly every case, the results are converted to quantitative estimates of body burden.

Some of these problems can be illustrated with data obtained following the exposure of nine persons to ^{238}Pu in 1971. They were disassembling a ^{238}Pu heat source for diagnostic measurements when one of the sleeves on their hot cell manipulator developed a split and permitted airborne contamination. The material in this source is formulated in a molybdenum-ceramic matrix to make it as insoluble as possible and undoubtedly accounts for the unusual excretion pattern in these cases. Data from two of the higher exposed individuals are presented here as examples. Initial (within the first two days) measurements of the chest burden (next section) showed high values but subsequent measurements were lower. Urine samples taken during the first few days after the exposure were low (<0.1 dis/min) indicating little uptake by the systemic circulation. Nasal swabs at the time of the incident showed several thousand counts per minute and suggested possible significant exposure, so the sampling program was continued. Fecal samples showed significant activity by direct counting techniques, but insolubility even in strong acids heated overnight created serious analytical problems with these samples. The results of urinalyses for two individuals are shown in Figures 1 and 2. As can be seen, there was a steady increase over the first 300-400 days followed by a leveling off. In gross appearance, this is the increase postulated in the models by Healy [3] and Beach and Dolphin [4]

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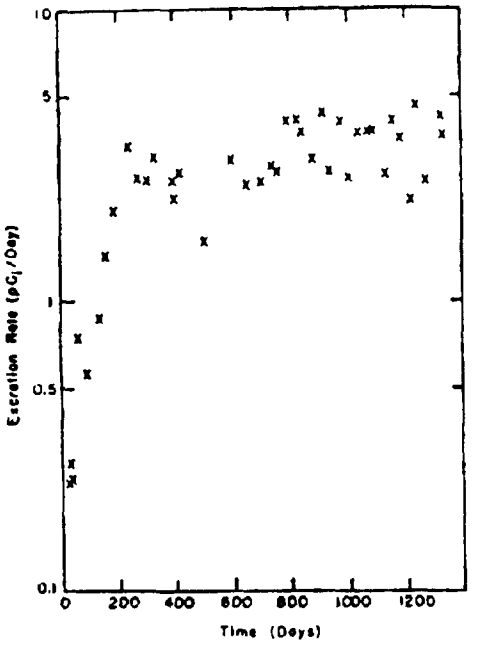

FIG. 1. Urinary excretion of ^{238}Pu by subject A following incident.

for movement from the lung into the body. These models assume a constant solubility or rate of transfer to the bloodstream and kidney where the plutonium is then excreted according to the pattern prescribed by Langham. They predict a much more rapid increase in urinary excretion with the maximum occurring at an earlier time.

The quantity of ^{238}Pu transferred to the bloodstream, and now considered as systemic body burden, has been calculated by the PUQFUA Code [5] which uses Langham's equations. This procedure can have wide uncertainties if samples are infrequent, but for these cases where sampling was done every 30-90 days, it should provide a reasonable estimate within the uncertainties noted earlier. Estimated quantities transferred in the first 1200 days are 100 nCi for Subject A and 50 nCi for Subject B.

IN-VIVO COUNTING

It is only in the past decade that equipment has been developed to permit estimation of quantities of plutonium in the lung (or more accurately, in the chest) by external measurements on the individual. In some cases, the more energetic photons (~60 keV) from ^{241}Am are used, but this depends upon prior information as to

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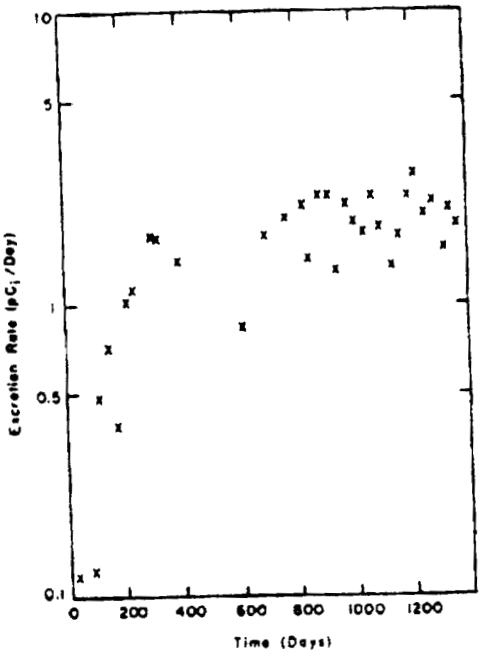

FIG. 2. Urinary excretion of ^{238}Pu by subject 8 following incident.

the quantity of ^{241}Am in the material inhaled and upon an assumption that the americium remains associated with the plutonium. Data on the metabolism of ^{241}Am in animals [6] makes the latter assumption questionable although the degree of uncertainty is not defined.

A full description of the LASL instrumentation and procedures is given elsewhere.[7] Essentially the procedure depends upon the measurement of the ~17 keV L x-rays (uranium) in 4.6% of the disintegrations. Two 12.7-cm-diameter Phoswich detectors are placed on the chest of a prone individual and measurements are normally made for 30 minutes. The difference in the pulse shape in the two crystals [NaI(Tl) and CsI(Tl)] making up the Phoswich permit discrimination of the energy band of interest from the normal Compton continuum and hence provide much reduced room backgrounds.

Because of the low energy and frequency of the photons which are detected, the sensitivity and accuracy of the procedure are not as good as could be desired and the interpretation of the measurements must include a number of correction factors, described below, each of which contains its own source of uncertainty and bias. The thickness of the individual's chest wall, measured by ultrasonic technique, determines a correction factor to account for differential absorption compared to that of the calibration

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phantom. The influence of higher energy gammas emitted by other isotopes in the body, such as the natural ^{40}K, is corrected by measuring a portion of the spectrum in the higher energy region and extrapolating to the contribution in the region of interest. The presence of even relatively small quantities of other isotopes, such as ^{137}Cs, can result in a correction of such magnitude that the sensitivity for plutonium determination is greatly decreased, or conversely, low indications of the presence of plutonium can result. Since all isotopes of plutonium emit the x-rays being measured, but with differing frequencies per disintegration, any absolute determination requires knowledge of the involved isotope(s) from other sources. Americium-241, a common contaminant of plutonium mixtures, also emits x-rays in the plutonium region. It is therefore necessary to determine the ^{241}Am content by a measurement of the 60-keV gamma rays and subtract the estimated contribution from this isotope from the reading in the plutonium x-ray region.

Once these corrections have been made to the measurement, the result is corrected finally to provide the estimated quantity of plutonium in the lung. This is done by a calibration using a phantom of known chest wall thickness and with the plutonium distributed uniformly through the lung. This may well be one of the more critical assumptions since it is known that the plutonium is not uniformly distributed. This is a particularly important consideration for plutonium chest counters because the detector sees, preferentially, activity from the outer ~2 cm of the lung due to the high absorption of the ~17 keV x-rays in tissue. Thus any mechanism which causes redistribution in this subpleural region, such as may occur in the early time period after exposure, can cause a change in the response of the detector.

Sensitivities of the LASL equipment, using a 30-minute measurement, are such that one can detect about 9 nCi of ^{239}Pu, 4 nCi of ^{238}Pu, and 0.2 nCi of ^{241}Am. The minimum detectable amount (MDA) is defined as a response equal to three standard deviations of the background.

A common operational problem with such devices is the possible presence of contamination on the skin of the individual being measured. Because of the large absorption in the chest wall and the geometry, a small amount of plutonium on the skin will give a greatly exaggerated response if interpreted as a lung burden. This problem is particularly noticeable when measurements are made following an incident where the individual may retain low levels of skin contamination even after extensive decontamination. For this reason positive values at such a time are regarded as suspect to be followed up by later checks. With experience it is often possible to recognize contamination that is essentially all on the surface by the shape of the spectrum.

The incident mentioned in the previous section involving ^{238}Pu provides an excellent illustration of some of the problems, as well as potential advantages, of this system. Measurements were made on these individuals for several days following the incident and periodically thereafter for about 215 days. These individuals already had small burdens of ^{137}Cs as a result of working with fission products prior to the plutonium exposure. At the end of 215 days they were all involved in another minor ^{137}Cs exposure which, while small compared to maximum permissible limits, was sufficient to mask the ^{238}Pu signal.

Data for these individuals are given in Figures 3 and 4. The results during the first several weeks varied beyond the estimated

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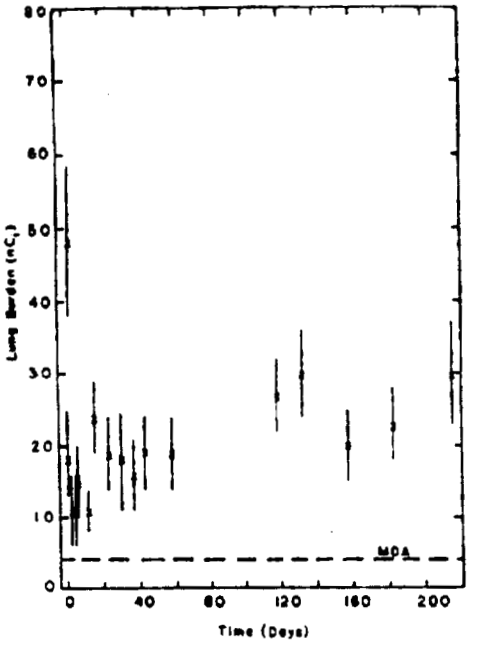

FIG.3. Chest measurements on subject A. (MDA = minimum detectable amount.)

limits of error and then settled into a reasonable pattern. We have not identified definitely the cause of the variability nor the reason for the apparent increase after about 100 days. There is a possibility that the instrumentation shifted calibration for the points past 100 days. The early variability does not seem to correspond with the fast clearance phase of particles from the bronchial tree, although this appeared a possible interpretation for the first three or four counts. Since detection of activity for plutonium is preferential for the outer 2 cm of lung tissue, it is possible, perhaps likely, that clearance of the bronchi will have little influence on the count. Alternatively, we have speculated that the plutonium particles may be translocating in the lung over several weeks to the subpleural region where they are more visible to the detector. Some evidence for this possibility will be discussed later.

As was noted earlier, interpretation of the body burden on these individuals from the urine data by use of the FUGFUA Code indicated that after 1200 days the systemic body burdens were estimated as 100 and 50 nCi. This material must have been transferred from the lung to the bloodstream as a result of this incident since the individuals did not work with plutonium after this exposure, yet the initial indication of quantity in the lung was only on the order of 10-30 nCi. We believe that this discrepancy may be due to the nonuniform distribution with heavier deposits in areas which are not "seen" by the detectors.

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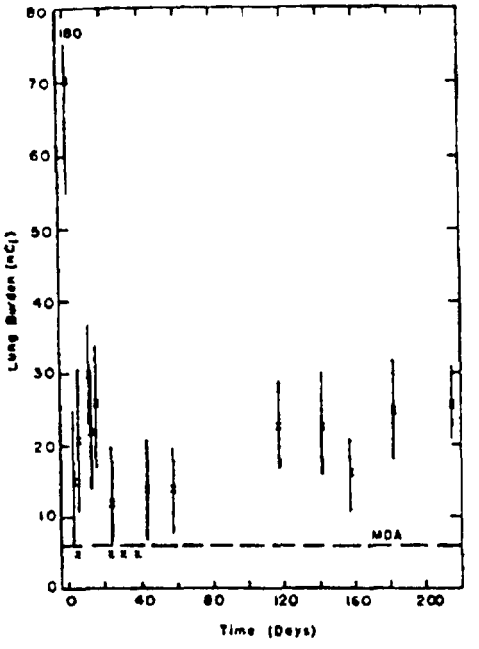

FIG.4. Chest measurements on subject B. (MDA – minimum detectable amount.)

AUTOPSY RESULTS

In view of the uncertainties in the urine evaluations of plutonium deposition in the body, a program of collecting tissue specimens from autopsies performed at the local hospital was started in the late 50's. This provided an ultimate check on the accumulation of plutonium by those working in the laboratory as well as background values from individuals who had lived in the area but had not been employed in the laboratory. In recent years, the program has been expanded to include tissue samples from the general population in other areas and the analytical techniques have improved so that the minute quantities available in individuals as a result of accumulation from fallout can be measured. Such data, over a wide range of exposures, are invaluable in determining the actual distribution of plutonium in the body from a wide variety of exposure situations. The procedures and the data obtained on this autopsy program have been published [8] and several additional papers are in preparation.

Information on the distribution of plutonium in lung can be gained from autopsy data and is valuable to study potential calibration problems for in-vivo measurements.

Complete lungs were obtained from a female who was exposed to plutonium less than a month before her accidental death and a total lung content of about 4 nCi is estimated by tissue analysis. We also analyzed lungs of a male accident victim who was employed in

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TABLE I. COMPARISON OF URINE AND TISSUE ANALYSIS ESTIMATES

Time From First Exposure to Death (years)	Number of Urine Samples	Body Burden (nCi)			Ratios	
		Urine Estimate	Tissue Estimate Systemic	Tissue Estimate Lung	Urine / Systemic	Urine / Total
13	71	26	18	3.0	1.4	1.2
15	49	12	3.5[a]	1.9	3.4[a]	2.2[a]
15	58	10	4.2	4.9	2.4	1.1
11	26	2.4	0.02	0.03	120	48
13	13	2.5	0.25	0.01	10	9.6
8	23	1.8	0.4	0.01	4.5	4.4
8	18	2.2	0.9	0.25	2.4	1.9
19	40	5.4	0.7	0.35	7.7	5.1
29	42	7.2	0.75	0.24	9.6	7.3
25	149	31	9.3	37	3.3	0.67

[a]No bone sample was obtained. This estimate does not include the skeleton.

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plutonium work for 26 years; his total lung content was about 36 nCi, estimated by tissue analysis. In each of these cases, the superior lobe of one lung was divided into 3-4 mm-wide tissue sections made horizontally, starting at the top. Individual sections were then divided into pleural (~1 cm) and parenchymal regions to obtain some indication of plutonium distribution. In the recent exposure case, there was little difference in the concentrations between the two regions. There was, however, an apparent increase in the plutonium concentration in both regions as one proceeds inferiorly from the apex of the lung. In this case, the parenchymal sections were also subdivided into a 1-2 cm outer region, 2-4 cm intermediate region, and an inner section containing the major bronchi in those sections close to the lung center where these structures were apparent. There was little difference between the outer two regions of parenchyma but the concentration in the region containing the bronchi appeared lower. In the lung from the individual with the long exposure time, the concentration in the pleura in the region from 12-24 mm from the apex was some 10-35 times that in the parenchyma. Unfortunately, the analytical results for the sections between this region and the middle levels of the lung did not permit an estimate of chemical recovery and, consequently, there are uncertainties as to their absolute value. They do indicate a decreased concentration in the pleural sections toward the middle levels of lung where concentrations are approximately those of the parenchyma. These results are similar to those of Nelson [9] who reported concentrations 59 times greater in the pleural and subpleural tissues than in the parenchyma in a former worker with 25 years of potential exposure.

While we cannot draw firm conclusions from only two cases, these data would suggest that the plutonium is fairly uniformly deposited throughout the lung at reasonably short times following exposure. As time passes, there seems to be concentration in some subpleural regions either by migration toward the pleura or preferential elimination from the parenchyma or a combination of both. More data is needed to confirm these observations.

The accumulation of plutonium in the lymph nodes has been well documented. [10] In our long-term exposure case, the tracheobronchial lymph nodes had concentrations about 40 times that of the lung as would be expected from past data, but in our acute exposure case the lymph node concentration was about one-fifth that of the lung as a whole, indicating that concentration in the nodes had barely started. This observation indicates that the kinetics of accumulation by the lymph node in humans may be similar to that described by Thomas [11] for the dog.

An estimate of the quantity of plutonium in the body by tissue analysis has been used to compare with antemortum estimates by other techniques. Such an estimate does involve considerable uncertainty, particularly in obtaining the total skeleton deposit. Autopsy samples of bone are only on the order of a few percent of the skeleton and calculation of total deposition requires large scaling factors. Furthermore, wide variations in the plutonium content of individual bones are possible. [12] Table I indicates the comparison between urine estimates and tissue estimates for those cases which had a sufficient plutonium in the body to be measurable and for which a reasonable number of urine samples were obtained.

The variation in the ratio between the two estimates (urine vs tissue) is similar to those reported by others [13,14] with the urine estimate being higher than that found at autopsy in almost

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every case. The value of 120 (line 4, Table I) is out of line with the others. However, investigation of the urine results indicates that the high urine estimate was entirely due to two samples in the 1950-60 era. Samples taken prior to and following the high readers were below the detection limit. Due to the rapid drop-off in time with Langham's equations one cannot assure that this was not a true exposure and it is carried in the records as such. However, in retrospect, it appears that the two samples in question may be due to external contamination. This again illustrates the difficulty of interpretation of results of this nature and the uncertainties in the answer.

In Table I, we have distinguished between the amount in the lung and lymph nodes and the so-called "systemic" burden, the quantity in other organs such as bone, kidney, liver, and muscle. This is customary since it is assumed that the Langham equations only reflect the quantity actually entering the bloodstream and, for this purpose, the lung and lymph nodes are considered to be outside the circulatory system. We are beginning now to question this concept as it applies to the long-term deposit in the lung since this material is probably no longer in the alveoli but is incorporated into the tissue structure. Since the long-term elimination in the Langham equation represents bone remodeling and elimination from other tissues, it appears reasonable that a similar elimination or exchange will occur with the plutonium embedded among the cells of the lung or in drainage from the lymphatic system. These locations also permit contact of plutonium with tissue fluids and would attain a transfer rate to the systemic circulation. The influence of this on the excretion curve may be as important in affecting the urinary results as the movement from the liver or bone. Only additional experimental data will permit verification and quantification of this hypothesis, but inclusion of long-term lung content in the body burden gives a better correlation with urinary excretion estimates.

DISCUSSION

The cases described in this paper illustrate a number of important and unresolved questions in attempting to measure the quantity of plutonium in the bodies of workers. The lung counting methods are still in process of development; these estimates are based on calibration with uniform distribution in the lung but the results are sensitive to nonuniform distribution effects that are largely unknown now. If our speculations as to redistribution with time leading to a higher concentration in the lung periphery are correct, this would lead to an increase in the response of the lung detector for a given quantity in the lung since the photons originating from this material on the surface of the lung will be less subject to absorption. Note that this does not necessarily mean an increase in response for a given case since elimination is occurring at the same time. If this is true, it may provide a basic limitation on the accuracy of such measurements unless further study can provide reasonable estimates of the distribution effect with time. In the cases of ^{238}Pu inhalation described, it was not possible to identify changes in the lung burden with certainty by chest counting due to variances in the data during the first 215 days. However, the subsequent analysis of urine data would indicate the transfer to the body of some 3-5 times as much

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plutonium over a period of 1200 days as the lung counter indicated as the initial deposit. Since the fraction transferred to the bloodstream was undoubtedly only a portion, and perhaps only a small portion, of that initially deposited in the lung, the response of the counter was even lower. This is puzzling to us since we cannot visualize a plausible distribution which would account for such a high factor. On a physical basis, the use of ^{241}Am instead of ^{239}Pu would appear to be advantageous in the early estimates following inhalation. In these ^{238}Pu cases, there was no measurable americium content for such use.

Another lesson which we have learned from these exposures is that one cannot rely on early urine results to even give an indication of a potentially serious exposure and that follow-up urine samples are necessary. We realize that this was an unusual material not encountered frequently, but this may also be true of reactor fuels to be used in the future. It was not until some months had passed that the potential intake by these individuals became apparent.

It is probably not surprising that estimates by urinalysis appear to be somewhat high compared to tissue analysis. They were designed for health protection purposes so assumptions that need to be made tend to be conservative and result in higher (safer) estimates. A high estimate can be less desirable if applied to problems other than radiological control purposes. Two such applications are assessing individual risk for continued work with plutonium or other radioactive materials and assessing health effects in workers by epidemiological methods for possible dose-effect relationships.

There are strong motivations for most employers to remove an individual from work with plutonium if the estimate of body burden exceeds a given value. In some instances the value chosen is less than the maximum permissible body burden. A false high body burden estimate may be used in an arbitrary fashion to affect an individual's career and future without proper weight being given to these uncertainties. It is not clear that personal plutonium risk should supersede all other considerations in the decision. Is the situation much different than if we require a truck driver to stop his driving because we recognize his risk is greater than some other vocation? Job displacement represents a drastic step for the individual, particularly if he has training and experience in the job for a period of time. We believe that this should be a matter for decision by the individual with the counsel of medical and management personnel and taking into account his age, state of health, potential problems (including risk) on an alternate job, and the certainty of his individual exposure estimate. The maximum permissible body burden is invaluable for health physics control purposes in the plant, but should be used as only one comparative piece of information in the complex decision of individual work assignment. For this purpose, body burden calculations should strive for the best possible estimate, rather than a conservative (high) estimate.

Improved techniques for body burden assessment are also needed for all studies used to assess the validity of the maximum permissible limits. If we are to understand better the risk involved in the use of plutonium, we need long term data on the health of individuals exposed to it as well as good estimates of the exposure. Estimates that are biased systematically on the high side could lead to false conclusions on the dose/effect relationships. Such studies need to be done to answer the questions of risk at various

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exposure levels. Only continuous surveillance of exposed populations correlated with adequate estimates of the body burdens will permit us to assess the validity or need for revision of the maximum permissible limits.

These experiences and results convince us that more research and study are needed in instrumentation development, measurement technology, and analysis of data on plutonium in man. Our knowledge of plutonium burdens in man continues to rest on a base that is not sufficiently secure to relax or to feel comfortable.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the work of Philip Dean, who developed the plutonium chest counting technology and data analysis at Los Alamos. The chest counting procedures presented in this paper were performed by Harold Ide. Tissue analyses for plutonium were performed by William Moss.

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DISCUSSION

P.M. LEGUEN: Three years ago at Fontenay-aux-Roses we observed a contamination incident of the same order as the one you have referred to, involving inhalation of plutonium-238. This incident concerned 17 workers and two of them were chosen for follow-up because of their initial lung burden as estimated from X-ray spectrometry and from the activity of faeces and urine samples. The estimated lung burden was of the order of 15 nCi for one of the patients and 10 nCi for the other. Over a period of two years 24-hour urine samples and 3-day faeces samples were measured radiochemically twice a month, a practice now carried out on a routine basis in the medical departments of the CEA. The urine activity was always slight, of the order of 1-4 pCi, and practically without variation, but the activity in the stools – initially of the order of 1 to 3 nCi – decreased steadily over the entire period of observation.

On the basis of this curve for excretion of the contaminant in the faeces the hygienist was able to estimate the dose to the lung. This dose was only a factor of 2 higher than that obtained using X-ray spectrometry in the first week.

If we had merely taken the urinary excretion curve, we should have found it difficult to estimate the dose received.

W.J. BAIR: It is of interest to note that the higher concentrations of plutonium which you find in the pleural and sub-pleural regions of human lungs, as distinct from the lung parenchyma, agree with observations in experimental animals such as rodents and dogs. Have you done any microdosimetry studies on the sections of human lungs you described? This would be a useful study in view of current controversy about the distribution of the alpha-radiation dose from inhaled plutonium in the lungs and speculations about the relative radiocarcinogenic sensitivities of the human bronchial epithelium and the bronchiolar and alveolar epithelia.

G. VOELZ: We have done radioautographic studies on the long-term exposure autopsy case discussed in my paper. Using a track-counting technique we hope to estimate the particle size of the plutonium deposited in the lung. So far, counts on about 120 such particles have been analysed and the results show only small particles in the lung, less than one micrometre. We are not convinced that the precision of such studies is adequate for microdosimetry calculations. Another complication is the probability that some movement of particles occurs with time. We have not found any technique that is more precise, and would welcome any ideas on how to make such measurements.

G.B. SCHOFIELD: I strongly endorse your plea for a reassessment of the methods used at present to estimate plutonium body content. During the past 10 years we have conducted autopsy examinations on 18 plutonium

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workers at Windscale and the findings reflect those reported by you and others in the United States in that the in-vivo urinary estimate of body content is usually between 2 and 10 times higher than the direct estimate from autopsy material. A more realistic method of in-vivo assessment is therefore needed, and this would result in less exacting restrictions on workers in the nuclear industry.

ASSE
BURI

R.C. SH
T. SURE
D.P. BH
Bhabha
Bombay

Abstract

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