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Part II

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RADIOLOGICAL AND ENVIRONMENTAL RESEARCH DIVISION ANNUAL REPORT

Center for Human Radiobiology

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FOREWORD

This Annual Report contains a summary of case notes on the six new cases of mastoid carcinoma which have been diagnosed in radium patients since 1969, when the Center for Human Radiobiology was established. It is noteworthy that no new cases of bone sarcoma were diagnosed in this population during the same period. There are now 54 cases of bone sarcoma and 27 sinus or mastoid carcinoma cases known among the 1568 persons whose body burdens of radium had been measured by the end of 1973. Exposure data for the measured radium cases are given in Appendix A at the back of this Report, and the cases of bone sarcoma and head carcinoma are listed in Appendix B. These tumors are considered to be radium-induced malignancies because of the strong correlation with radium burden and because the total number expected for the 1568 patients on the basis of natural incidence rates would be less than one.

It should be noted that several papers in this Report deal with studies of plutonium in humans or with methods for measuring plutonium. Rates of excretion of plutonium are reported for three persons who received injections of plutonium in 1945–47, and another paper describes the microscopic distribution of plutonium in bone from a patient who died with Cushing's syndrome 17 months after injection of plutonium in 1945.

During the past year considerable effort was given to explicit formulation of the epidemiological plan for the radium studies of the Center. Of particular concern was, and is, the problem of sampling bias due to incomplete follow-up of many persons who have been identified by name as exposed to intake of radium (cf. p. 1 of this Annual Report). This includes persons whom we have not been able to locate because of insufficient personal data and other persons whose radium burdens have not been measured. Many of the latter died before the Center was established in 1969. A detailed Plan of the Radium Project was prepared by CHR staff members in May 1974, and it is now being reviewed by several epidemiologists and radiobiologists.

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Center for Human Radiobiology

THE MICROSCOPIC DISTRIBUTION AND INITIAL DEPOSITION OF ^{239}Pu IN HUMAN BONE

Robert A. Schlenker and Billie G. Oltman

The initial concentrations of ^{239}Pu on endosteal surfaces of compact and cancellous bone from a person who had received a ^{239}Pu injection were determined by quantitative autoradiography. The concentrations were higher in cancellous bone than in compact bone, and concentrations in the axial skeleton were greater than in the appendicular skeleton. The values ranged from 0.4 to 4.6 pCi/cm^2, and the average over the entire endosteal surface of the skeleton was 3.0 pCi/cm^2. The bone surface to bone volume ratio was measured in a cervical vertebra and in a lumbar vertebra and was found to be 114 cm^2/cm^3 in the former and 101 cm^2/cm^3 in the latter, close to the value of 110 cm^2/cm^3 found for a normal adult male by Lloyd and Hodges. The total skeletal bone mass was estimated to be 2100 g and is considerably subnormal, probably because the subject suffered from Cushing's syndrome. The initial uptake of ^{239}Pu in the marrow-free skeleton was estimated to be 26% of the injected amount. The data were used, in conjunction with a model by Marshall and Lloyd, to predict that the $^{239}Pu/^{226}Ra$ RBE in man will be four times the $^{239}Pu/^{226}Ra$ RBE observed in the Utah beagles.

Introduction

In September, 1973, the Center for Human Radiobiology received the remains of a woman (case no. 40-010) who had been injected at age 18 with 0.3 μCi ^{239}Pu(IV) in citrate solution and who died 17 months later in April, 1974 with Cushing's syndrome (case HP-4, Refs. 1 and 2). We have used autoradiography to study the microscopic distribution of ^{239}Pu in bone. From the observed deposition, plus measurements of surface-to-volume ratio and bone mass, we have estimated the initial uptake in the marrow-free skeleton. In addition, we have used the initial surface concentrations and uptake to estimate the initial ratio of surface dose rate to uniform dose rate. This has been used in conjunction with a model proposed by Marshall and Lloyd to predict a value for the $^{239}Pu/^{226}Ra$ RBE in man relative to that in beagle.[3]

Methods

Bone samples were defatted, dried, embedded in methyl methacrylate, and cut into 100-μm sections. The sections were apposed to pieces of 300-μm

thick Lexan sheet and exposed to the neutron flux in the Argonne CP-5 reactor, to produce fission track autoradiographs.[4] Tracks were counted on the autoradiographs and used to provide estimates of initial surface concentrations.

Figure 1 shows the sites on the skeleton which were sampled. They included both cancellous and compact bone, and sites in the axial and appendicular skeleton.

Autoradiographic Appearance of ^{239}Pu in Human Bone

The distribution of ^{239}Pu in human bone shows features similar to those observed in the bone from other mammals:[5] Deposition on bone surfaces, displacement from the bone surfaces by apposition and resorption and deposition in the volume of growing bone. Figure 2 shows intense lines of fission tracks below the bone surface. These are believed to represent ^{239}Pu which was initially deposited on bone surface and then displaced from it by apposition. In addition, Figure 2 shows regions in which the bone volume contains a low fission track density and regions where the density is much higher. These are believed to be, respectively, preinjection bone, which was formed when there was no circulating ^{239}Pu and postinjection bone which incorporated circulating ^{239}Pu as it formed.

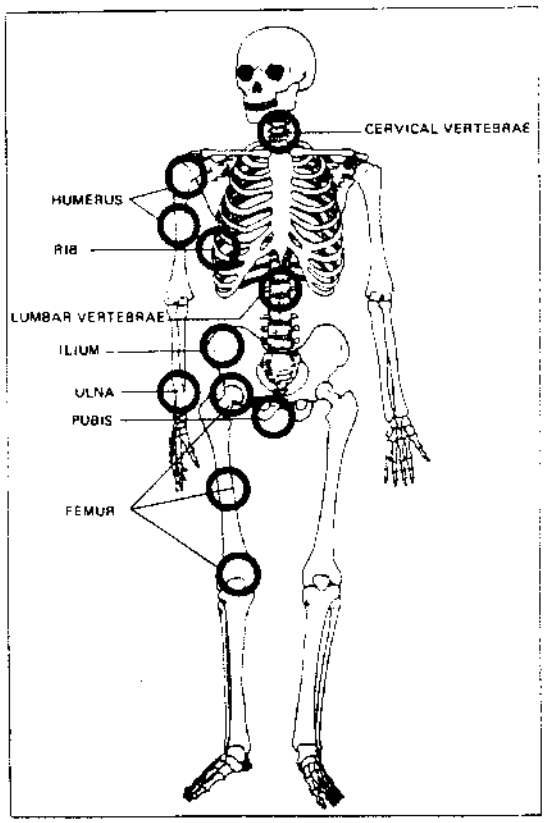
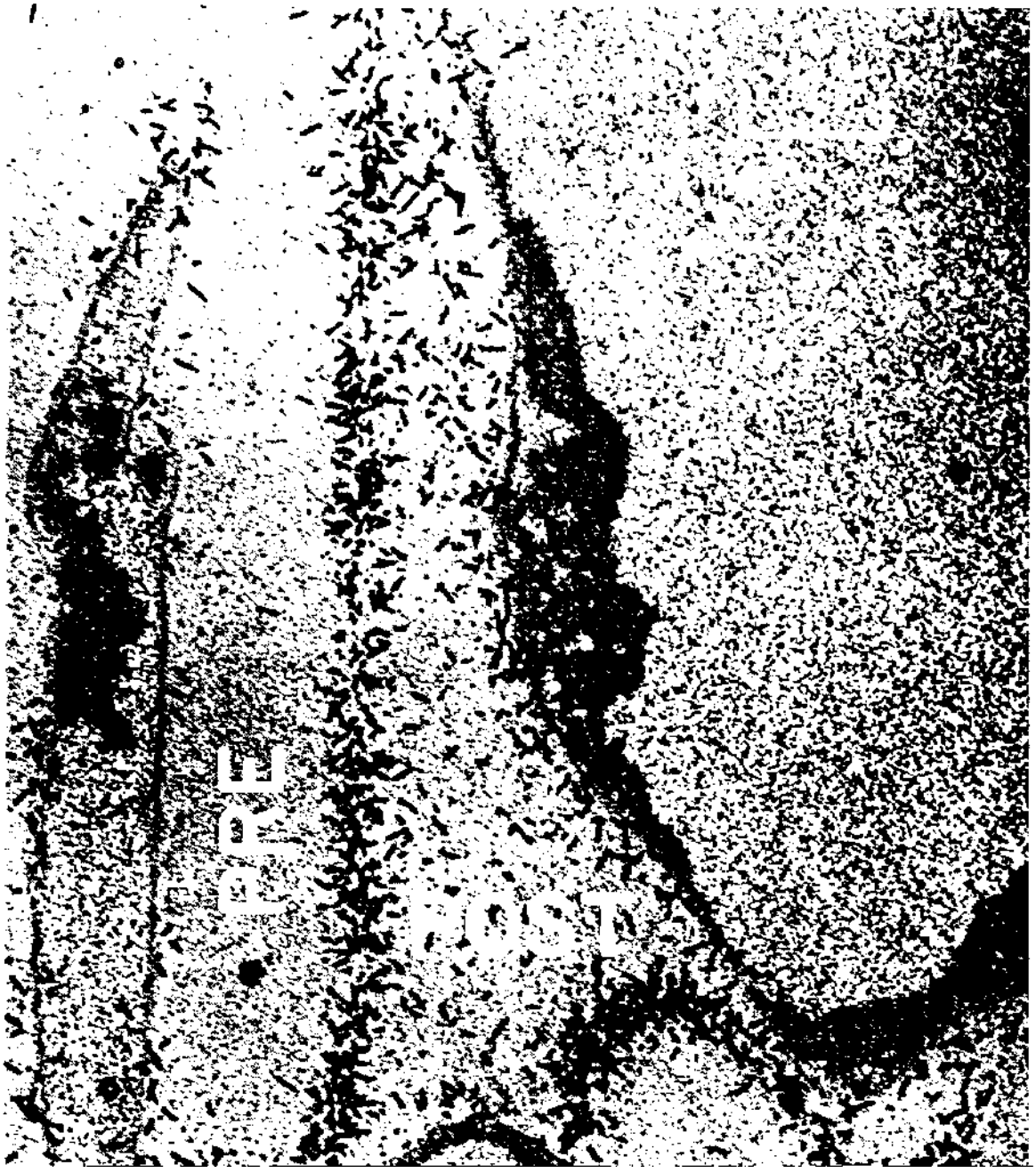


FIG. 1.--Skeletal sites sampled for ^{239}Pu autoradiography. (Neg. 149-6296K)

Initial Surface Concentration

In studies with beagle dogs at Utah, the initial concentration is observed to be greatest on the endosteal surfaces, less on the surfaces of Haversian canals and resorption cavities, and least on the periosteal surface.



In the sections of human bone which were studied, the endosteal surface concentration is extremely low. Much higher concentrations are observed in some burial lines (e.g., Figure 2), and we have assumed that these lines represent the original surface deposit displaced by apposition bone growth. The surface concentrations represented by these intense burial lines were determined by track counting. Attention was limited to those lines which have a width of 40 μm or less, since this is the width which would be produced by a deposit on a surface which was perpendicular to the plane of the autoradiograph.

The average surface concentrations determined from track counting are listed in Table 1 with the range of concentrations observed at each site.

From the table it is evident that the concentrations are higher in cancellous bone than in compact bone and that among the cancellous sites the concentrations are higher in the axial skeleton than in the appendicular skeleton. The ratio of the concentrations in the lumbar verbetra to the femoral metaphyses is 1.7, which compares with a value of 1.5 reported by Jee for beagle.[6]

A small sampling of the surface concentrations on Haversian canals was made and the concentrations were found to run about 10% of the average value for endosteal surfaces shown in Table 1. Occasional burial lines were noted in association with Haversian canals, but these did not appear to have higher concentrations than the surfaces. Low concentrations on existing endosteal surfaces plus high concentrations in burial

TABLE 1. Initial Surface Concentrations of Plutonium in Human Bone

Bone type	Site	Average, pCi/cm^2	Range, pCi/cm^2
Cancellous	Cervical vertebra	4.6	2.4 — 7.9
	Lumbar vertebra	4.1	1.7 — 7.0
	Rib	3.8	1.8 — 5.8
	Iliac crest	3.4	2.3 — 6.0
	Pubis	2.9	1.6 — 4.8
	Humerus	2.6	0.8 — 4.3
	Femur	2.3	0.8 — 4.5
	Ulna	1.1	0.7 — 2.0
Compact	Humerus	1.8	1.0 — 2.6
	Femur	0.4	0.3 — 0.6
Average over entire endosteal surface		3.0	

lines suggest a rapid rate of remodeling for endosteal surfaces, which in turn suggests that Haversian canal surfaces have remodeled rapidly as well. Thus, the concentrations observed probably do not reflect initial uptake. In beagle femur, the initial Haversian canal concentrations are 40% of the trabecular surface concentrations.[6] If the same were true in man, the Haversian canals would have a concentration about 1/3 of the average for endosteal surfaces shown in Table 1.

Initial Skeletal Uptake

The initial skeletal uptake can be estimated from the surface concentration data as

$$U = C_E A_E + C_H A_H + C_P A_P ,$$

where C_E, C_H, and C_P are average initial concentrations on the endosteal, Haversian canal, and periosteal surfaces; A_E, A_H, and A_P are the areas of the endosteal, Haversian canal, and periosteal surfaces. Concentration data have not been collected for the periosteal surface. However, the concentrations are no higher than for Haversian canals, and the periosteal surface area is small compared to the areas of the other surfaces so that neglect of the last product in the equation introduces only a small error. In order to determine the surface areas, values are required for bone density, mass, and surface-to-volume ratio. The density was assumed to be normal, i.e., 2.0 g/cm^3.

Bone Mass

An estimate of bone mass was made by measuring the masses of the left femur and tibia with the bone mineral analyzer[7] and extrapolating these masses to the whole skeleton using the ICRP Standard Man data.[8] A value of 2100 g was obtained. This is considerably below normal, probably because of bone loss associated with Cushing's syndrome.

Surface-to-Volume Ratio

Measurements of surface-to-volume ratio in the cervical and lumbar vertebrae were made using the CHR microanalyzer.[9] For the lumbar vertebra a value of 114 cm^2/cm^3 was obtained and for the cervical vertebra the value

was 101 cm^2/cm^3. These compare with a value of 110 cm^2/cm^3 reported by Lloyd and Hodges for the third lumbar vertebra in a normal adult male.[10] Because of this close agreement in cancellous bone, the Lloyd and Hodges value of 33 cm^2/cm^3 for compact bone was assumed to apply.

Initial Uptake

With the use of the concentration measurements, surface-to-volume ratio, and bone mass, the initial skeletal uptake was estimated to be 26% of the injected amount. (If the initial concentration on Haversian canal surfaces is 33% of the endosteal surface average, the uptake is 32% of the injected amount.) These values compare with an average of 49 ± 8.3% estimated by Durbin[2] for six human cases 4 to 457 days after injection.

Initial Ratio of Surface Dose Rate to Uniform Dose Rate

The surface concentration and initial uptake data can be used to determine the initial ratio of surface dose rate to uniform dose rate. This ratio is dependent on the amount of ^{239}Pu initially deposited in the skeleton on the assumption that Cushing's syndrome does not alter the way in which ^{239}Pu is distributed throughout the skeleton. Thus data and conclusions drawn from this case should be applicable to other human cases as well. Since the ratios are directly proportional to the surface concentrations, they show the same trend as the concentration data, being highest in the axial skeleton, intermediate in the metaphyseal portions of the long bones, and least in the diaphyseal portions. The range of ratios is 3.3 to 39. In the lumbar and cervical vertebrae they are 30 and 39 respectively. This compares with a range of 10 to 23 quoted by Jee for the third lumbar vertebra in beagles.[6] Thus the ratios are about twice as high as in the beagle. This is probably a reflection of the fact that the beagle has a higher surface-to-volume ratio in vertebral bone than does man.[10]

Since the observed surface-to-volume ratio in the plutonium case is the same as that observed in normal man, the surface to uniform dose rates for this case apply to normal man as well.

The surface to uniform dose rate plays a central role in Marshall and Lloyd's model for prediction of the $^{239}Pu/^{226}Ra$ RBE in man.[3] In the absence of human data they assumed that the surface to uniform dose rates in man and beagle were the same and predicted that the RBE in man would be 3 ± 1 times the RBE in dog. When the computation is carried out using the surface to uniform dose rate ratio obtained in the present work for the human cervical vertebra, it predicts a RBE in man of 4.5 times that in the beagle. Use of the lumbar vertebra value gives 3.5 times and the rib value gives 3.7 times. Thus these data predict that the RBE in man will be about four times that in beagle, in good agreement with Marshall and Lloyd's prediction.

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PLUTONIUM IN THE EXCRETA OF THREE SUBJECTS 10^4 DAYS AFTER INJECTION

J. Rundo, P. M. Starzyk, J. Sedlet,* R. P. Larsen, R. D. Oldham, and J. J. Robinson*

Substantial amounts of ^{239}Pu were found in the daily excreta of two subjects who had been injected intravenously with plutonium citrate (^{239}Pu) 10^4 days previously. The urine of a third subject injected intramuscularly with ^{238}Pu contained just measurable amounts of this nuclide.

Introduction

Three persons who had received injections of plutonium in 1945–1947 were hospitalized on a metabolic ward in 1973. Complete collections of urine and feces were made for periods of 8 to 14 days, and these excreta were shipped to ANL for plutonium analysis. Two of the individuals received intravenous injections of about 0.3 μCi of plutonium (IV) citrate; the third individual received an intramuscular injection of 0.095 μCi of plutonium (VI) nitrate. The intravenous injections were of ^{239}Pu, while the intramuscular injection was of ^{238}Pu.

The intramuscular injection was made in the gastrocnemius muscle of a leg having a bone sarcoma; four days after the injection, the leg was amputated. Analysis of a 69-g sample of tissue from what was described as the "injection site" showed that it contained 0.044 μCi. Because of the possibility that tissue adjacent to the "injection site" also contained unabsorbed plutonium, it is impossible to establish an accurate value for the initial systemic burden.

This report is confined to the description of the methods used for the analyses of these unique and important samples, together with the results. Interpretation will be presented elsewhere. For a description of the early experiments and their results, the reader is referred to the extensive review prepared by Durbin.[1] Some pertinent details of the three subjects are set out in Table 1.

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TABLE 1. Some Details of the Three Subjects Who Survived Their Primary Diseases.

CHR case No.	Literature[a] case No.	Sex	Original diagnosis	Age in 1973, yr	Amount injected, μCi
40-003	Cal-3	M	Osteo-fibro myxochondrosarcoma	62	0.095 (^{238}Pu)
40-009	Hp-3	F	Hepatitis, dermatitis, hypoproteinemia	77	0.301 (^{239}Pu)
40-012	Hp-6	M	Addison's disease	72	0.331 (^{239}Pu)

[a]Literature case numbers are those in Reference 1.

Urine: Sample Treatment, Aliquoting, and Analysis

During each 24-hr collection period the individual urine specimens were transferred to a polyethylene bottle; at the end of the collection period the urine was frozen. The samples were shipped to ANL and kept frozen until they were aliquoted.

To aliquot a 24-hr urine specimen, it was thawed and transferred, along with several concentrated nitric acid washes of the original container, to a tared mixing cylinder. The amount of nitric acid used was such that the final acidity of the urine was about 2.0 N. After the urine had been mixed with the acid and the mixing cylinder reweighed, the solution was apportioned about equally to 12 tared polyethylene bottles. The bottles were then retared and their contents frozen. These portions were individually analyzed; the fraction factor for each portion was calculated from the weight of each portion and the total weight of acidified urine.

The plutonium content of the urine was determined by alpha spectrometric-isotope dilution analysis using ^{242}Pu as the spike isotope. The aliquot was thawed, the ^{242}Pu spike was added, the urine was transferred, along with nitric acid washes of the container, to an erlenmeyer flask and the urine was wet-ashed. The ashing was considered to be complete if the salt residue was white when evaporation was carried to dryness. The salts were then dissolved in 2 N nitric acid.

The plutonium was separated from the other inorganic constituents of the urine by first coprecipitating it with cerous fluoride and then subjecting it to an anion exchange separation procedure. Hydroxylamine was added to the

nitric acid solution, the solution was heated to reduce the plutonium to the trivalent state, cerous nitrate was added, and cerous fluoride was precipitated by the addition of hydrofluoric acid. After separation of the cerous fluoride by centrifugation, it was dissolved by heating with 8 N nitric acid that had been saturated with aluminum nitrate. This solution was passed through a column of Dowex 1 × 8, and the column was washed, first with 8 N nitric acid and then with 12 N hydrochloric acid. The plutonium was eluted from the column with 0.1 N hydrochloric acid-0.01 N hydrofluoric acid.

The plutonium was transferred from solution to the surface of a polished stainless steel planchet for alpha spectrometric assay by an electrodeposition procedure. Sulfuric acid was added to the eluant solution, the solution was evaporated to fumes of sulfuric acid, diluted with water, and neutralized with ammonia gas to a pH of 2.0. The electrodeposition was carried out for 1.5 hr at 1.2 amp. The planchets were counted until about 300 counts had been accumulated in the ^{239}Pu peak. The amounts of activity in the aliquots ranged from about 0.3 pCi to 0.75 pCi; the counting efficiency was about 35%.

The alpha spectrograms ranged in quality from good to excellent, a "good spectrum" being defined as one in which the FWHM of the ^{242}Pu peak is 0.12 MeV and an "excellent spectrum" as one in which the FWHM is the same as that obtained in the electrodeposition of standards, i.e., 0.06 MeV.

As the analysis of several aliquots of the urine from case 40-003 showed that there was too little plutonium for measurement, the aliquots that had been made from each of three 24-hr collection periods were recombined and analyzed. In the alpha spectrograms, integration of the ^{238}Pu peak at 5.48 MeV was impeded by the presence of a peak at 5.43 MeV. The radionuclide producing this peak was identified as ^{228}Th. By counting the plates after 3.62-day ^{224}Ra had reached secular equilibrium with its ^{228}Th parent and integrating the counts in the ^{224}Ra peak at 5.68 MeV, we could calculate the ^{228}Th contribution to the ^{228}Th–^{238}Pu peak. This contribution ranged from 20 to 25% of the total. It was subsequently established that ^{228}Th as well as ^{230}Th and ^{232}Th were present in the reagents. The hydrochloric acid wash of the anion exchange column, although extensive, had not been sufficient to wash

all the thorium away from the plutonium.

Feces: Sample Treatment and Analysis

At the time the fecal samples were obtained they were bagged and frozen. They were kept in this condition until the time of analysis.

To prepare the samples for analysis, they were thawed, the ^{242}Pu spike was added, and the organic matter destroyed by first dry-ashing them for 16 hr at 500°C and then wet-ashing by repeated additions of nitric acid and evaporation to dryness. When the residues from the nitric acid treatment were judged by their appearance to contain no residual organic material, they were dissolved by adding concentrated hydrochloric acid and heating to 80°C. These solutions were analyzed by the radiochemical procedure described above for the urine samples.

For 22 of the 24 samples analyzed, the ^{242}Pu recovery ranged from 66 to 100%. Although the recoveries in two of the analyses were only 10%, the ^{239}Pu excretion rates obtained did not appear to be significantly different from the rates obtained where the recoveries of ^{242}Pu were much higher. From this it is inferred that isotopic exchange between the ^{239}Pu and ^{242}Pu had been established in all the samples during the operations used to destroy the organic material.

Results

To establish the precision of the analysis three aliquots from each of three urine samples were analyzed, and the values were compared. In each comparison all values were within the 95% confidence limits calculated from the average value and the number of counts in the ^{239}Pu peak.

The amounts excreted in the 24-hr urine samples are summarized in Table 2, while the results for the fecal samples are given in Table 3. One aspect of the entries in these tables calls for comment. For cases 40-009 and 40-012 the statistical errors on the results in Table 3 are all substantially lower than on those in Table 2, yet the numbers are lower in Table 3. This is because only small aliquots (5–10%) of the 24-hr urine samples were analyzed,

while the whole of each fecal sample was assayed.

Day-to-day variations in the urinary output of plutonium-239 were comparatively small; the ratio of highest to lowest daily output was 1.48 for case 40-009 and 1.36 for case 40-012. There were much larger sample-to-sample variations in the fecal output. The number of days of excretion represented by the sample was determined by identifying the beginning and end of each of two periods when a carmine dye appeared in the stool. For case 40-012 the results for the two periods were in complete agreement, and the daily fecal excretion was 38% of the mean daily urinary excretion. The results for case 40-009 were not so straightforward; the mean daily fecal excretion was substantially higher in the first period than in the second period, and a sample voided just before the start of the first period contained a remarkably large amount of plutonium (sample 2, Table 3). This patient had been suffering from diverticulitis with paralytic ileus which ended the day before sample 1 was collected. It seems likely that the high excretion rate of plutonium just prior to and during part of the first marker period reflected the voiding of feces containing plutonium which had continued to be secreted into the gastrointestinal tract during the period of constipation. The mean daily excretion during the second period may thus be our best estimate of the true fecal elimination rate; it was 42% of the mean daily

TABLE 2. Plutonium in the 24-hr Urine Samples.

Day	Plutonium content of urine samples, pCi/day		
	Case 40-003[a] (^{238}Pu)	Case 40-009	Case 40-012
1	-	6.50 ± 0.24	4.62 ± 0.25
2	-	9.00 ± 0.34	3.94 ± 0.28
3	-	8.23 ± 0.21	4.56 ± 0.26
4	-	7.91 ± 0.25	5.33 ± 0.26
5	0.062 ± 0.005	7.63 ± 0.54	4.42 ± 0.32
6	-	7.72 ± 0.37	4.90 ± 0.28
7	-	7.47 ± 0.39	5.35 ± 0.34
8	-	7.38 ± 0.38	4.46 ± 0.25
9	0.059 ± 0.005	6.59 ± 0.34	
10	0.055 ± 0.010	7.37 ± 0.47	
11	-	8.41 ± 0.49	
12		7.77 ± 0.38	
13		6.09 ± 0.43	
14		8.05 ± 0.39	
Weighted mean ± S.E.	0.060 ± 0.003	7.60 ± 0.21	4.68 ± 0.17
Time since injection, days	9474	9934	10,008

[a] Small aliquots did not provide sufficient ^{238}Pu for analysis of samples from case 40-003; only 3 of the 11 samples were analyzed in toto.

TABLE 3. Plutonium in the Fecal Samples from the Two Patients Who Received ^{239}Pu

Sample No.	Weights and plutonium contents of fecal samples: Case 40-009[a] Wet weight, g	pCi	Case 40-012[a] Wet weight, g	pCi
1	20	1.94 ± 0.06	33.5	0.43 ± 0.02
2	222	18.7 ± 0.4	50.5	0.77 ± 0.03
3	135.5	9.18 ± 0.30	178.5	1.87 ± 0.06
4	75	2.92 ± 0.11	217	2.09 ± 0.08
5	167	4.96 ± 0.16	269.5[b]	1.46 ± 0.08
6	161.5	6.27 ± 0.10	90 [b]	0.91 ± 0.06
7	95.5	2.79 ± 0.11	98	2.21 ± 0.09
8	170	3.90 ± 0.10	53	0.85 ± 0.03
9	94.5	3.10 ± 0.15	125	1.72 ± 0.06
10	83	2.51 ± 0.08	132.5	2.29 ± 0.10
11	324	7.34 ± 0.50		
12	54	2.18 ± 0.10		
13	143	3.30 ± 0.10		
14	53.5	1.35 ± 0.08		
Mean for period I		5.22 pCi/d (5 days)		1.78 pCi/day (4 days)
Mean for period II		3.17 pCi/d (6 days)		1.77 pCi/day (4 days)

[a] The horizontal lines indicate the starts and stops of time periods defined by the appearance of a dye marker in the stool.

[b] Combinations of 2 or 3 smaller samples voided at short intervals.

urinary excretion. This is similar to the result for case 40-012.

Because of the importance of these analyses, large numbers of aliquots of the urine samples were analyzed by two of us independently, and also by the Bio-Analytical and Chemical Section of the Industrial Hygiene Group at the Los Alamos Scientific Laboratory. With only two exceptions all the values from the aliquots of one 24-hr urine sample agreed within the statistics of counting. The averages of the three sets of values also agreed within this limit.

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PROGRESS IN CALIBRATION FOR THE EXTERNAL COUNTING OF PLUTONIUM IN THE LUNGS

R. E. Toohey and J. Rundo

Calibration factors for the proportional counter have been obtained from a pair of phantom lungs containing 1.0 μCi ^{239}Pu. These factors agree well with the results of a "mock Pu" in vivo intercalibration experiment previously performed. Some of the more important variables affecting calibration for the detection of plutonium in the lungs are discussed.

Introduction

A continuing program is under way at the Center for Human Radiobiology to develop a system for the measurement in vivo of low-energy, x-ray emitters. The transuranic elements are the most important group, and plutonium is of particular interest. ^{239}Pu emits conversion L x rays of uranium following 4.6% of its α decays; the energies of the x rays are 13.6, 17.2, and 20.2 keV. For the combination of x rays of these energies in the proportions 1.2:2.4:1, the first half-value layer for attenuation by soft tissue is about 0.6 cm, and a few mm of bone are effectively opaque. Consequently, severe problems arise in calibrating the detection system measurements in vivo. One would like to be able to detect the smallest possible fraction of the maximum permissible lung burden of ^{239}Pu, 16 nCi (about 0.25 μg).

We are currently exploring the potential of an 18-cm diameter proportional counter filled with a mixture of 90% xenon and 10% methane at atmospheric pressure. The counter has an internal anticoincidence chamber above the main chamber and is operated with pulse-shape discrimination. The advantages of a proportional counter for this work are its large sensitive area, low background, and improved resolution compared to a scintillation detector. The proportional counter is located in one of the three steel rooms of the underground vault.

The counting geometry we have tentatively adopted as standard is with the counter centered over the mid-sternum of the supine subject and oriented parallel to the sternum. The counter window is separated by a distance of 1 cm from the chest surface and a total counting time of 30 min is used. After the

first 10 min of counting, the counter is repositioned if the chest wall has receded from it, as often occurs with subjects of medium-to-heavy build in the supine position. Since the 13.6-keV x rays are usually completely absorbed by the chest wall of even a slender subject, the energy band from 15.6 to 23.9 keV is integrated to obtain the counting rate from the subject's radioactive content.

Calibration Factors

The calibration factor is primarily a function of the amount of tissue covering the source, which in turn is a function of body build. The chest wall thickness, that is, the mean thickness of soft tissue overlying the rib cage, may be estimated from Ramsden's expression [1]

$$CWT = 15.3\ W/H - 0.01C - 3.55\ ,$$

or from Dean's expression[2]

$$CWT = 0.0071 + 5.12\ W/H,$$

where CWT is the chest wall thickness in cm, W is the weight in kg, H is the height in cm, and C is the chest circumference in cm. Either expression, based on data obtained by ultrasonic measurements of the chest wall thicknesses of volunteer subjects, reportedly predicts CWT with a standard error of 0.2 cm. Because of the severe attenuation of plutonium x rays by soft tissue, an error of ± 20% in estimating the chest wall thickness of a subject of average build leads to an error of a factor of $\mp$ 2 in determining the body burden.

We were fortunate during the past year to acquire on loan from Harwell a pair of phantom lungs loaded with 1 μCi ^{239}Pu. The lungs are molded from "Temex," a rubber material of the same effective atomic number as soft tissue,[3] which had been foamed to have about the same density as lung tissue. Calibration factors were obtained by arranging these lungs in the correct anatomical configuration under the counter and placing successive layers of Alderson-Rando tissue-equivalent absorber over them. Figure 1 shows the data obtained, joined by a smooth curve. The curve departs from a simple exponential because of a) forward scattering, b) the inclusion of two x rays of different energies in the counting band, and c) the fact that the source is almost

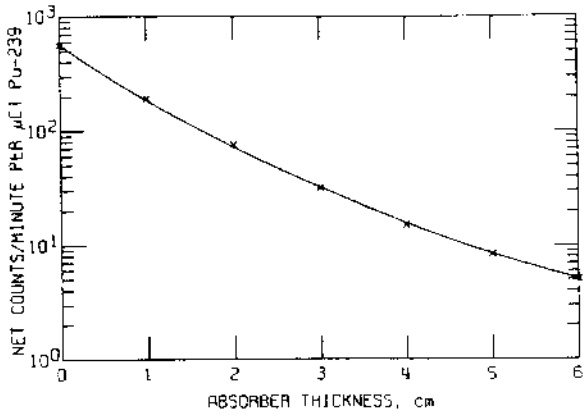

FIG. 1.--Calibration factors derived from the Pu-loaded phantom lungs. These factors must be multiplied by 0.57 to account for the effects of bone in a subject.

infinitely thick at these low energies.

Before these calibration factors can be applied to a subject, however, they must be corrected for the effects of the rib cage. In making this correction, we assume that the bones of the chest are completely opaque to the x rays, and consequently, that no x rays are counted from that fraction of the chest covered by bone. This fraction has been determined by projecting the area of the proportional counter window on an anatomical drawing of the thoracic skeleton and measuring the area covered by bone with a planimeter. This measurement showed that 41.5% of the area viewed by the proportional counter was obscured by bone. Similar measurements have been made by Ramsden[1] on chest radiographs of 19 subjects yielding a mean of 42.9 ± 4.8% of the chest covered by bone.

Table 1 presents a comparison of the calibration factors with the results of the IAEA-sponsored intercalibration experiment with "mock Pu" in vivo reported last year.[4] The uncertainties in the calibration factors are approximately ± 20%. The agreement is encouraging in view of the rather simple calibration technique.

TABLE 1. Comparison of in Vivo and Phantom Lung Calibration Factors

Subject	Chest wall thickness, cm	cpm/μCi ^{239}Pu: Revised results of "mock Pu" exper.[a]	cpm/μCi ^{239}Pu: Calibration factors from phantom lung
DN	1.04	97	103
JR	2.38	37	30
KB	3.23	15	15

[a] The results reported last year have been revised due to a change in the reported strength of the "mock Pu" used.[5]

Room and Subject Backgrounds

Another source of difficulty in measuring the plutonium content of a contaminated subject is the determination of the correct background to subtract from the gross counting rate. The room background, measured

with no subject present, has been found to be quite constant and may be lowered by pulse-shape discrimination techniques. The more important component of the background is that coming from the subject himself. Higher energy γ rays from ^{40}K and ^{137}Cs in the body are scattered and degraded in energy by body structures and contribute to the counting rate in the x-ray band; the amount of this contribution varies from subject to subject.

Backgrounds have been measured from 10 control subjects in an effort to predict the counting rate from some function of the subjects' physical dimensions. The best correlation was obtained between the counting rate in the x-ray band and the ratio of weight to height. However, the correlation coefficient was only +0.60, so the subject background cannot be predicted accurately enough in this manner, and an alternative method has been developed. The background measured with the counter positioned over a subject's abdomen has been found to agree to within a few percent with that measured over the chest. Since no radiation from contamination in the lungs reaches the counter when it is positioned over the abdomen, this method seems the most direct and simple way of determining the subject's background, but it is limited in its application to those cases where there is no plutonium in the gut or liver of the contaminated subject.

Instrumentation

The instrumentation for the proportional counter performs two analyses before routing the output pulse of the counter to a multichannel analyzer. First, the output of the main chamber is rejected if a simultaneous output is received from the anticoincidence chamber. The use of this anticoincidence technique reduces the room background from 29.3 to 4.4 cpm in the x-ray band (15.6 – 23.9 keV). Second, the output of the main chamber is subject to pulse-shape discrimination; that is, the risetime of the pulse must fall within a narrow range.

A block diagram of the electronics is shown in Figure 2. Briefly, the output of the main chamber preamplifier is fed to two separate active filter (RC) amplifiers with different time constants. Although the crossover time of the bipolar output pulse of these amplifiers is largely determined by the shapping network, it also remains proportional to the input pulse risetime. Con-

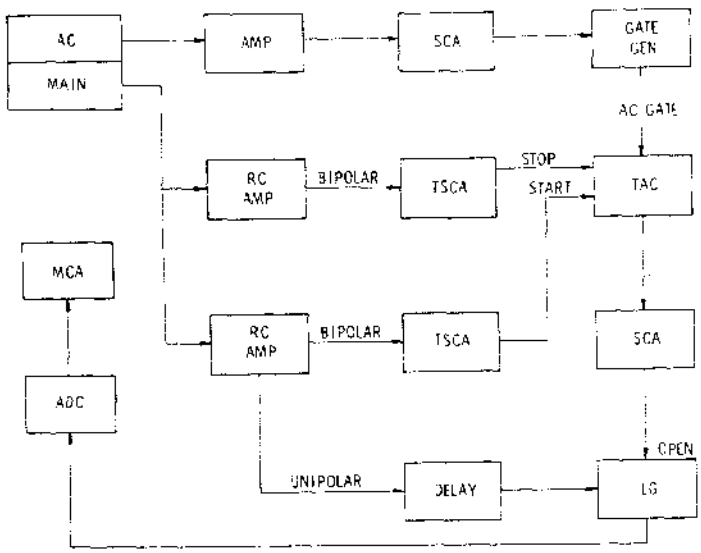

FIG. 2.--Block diagram of the counting electronics for the proportional counter. AC, anticoincidence chamber; AMP, amplifier; SCA, single-channel analyzer; TSCA, timing single-channel analyzer; TAC, time-to-amplitude converter; LG, linear gate; ADC, analog-to-digital converter; MCA, multichannel analyzer. (ANL Neg. 149-6374)

sequently, the difference in the crossover times of the outputs of the two amplifiers is a function of risetime of the input pulse. The output of each amplifier is fed to a timing single-channel analyzer (TSCA), which produces an output at the crossover point of the bipolar input pulse. These outputs start and stop a time-to-amplitude converter (TAC), whose output is, therefore, proportional to the risetime of the main chamber output. The TAC is inhibited by a pulse from the anticoincidence chamber. If the TAC output falls within a preset range determined by a single-channel analyzer, a linear gate is opened, and the delayed unipolar pulse from the main amplifier is allowed to pass to an analog-to-digital converter and multichannel analyzer (Nuclear Data 4410).*

The distribution of the risetimes of x-ray pulses from a plutonium source has a peak at 750 nsec and a half-width of 20 nsec. The risetimes of the room background pulses also peak at 750 nsec, but the distribution is asymmetric. The half-width (from the center to the half-maximum) of the peak is 30 nsec to the fast side, but 80 nsec to the slow side. Consequently, the signal-to-background ratio can be optimized by proper selection of the range of risetimes to be accepted.

*This instrumentation system was originally designed by K. Eckerman.

Figure 3 shows the dependence of S^2/B (the ratio of the counts squared from a plutonium source to the room background counts) on the range of risetimes accepted, centered on 750 nsec. The optimum value occurs for a range of 40 nsec. At this value, the room background in the x-ray band is 1.5 cpm, compared to a background of 4.4 cpm if no pulse-shape discrimination is used. However, the subject background is not appreciably reduced by pulse-shape discrimination, since it has the same photon energy and pulse risetime characteristics as the plutonium x rays.

^{238}Pu in Vivo Intercomparison Experiment

We have recently participated in another intercomparison experiment, this one involving a subject who was exposed to ^{238}Pu in an industrial accident. The subject has visited six in vivo plutonium counting laboratories in the U.S. and served as a means of comparing their techniques and results, as in the "mock Pu" experiment.

The detection in vivo of ^{238}Pu is simplified somewhat by the fact that ^{238}Pu emits approximately two and a half times as many x rays as does ^{239}Pu per α disintegration. The chest wall thickness of this subject was estimated at 3.02 ± 0.20 cm (Ramsden's expression[1]) or 2.55 ± 0.20 cm (Dean's expression[2]), and a calibration factor from the phantom lungs was multiplied by 2.48 to apply to ^{238}Pu. The gross counting rate from the subject's chest was 9.25 ± 0.48 cpm. The background counting rate measured with the detector over the subject's abdomen was 8.25 ± 0.52 cpm, including the room

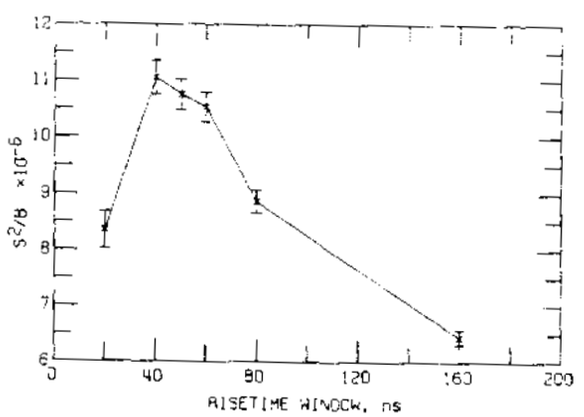

FIG. 3.--The dependence of S^2/B on the width of the pulse-shape discrimination window. The signal was obtained from a 1-μCi ^{238}Pu source at 10 cm from the detector.

background. Thus the net counting rate due to plutonium in the chest was 1.00 ± 0.71 cpm, corresponding to 23 ± 16 nCi (Ramsden) or 15 ± 11 nCi (Dean) ^{238}Pu. This subject has been estimated to have a lung burden of between 11 and 30 nCi ^{238}Pu at other laboratories.[6] Measurements of the chest wall thickness by the ultrasonic echo technique at these laboratories gave an average value of 2.37 cm (range 2.2–2.6 cm). The calculated value from Dean's expression, derived from data on almost 400 cases, is in this range; Ramsden's expression was much less solidly based (19 cases), so it would seem reasonable to place more reliance on the lower estimate. This emphasizes the importance of the effect of the chest wall thickness and points to the need for measurements by the ultrasonic method.

Measurements of ^{241}Am Related to Plutonium Calibration

The measurements with the proportional counter of case 30-041 are described in detail elsewhere in this report. These measurements were of interest in the calibration of plutonium detection since ^{241}Am is commonly present in modern samples of plutonium to which workers may be exposed. Under these circumstances the contribution of the ^{241}Am x rays and of scattered radiation from the 60-keV γ ray to the x-ray band must be subtracted before the plutonium content can be determined. Measurements of the radiation from the subject and from a ^{241}Am source covered with absorber indicated that the ratio of the counts in the x-ray band to the counts in the 60-keV peak was constant to within a few percent over the range of 3 to 6 cm of absorber. This is the range of interest for measurements in vivo, since the absorption in lung tissue must be added to that of the chest wall. In this range of absorber thickness the increased absorption of the x rays is just offset by increased scattering of the 60-keV γ rays. With this ratio, the contribution of ^{241}Am in a plutonium case can be determined.

Conclusions

The calibration of an in vivo plutonium detection system is a formidable problem. We may estimate our progress toward its solution by considering

some of the remaining difficulties.

At the present time, the subject background is the limiting factor on the minimum amount of plutonium which can be detected. In the ^{238}Pu intercalibration experiment mentioned above, the net response to plutonium was equal to only two standard deviations of the background counting rate. Consequently, not only is a method of predicting the subject background required, but efforts to reduce it are also necessary. This background cannot be appreciably reduced by anticoincidence or pulse shape discrimination techniques without at the same time lowering the sensitivity of the detector to Pu.

The Pu-loaded phantom lungs are preferable to a point source for deriving calibration factors; however, it is still necessary to determine more exactly the effects of a) the ribs and sternum, b) a nonuniform deposition of Pu in the lungs, and c) the translocation of Pu to the tracheobronchial lymph nodes by physiological clearance processes.

Finally, a direct measurement of a subject's chest wall thickness is preferable to an estimate based on body build. This may be done by using an ultrasonic probe.[1]

In summary, we now have a reliable system to detect Pu contamination in the lungs down to a level below the maximum permissible lung burden for subjects of small body build and in the region of the maximum permissible burden for subjects of average build. However, much work remains in order to lower the minimum detectable activity and improve the calibration of the system.

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5. Newton, D., AERE Harwell, England, private communication, 1973.
6. Tomlinson, K., Mound Laboratory, private communication, 1974.

Note Added in Proof

Since this report was prepared a further revision of the results (Table 1) of the IAEA-sponsored "mock Pu" in vivo intercomparison experiment has become necessary. The standardization of the ^{103}Pd used in the experiment was based on concordant published values for the intensity of the weak 357-keV γ ray; these values have since been discovered to be incorrect.* A new standardization, based on the Rh K x rays, has been obtained by several laboratories, and we have verified this standardization with a ^{103}Pd source supplied by Dr. A. L. Anderson of Lawrence Livermore Laboratory.

In addition, we have also obtained values for the chest wall thickness of each volunteer, measured by an ultrasonic echo technique. The calibration factors from the phantom lungs have been modified accordingly. The two sets of calibration factors are compared in the table below, which now should be considered as replacing Table 1.

Comparison of Current Calibration Factors for ^{239}Pu

Subject	Ultrasonically-measured chest wall thickness, cm	cpm/μCi ^{239}Pu	
		Results of "mock Pu" experiment	Calibration factors from phantom lungs
DN	1.7	47	54
JR	2.5	22	27
KB	3.1	9.5	16.5

*D. Newton, AERE, Harwell. Private communication, 1974.

MEASUREMENTS OF ^{241}Am IN VIVO SEVEN YEARS AFTER INHALATION

R. E. Toohey, M. A. Essling, and J. Rundo

Case 30-041 was remeasured to investigate the retention and possible redistribution of a body burden of 1.8 μCi of ^{241}Am, last measured in 1967. Long-term chelation therapy seems to have reduced the amounts of radioactive material in the skeleton, but to have reduced the lung burden only slightly.

Introduction

In 1967, four individuals, who had been exposed in an industrial plant to airborne americium particles over a period of several months, were investigated at ANL for possible internal contamination. One man was found to have a burden at least fifty times that of the others, from measurements of the 60-keV γ ray with a NaI(Tl) crystal. A detailed series of measurements was then made to determine the apparent distribution of the isotope within his body. The results indicated widespread deposition in the skeleton in addition to material remaining in the lungs. The total body burden was estimated to be 1.8 μCi. The relative uptakes of ^{241}Am in different parts of the skeleton were consistent with the assumption that americium was deposited on bone surfaces and, therefore, was concentrated more in trabecular bone than in cortical bone.[1] Since that time, this subject has undergone weekly chelation therapy with DTPA at a hospital.[2]

The subject visited the Center for Human Radiobiology in 1973 and an attempt was made to reproduce some of the previous measurements. In addition, several new measurements, including "profile scane," were made. A proportional counter was also used since it offered the advantage of detecting the conversion L x rays emitted by ^{237}Np following the α decay of ^{241}Am, as well as the 60-keV γ rays detected by the crystals.

γ-ray Measurements

Seven-Position Scans

A measurement of the 60-keV γ ray was made at each of seven positions along the body of the supine subject. Two 11-1/2" × 4" NaI(Tl) detectors were used, one at 30 cm above the bed and the other at 10 cm below the bed. The

standard seven-position scan was made, with an interval between positions equal to 15% of the subject's height; the fourth position was at the midpoint of the subject's height. The results of these measurements indicated the general longitudinal distribution of ^{241}Am in the body and are shown in Figure 1. The peak at position 2 arises from activity in the vertebrae, ribs, and thorax. The higher counting rates from the upper crystal at positions 5 and 6 indicate activity in the bones of the legs, and especially in the knees, since more soft tissue shields the lower crystal than the upper, and the legs are much closer to the lower crystal than to the upper.

These scans indicate that much of the activity in the thorax actually lies in the lungs for the following reasons: 1) a higher counting rate is observed from the back than from the front; 2) the maximum counting rate appears to be nearer the vertex when measured from the front than from the back; and 3) a broader peak in the counting rates is observed from the back than from the front. All these characteristics can be predicted from a consideration of the size, shape, and positioning of the lungs in the thoracic cavity.[3]

Profile Scans

A lead collimator with a one-inch-wide slit transverse to the long axis of the body was then placed over the lower detector, and a longitudinal profile scan was made with 10-cm intervals. The interval was decreased to 5 cm in regions where the response from ^{241}Am differed markedly from one position to the next. Figure 2 shows the profile obtained. Peak counting rates occur in the regions of the skull, chest, pelvis, knees, and feet, consistent with the 1967 results, which indicated labeling of the entire skeleton, but especially trabecular bone. The large peak from the chest is due to material both in bone and in the lungs, and the asymmetry of this peak at 50 cm from the vertex may be due to material in the liver and spleen, or in the rib cage and vertebrae. The aperture was then turned through 90° so as to be parallel to the long axis of the body, and a transverse profile scan was made of the chest at 40 cm from the vertex, the position which yielded the highest counting rate in the longitudinal scan. The transverse scan is shown in Figure 3. The asymmetry of this scan indicates that the response came from material in the lungs

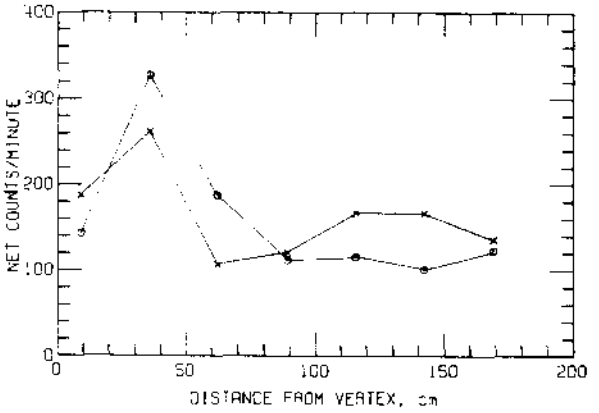

FIG. 1.--Seven-position scans of case 30-041. The counting rate from the upper crystal is shown by the symbol (x), and the counting rate from the lower crystal by (O). (ANL Neg. 149-6429)

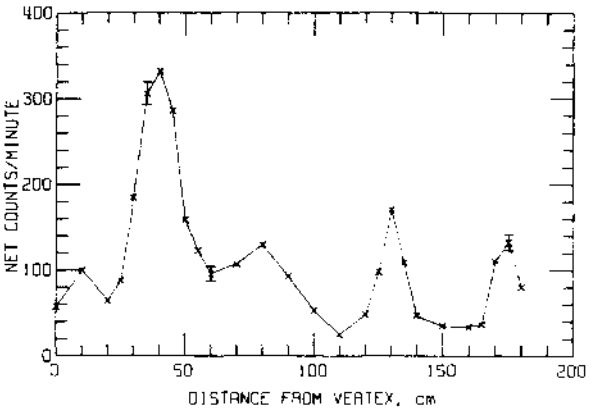

FIG. 2.--Longitudinal profile scan of case 30-041. These measurements were made with a 1-inch-wide aperture on the lower crystal. (ANL Neg. 149-6427)

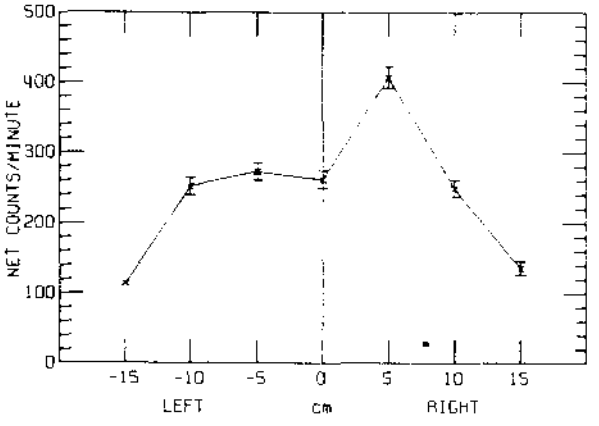

FIG. 3.--Transverse profile scan of case 30-041 at 40 cm from the vertex. The positions are left and right of the median line of the body. (ANL Neg. 149-6430)

(the right lung is larger than the left), rather than in the bone. Unfortunately, these γ-ray measurements with the crystals offered no direct method of determining the effective or average depth of the source in the body or of distinguishing between deposition in soft tissue and in bone.

Whole Body Content

The subject's radioactivity was measured with an 11-1/2" × 2" NaI(Tl) crystal while he lay on a curved bed with a 1.5-m radius. Measurements were made both with the subject facing the detector and with his back to the detector, which was mounted 1.37 m above the bed. The efficiency of detection of the 60-keV γ ray in this configuration varied along the length of the bed. A correction was made for this by weighting the efficiency observed at

each of several positions along the curved bed with the relative radioactivity of the subject at that position, obtained from the seven-position scan.

Transmission measurements using a phantom of Alderson-Rando tissue-equivalent material established the mass attenuation coefficient for the 60-keV γ ray to be 0.130 cm^2/g, with a forward scatter correction of 1.28. With the use of these values and of transmission data for the subject (measured at the upper back, lower back, and knee), the effective average thickness of the body was determined to be 12.4 cm.

With the assumption that the effective center of the radioactivity was at the midplane of the body, the whole body burden of ^{241}Am was calculated to be 1.00 ± 0.03 μCi.

Repetition of Previous Measurements

When the subject was measured in 1967, the general distribution of ^{241}Am was determined with one of the same detectors used for the profile scans. Sheets of 1/16-inch-thick lead were placed over the supine subject so as to define a 10-cm-wide unshielded rectangular slit. The long axis of the slit was perpendicular to the long axis of the body and the upper 11-1/2" × 4" crystal was centered over the slit at a height of 35 cm over the bed. Counts were taken with the slit centered approximately at the level of the first and seventh thoracic, and first lumbar vertebrae (T1, T7, and L1). A fourth count was taken over the legs, with the upper body shielded to the level of the first lumbar vertebra. These measurements were repeated during the subject's recent visit. The results of the two sets of measurements are presented below in Table 1. These results indicate that the subject has retained approximately 33% of the activity in the thorax and 45% of the activity in the pelvis and legs.

TABLE 1. Comparison of Measurements Made in 1967 and 1973

	Counts/min		
Position	1967	1973	Ratio
T1	6275	1907	0.30
T7	6190	2056	0.33
L1	3000	1149	0.38
Legs	5170	2316	0.45

X-ray Measurements

Introduction

A series of measurements was made on the subject with the 18-cm diameter proportional counter. This counter is intended for the detection of low-energy x-ray emitters in vivo, particularly, inhaled plutonium. The measurements of this subject are not only interesting in themselves, therefore, but also serve as a valuable aid to calibration of the detector. Incidents of plutonium inhalation often also involve americiam, since ^{241}Am is the daughter of ^{241}Pu present in spent reactor fuels. The 60-keV γ ray of ^{241}Am is scattered and degraded in energy by body structures and makes a significant contribution to the counting rate in the x-ray band. In addition, the contribution of the ^{241}Am x rays must also be taken into account.

The proportional counter spectrum obtained from the chest of this subject is shown in Figure 4. The peak at 30 keV results from the escape of a 30-keV xenon x ray following complete ionization of an atom of the counting gas by a 60-keV γ ray. The peak at 26.5 keV is from another ^{241}Am γ ray, and the conversion x rays are at 13.9, 17.8, and 20.8 keV.

Determination of Effective Source Depth

In order to determine the amount of ^{241}Am present in the chest of this subject, the amount of absorber interposed between the source and detector must be known. We estimated this quantity (the effective soft tissue thickness) from the equation suggested by Rundo et al.[4] For this subject the effective soft tissue thickness, which allows for the lower density of lung tissue, was 5.17 ± 0.73 cm. Normally, a calibration factor is then obtained by layering Alderson-Rando tissue-equivalent absorber over a ^{241}Am source to obtain a broad-beam attenuation curve, and a calibration factor (cpm/μCi ^{241}Am) is interpolated from this curve.

However, this procedure could not be applied directly to this subject because of the possibility of an unknown fraction of the total chest burden lying in the ribs, with the rest lying in the lungs. Consequently, we developed an alternative empirical method of determining the amount of absorber through which the ^{241}Am radiations from this subject had passed. In the spectrum of

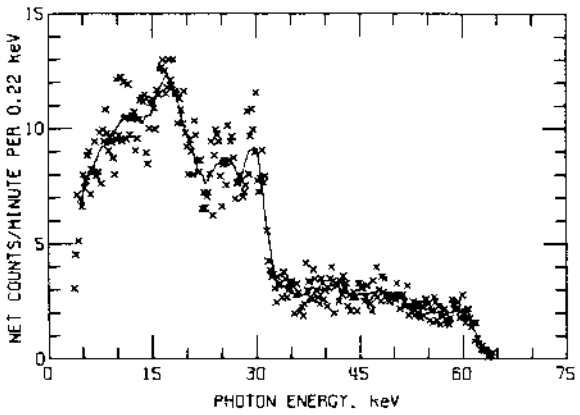

FIG. 4.--Spectrum obtained with the proportional counter centered over midsternum of case 30-041. The solid line was obtained by smoothing the experimental points over eleven-channel intervals; this results in considerable loss of resolution in the region below 35 keV.
(ANL Neg. 149-6428)

Figure 4, the region from approximately 35 to 45 keV consists entirely of scattered radiation from the 60-keV peak. Since the amount of scattering should increase with an increasing amount of scattering material present, the ratio R_{sp} of counts in the 35- to 45-keV region to the counts in the 60-keV peak was determined as a function of absorber thickness covering the source. This curve is shown in Figure 5. These measurements were repeated with several different source-to-detector geometries, with little effect on the results. The inclusion of bone as part of the absorbing material, as would be the case in vivo, also had little effect.

In the spectrum of Figure 4, the ratio of scattered counts to peak counts is 2.68 ± 0.13, indicating an absorber thickness of 4.86 ± 0.49 cm. This figure agrees quite well with the estimated effective soft tissue thickness of 5.19 ± 0.73 cm and, therefore, indicates that the burden lies primarily in the lungs. A small fraction of the total amount present lying in the rib cage cannot

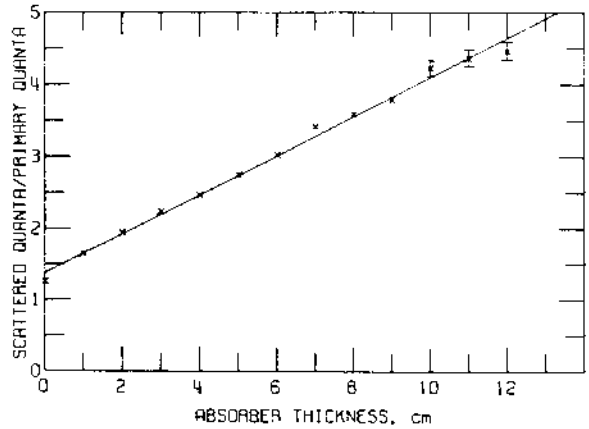

FIG. 5.--The counting rate ratio R_{sp} of scattered to primary counts in the 60-keV peak of ^{241}Am as a function of absorber thickness covering the source.
(ANL Neg. 149-6426)

be ruled out, however, because of the rather large limits of precision of these numbers. For instance, a combination of 80% of the total amount of ^{241}Am present under 5.25 cm absorber and 20% of the total under 2.5 cm absorber gives a scattered-to-peak ratio lying within the lower limit of that obtained from the subject.

Calibration Factors

The half-thickness of the subject's chest was 12.5 cm, which is the depth we assume for material lying in the lungs. This agrees well with the estimated effective soft tissue thickness if we allow 2.5 cm for the chest wall thickness and multiply the remaining 10 cm by 0.25 to account for the lower density of lung tissue. Since the counter is positioned 1 cm from the chest surface, material in the rib cage is almost four times closer to the counter than material in the lungs and so gives a much higher counting rate/μCi. Of course this is an oversimplification, since material in the lungs is more correctly represented by a distributed source. In the absence of a realistic chest phantom, however, this assumption is useful in obtaining calibration factors.

Another large correction factor arises when the x-ray band is considered. Since the rib cage is nearly opaque to the x rays, the calibration factor obtained from source and absorber measurements must be reduced by an amount which allows for the shadowing of lung tissue by bone. We presently use a factor of 43%; that is, we assume 43% of the chest viewed by the proportional counter is covered by bone. This factor has been found to give good results when applied to calibration factors for the in vivo counting of plutonium. The correction does not apply to the counting rate of the 60-keV γ rays, since the ribs are relatively transparent to photons of this energy.

The effect of activity in the ribs on the calibration factor is illustrated in Table 2. The first column gives the percent of the total chest burden assumed to lie in the ribs, with the remainder in the lungs. The next three columns give the total chest burden in μCi computed for this subject, derived respectively from the 60-keV γ-ray peak, the x-ray band omitting the correction for bone shadowing, and the x-ray band including the correction for bone shadowing:

TABLE 2. Effect of Activity in the Ribs on the Calibration Factor

% activity in ribs	Total chest burden, μCi		
	γ-ray band	x-ray band without shadowing	x-ray band with shadowing
0	0.23 ± 0.03	0.23 ± 0.02	0.41 ± 0.04
1	0.20 ± 0.03	0.19 ± 0.02	0.30 ± 0.03
5	0.12 ± 0.02	0.11 ± 0.01	0.14 ± 0.01
10	0.08 ± 0.01	0.08 ± 0.01	0.09 ± 0.01
15	0.06 ± 0.01	0.06 ± 0.01	0.06 ± 0.01
20	0.05 ± 0.01	0.05 ± 0.01	0.05 ± 0.01

All three estimates of the total chest burden agree when 10% or more of the total lies in the rib cage, simply because the counting rate from activity in that region would overwhelm that from deeper material in the lungs. However, large discrepancies occur when the correction for shadowing of the material in the lungs by bone is applied to the cases where 95% or more of the total activity is in the lungs. This situation indicates that the correction for bone shadowing may not be simply applied to americium contamination. One could predict this by considering the fact that scattered radiation from the 60-keV peak makes a substantial contribution to the counting rate in the x-ray band when several centimeters of absorber are positioned between the source and the detector. It is evident from Figure 4 that scattered radiation may be contributing at least one-half of the counting rate in the x-ray band. An exact method of determining the amount of this contribution remains to be developed. Consequently, estimates of activity based on the 60-keV peak are more reliable.

Conclusion

An interesting comparison may be made between the results obtained with the proportional counter and those obtained with the crystals. The longitudinal profile scan of Figure 2 indicates that approximately 25% of the total body burden resides in the chest region. If the proportional counter results are considered in light of this figure, then no more than a few percent of the chest burden lies in the rib cage. This same conclusion is supported

by the ratio of scattered-to-peak 60-keV radiation measured with the proportional counter, by the seven-position scans, and by the transverse profile scan of the chest.

Comparison of the current results with those of the 1967 measurements indicates that with prolonged chelation therapy, this subject has excreted approximately one-half of the initial body burden of ^{241}Am. Other measurements made in 1967 indicated that, of the total activity in the body, 28% was in the bones of the thorax, and 16% in the lungs.[1] Although time did not permit the repetition of those measurements, the recent measurements indicated that the situation has been reversed: the majority of activity now in the thorax resides in the lungs.

References

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