ENP

702257

ANL-7489

ANL-7489

Argonne National Laboratory

RADIOLOGICAL PHYSICS DIVISION ANNUAL REPORT

July 1967 Through June 1968

RETURN TO REFERENCE FILE
TECHNICAL PUBLICATIONS
DEPARTMENT

REPOSITORY Argonne Lab
COLLECTION
BOX No.
FOLDER

PARTIAL DOCUMENT

001652b

TABLE OF CONTENTS

TOXICITY OF RADIOELEMENTS

1 Localization of Radioactivity *in Vivo* by Photon Time-of-Flight Techniques: A Preliminary Report
L. D. Marinelli, G. F. Clemente, I. K. Abu-Shumays, and O. J. Steingraber

13 Division Delay and Recovery in Irradiated Sea Urchin Gametes
Patricia Failla

[illegible] Preliminary Report on the Distribution of Americium-241 Following Accidental Inhalation
Harold A. May

24 Comparative Kinetics and Autoradiography of Ca^{45} and Ba^{133} in 10-Year-Old Beagle Dogs
J. C. Ellsasser, J. E. Farnham, and J. H. Marshall

30 Thorotrast: An Intraperitoneal Injection
Henry F. Lucas, Jr. and Frank Markun

33 The Deposition of Uranium in Bone
R. E. Rowland and J. E. Farnham

[illegible] Sources of ^{210}Pb in Uranium Miners
Richard B. Holtzman

43 The Uptake of Calcium-45 in the Acellular-Boned Toadfish
David J. Simmons, Natalie B. Simmons, and John H. Marshall

53 The Effect of Deladroxone and Deladroxate on the Skeletons of Rats and Mice
David J. Simmons and Helen Cummins

68 Alopecia and Contraceptive Drugs in Mice
David J. Simmons and Helen Cummins

[illegible] Rickets in Rats: Progress Report II
David J. Simmons, Arthur S. Kunin, and Gerald B. Burg

77 A Comparison of the Relative Rates of Transfer of Barium, Radium, and Strontium between Blood and Bone
Elizabeth Lloyd

81 Microscopic Distribution of Radium in Bone by the Method of Thick-Source Alpha-Ray Spectroscopy
Frank L. Bordell

METEOROLOGICAL STUDIES

[illegible] Chicago Air Pollution Meterorological Studies
James E. Carson and Donald F. Gatz

94 The Validity of Currently Popular Plume Rise Formulas
James E. Carson and Harry Moses

95 Wide-Band Spectra of Atmospheric Turbulence
Paul Frenzen and Richard Hart

RADIOLOGICAL AND HEALTH PHYSICS

[illegible] Specific Primary Ionization by High-Energy Electrons
F. F. Rieke and William Prepejchal

100 The Effect of Pressure upon Ionization in Pure Rare Gases
Harvey A. Schultz

0016527

101 Total Cross Sections for Inelastic Scattering of Charged Particles by Atoms and Molecules. II. Negative Hydrogen Ion.
MITIO INOKUTI AND YONG-KI KIM

101 Generalized Oscillator Strengths of the Helium Atom. I.
YONG-KI KIM AND MITIO INOKUTI

102 Generalized Oscillator Strengths of the Helium Atom. II. Preliminary Report
Y.-K. KIM, D. DOUTHAT, AND M. INOKUTI

104 Optical Properties of Metallic Alluminum over the Entire Spectral Region
TAIZO SASAKI AND MITIO INOKUTI

104 Minima of Generalized Oscillator Strengths
YONG-KI KIM, MITIO INOKUTI, GEORGE E. CHAMBERLAIN, AND S. R. MIELCZAREK

105 Photoionization Studies
JAMES C. PERSON

105 Isotope Effects in the Photoionization Yields and the Absorption Cross Sections for Ethylene and *n*-Butane
JAMES C. PERSON AND PAUL P. NICOLE

105 Isotope Effects in the Photoionization Yields and Absorption Cross Sections of Several Gases
JAMES C. PERSON AND PAUL P. NICOLE

116 Ecological Dosimetry
J. KASTNER, B. G. OLTMAN, W. C. ASHBY, AND C. PADEN

117 Thermoluminescence in Bone and Bone-like Materials
F. L. BORDELL AND J. KASTNER

124 Linearity of Response for LiF at Low Exposures
P. TEDESCHI, B. G. OLTMAN, AND J. KASTNER

125 High-Dose Gamma-Ray Dosimetry with Single-Crystal Thermoluminescent Dosimeters
CHARLES PADEN

127 PUBLICATIONS

0016528

sea urchins. Argonne National Laboratory Radiological Physics Division Annual Report, July 1966 through June 1967. ANL-7360, p. 78.
6. Paden, Charles. High dose gamma dosimetry with single-crystal thermoluminescent dosimeters. This report.
7. Cormack, D. H. Site of action of ribonuclease during its inhibition of egg cleavage. *Nature* **209**, 1364–1465 (1966).
8. Elkind, M. M. Repair of x-ray damage in mammalian cells. *Mechanisms of the Dose Rate Effect of Radiation at the Genetic and Cellular Levels*, Proc. Conf. Oiso, Japan, November 1964. *Jap. J. Genet.* **40**(Suppl.) 176–193 (1965).
9. Sinclair, W. K. Report of trip to conference on Comparative Cellular and Species Radiosensitivity in Animals, Kyoto, Japan, May 20–23, 1968, unpublished.

PRELIMINARY REPORT ON THE DISTRIBUTION OF AMERICIUM-241 FOLLOWING ACCIDENTAL INHALATION

Harold A. May

INTRODUCTION

Four individuals who had inhaled unknown amounts of Am^{241} as a result of metallurgical procedures carried on by an industrial processor were brought to the Whole-Body Counter Facilities of this Division. Apparently they had been exposed repeatedly to airborne americium vapor and/or particles over a period of several months, but the total duration of exposure was quite uncertain. Initial measurements with an 11½″ × 4″ crystal over the chest and thorax of one individual resulted in a net counting rate of 13,800 counts/min from the 60-keV gamma ray. This was at least fifty times that of the others and suggested a body burden of many times the recommended MPL. Almost all of the subsequent examinations were devoted to ascertaining the apparent distribution of the isotope within the body of this individual. The present discussion is restricted to these measurements.

MEASUREMENTS

The first series of measurements was made to obtain an indication of whether the americium burden was confined to the lungs or had become more uniformly distributed. Since facilities for continuous linear scanning are not included in our low level room, and the size of the NaI crystals in everyday usage is too large to yield optimum spatial resolution and signal-to-background ratio, the methods employed were practically improvised.

Sheets of thin (1.6 mm) lead were placed horizontally over the supine patient with straight edges parallel so as to define a 10-cm wide, uncovered rectangular slit, the long dimension of which was always perpendicular to the spinal column. Three successive 10-min counts were made with the slit (and crystal above it at a height of 35 cm from the flat bed) centered approximately at the level of the first and seventh thoracic, and the first lumbar vertebra; this way counting over adjacent, nonoverlapping portions of the chest and abdomen was achieved. The distal boundary of the third region thus defined passes approximately through the distal limits of the lowest rib. A fourth count included everything below this level. The net counts above room background in the energy region from 32 to 70 keV were:

Proximal 10-cm zone (over T1)	6275 cpm
Median 10-cm zone (over T7)	6190 cpm
Distal 10-cm zone (over L1)	3000 cpm
Remainder (over hips and thighs)	5170 cpm

These results suggested that the existing burden was not located primarily in the lung. With the lead sheets positioned to define a 5-cm slot parallel to the spinal axis, the following readings were observed:

Subject prone		*Subject supine*	
Centered over spine	2520 cpm	Centered over sternum	2950 cpm
5-cm lateral displacement	2400 cpm	5-cm lateral displacement	3055 cpm
15-cm lateral displacement	1160 cpm	15-cm lateral displacement	2200 cpm

Other readings obtained with the uncollimated detector at a distance of 27 cm above the bed were: centered over knees—5300 cpm, over ankles and feet—4250 cpm, over skull—3325 cpm.

Considering the known affinity of americium for selective deposition upon bone surfaces, the findings strongly suggested a rather uniform labeling of the entire skeleton. The measurements taken over the lungs and abdomen are in agreement with this interpretation, but do not rule out a concurrent soft tissue burden. Hence, a second series of counts was taken.

Again, lead sheets 1.6 mm in thickness were placed over the body to permit selective counting of radiation originating in specific regions. Instead of exposing successive slits of constant width to the detector, the shape and size of the shielding was adjusted so as to expose portions of the body more nearly related to natural divisions or "compartments." These were (I) the head, five upper cervical vertebrae, and arms;

0016529

TABLE 8. NET COUNTS PER MINUTE IN 40- TO 72-keV BAND

Compartment	Count rate, cpm	% of total
I. Head and arms—supine	330 ± 4.6	13.5
Head and arms—prone	280 ± 4.4	11.2
II. Chest, ribs, shoulders—supine	1335 ± 8.5	55
Chest, ribs, shoulders—prone	1410 ± 8.5	56
III. Pelvis—supine	375 ± 4.3	15
Pelvis—prone	570 ± 6	22.5
IV. Legs and feet—supine	380 ± 4.4	15.8
Legs and feet—prone	250 ± 3.5	10.0
Sum I–IV—supine	2420 ± 11	
Sum I–IV—prone	2510 ± 11	
Whole body—supine	2370 ± 9	
Whole body—prone	2820 ± 10	

Uncertainties are standard deviation from counting statistics for counting times of 20 min.

(II) the shoulders, sternum, ribs, and spinal column down to the third lumbar vertebra; (III) the pelvis and remaining lumbar vertebrae; and (IV) the legs and feet. Care was taken to insure that the boundaries of each "compartment" were maintained uniquely, i.e., that no overlap occurred and every point in the body was counted only once. In view of the complexities of human anatomy, achievement of this idealized situation was only approximate. The crystal was fixed in position at 1 meter above the table and centered over the abdomen for all runs. Counts were made with the subject both supine and prone. The unique contribution of Am^{241} within each compartment was obtained by comparison with exactly similar data runs of an unexposed control subject of similar height, weight and *habitus*. The resulting net counting rates are given in Table 8.

The results appear to be consistent with the hypothesis advanced above, the large difference between prone and supine positions for compartment III suggesting that most of the activity was in the pelvic bone and vertebrae, with very little contribution from such possible soft-tissue sites as the liver or kidneys. An opposite relationship is noted in compartment IV, due to shielding of the long bones by calf and groin muscles.

DATA ANALYSIS

The metabolic behavior of the actinide series has been studied extensively in laboratory animals during the past two decades. Patterns of deposition and retention in various tissues following intramuscular injection, as well as modes of excretion, have been summarized by Durbin.[1] The physicochemical properties upon which the differing properties of the series depends are (1) oxidation state which is stable at the pH of body electrolytes, (2) solubility of the stable state, and (3) tendency to form organic compounds, in particular to be chelated by or bound into complex protein molecules or colloidal aggregates. The data suggest that Pu, Np, Pa and Th, having oxidation states of 4^+ or 5^+, are predominantly deposited within the skeleton, whereas the tripositive members of the group (Ac, Am, and Cm) appear to a greater extent in the liver. The biological behavior of the rare earths and of yttrium is also expected to shed light on these questions. The conference proceedings edited by Kyker and Anderson[2] summarize the results of many early studies. Herring, Vaughan, and Williamson[3] have emphasized the similarity in localization of yttrium, americium, and plutonium upon bone surfaces, i.e., those portions of bone most accessible from the vascular system, within a few days after injection, suggesting, however, that the mechanisms of binding may indeed be different. The conflicting evidence concerning yttrium was reviewed by Ramsden,[4] who emphasized the important role of mass concentration of injected material, which, by influencing aggregate formation and particle size, may well determine the amount of isotope found within the reticuloendothelial system.

Direct evidence on the deposition pattern of americium in man following exposure by inhalation is conspicuous by its rarity. In the present context, the large counting rates observed over the skull, knees, and shins appear to indicate predominant deposition upon bone surfaces (in violation of the systematics stated by Durbin), although an alternative bone marrow site cannot be ruled out. Unlike the usual situation involving animal experiments, there is no means of resolving the conflict by direct measurement. It should, however, be possible to estimate the *relative* surface areas available for americium uptake by each bone, and hence for each of the experimental compartments. A comparison of these with the corresponding observed disintegration rates would serve to indicate the credibility of the assumed model. Possible sources of discrepancy are (1) uncertainty in our knowledge of bone surface areas, (2) inability to evaluate the variables of counting geometry and self-absorption properly, and (3) additional activity within soft tissues. Deposition to the latter is expected to be significant only within the lungs, liver and spleen; hence, only compartments II and III are affected.

0016530

Estimates of the available bone surface have been made by two essentially independent methods. In the first, reported values of the trabecular and cortical components for each of the principal bones are used. Let M_i be the total mass of the ith bone, t_i and c_i the mass percentages of the two components. The surface area, s_i, available for deposition in trabecular bone per gram dry weight is assumed to be greater than that in the cortical portion by a factor K. Then

$$s_i = CM_i(Kt_i + c_i), \qquad (1)$$

where the proportionality constant C has the dimensions of surface area per gram and a numerical value which need not be evaluated since only relative numbers are required. The total area made available by each of the j compartments is then

$$S_j = \sum_i S_{ij} (j = 1, \cdots, 4) \qquad (2)$$

which, when normalized to total available area, becomes

$$R_j = \frac{S_j}{S_1 + S_2 + S_3 + S_4}. \qquad (3)$$

Values for the mass of the major human bones have been given by several authors, although without exception they have certain limitations. The data of Mechanik[5] and Trotter and Peterson,[6] while representing a sufficient number of individuals to give a meaningful mean, are weighted with those of advanced age or pathology for which the possibility of extensive bone resorption and remodelling cannot be overlooked. Ages ranged from 16 to 86 (Mechanik) and 25 to 97 (Trotter and Peterson) with means of 44.5 and 63 years respectively. Data of the latter reference are extensive enough (a total of 120 skeletons were measured) to permit calculation of mean regression with age and to thus obtain values more typical of the reference subject. Mechanik measured wet bone mass; Trotter and Peterson encountered difficulties in the consistency of such measurements (apparently because the source material originated in the anatomy laboratories of a medical school, and was subject to varying degrees of dehydration) and instead report dry, fat-free weight. Spiers[7] gives the dry weight of one set of bones of unspecified origin; the total mass of 3.4 kg appears significantly

TABLE 9. ANATOMICAL EVALUATION

Compartment	Average bone mass, M_i, g	% Trabecular component, t_i	Total trabecular surface, CKM_it_i,	Cortical surface, CM_ic_i	Total in compartment	% of total (skeletal) available
I						
Skull	720	10.0	288	648		
Radius, ulna, hands	370	12.5	185	324		
Humerii	300	24.1	289	227		
5 Cervical vertebrae	50	16.0	32	42		
Subtotal	1440		794	1241	2035	23.2
II						
Clavicle, scapula, sternum	200	10.0	80	180		
Ribs	580	51.0	1184	284		
Vertebrae						
2 Cervical	20	80.0	1088	68		
12 Thoracic	215					
3 Lumbar	105					
Subtotal	1120		2352	532	2884	32.8
III						
2 Lumbar vertebrae	70	16.0	45	59		
Pelvis	500	21.0	420	395		
Femur, head and neck	160[a]	100.0[a]	640	—		
Subtotal	730		1105	454	1559	17.8
IV						
Femur, shaft	640[a]	15.0[a]	400	540		
Tibiae	620	23.0	572	477		
Fibulae	100	10.0	40	90		
Feet	150	7.0	42	140		
Subtotal	1510		1054	1247	2301	26.2
Total	4.8 kg		5305	3474	8779	100.0

[a] Estimated division in mass and structure.

0016531

less than that of a normal, adult white male. Johnson[8] recognized these difficulties and lists values representing a small sample (3 to 5 bones of each kind) deliberately chosen to be typical of a well-developed, adult male. His values for dry, but not necessarily fat-free bones, were adopted for these calculations. Since only relative values are required by the analysis, it must be admitted that the other tabulations would probably be as acceptable. Values of t_i and c_i were also taken from Reference 8; in several instances these were not consistent with recent measurements by members of this Division and were adjusted accordingly. Perhaps the greatest uncertainty exists with the proper value of K. After considerable discussion with the more knowledgeable members of the Division, a value of 4 was assumed. Table 9 summarizes the resultant calculations of relative bone area.

A second estimate of bone surface in each compartment may be obtained by reference to the experimental data of Atherton, Mays, and Stover[9] on the deposition pattern of plutonium in the bones of adult beagles. To assume similar metabolic behavior for Am and Pu defies the commonly accepted differences in their uptake discussed above. Nevertheless it is tempting in view of the apparent uniform distribution observed, and may perhaps be justified since inhalation of americium from the vapor phase may well result in behavior quite different from that resulting from injection. While the partition of activity between skeleton and soft tissue is certainly dependent upon mode of ingestion, it seems reasonable to assume that distribution *within* the skeleton is substantially independent of that variable.

These authors list (Reference 9, Table III, p. 122) the activity of the principal bones or divisions of the skeleton, expressed as percent of total skeletal radionuclide divided by fraction of total ashed bone weight. It is convenient to think of this as percent of skeletal activity per gram bone (ashed weight) normalized to total skeletal (ashed) weight of 100 g. The data were averaged over four animals sacrificed at 1, 400, 499 and 610 days post-injection. Since it appears unlikely that for each bone the ratio ashed weight to wet weight (the value of which is certainly determined in part by the relative amounts of trabecular vs. cortical tissue) is the same for man and beagle, some hesitation may be felt in applying the data to man, although the errors resulting from the assumed relationship are probably no greater than those inherent in the data themselves. It did seem, however, preferable to convert the beagle data to percent skeletal activity per unit wet mass before combining with the dry, fat-free values for human bones. The calculations and resultant relative radioelement uptakes are summarized in Table 10.

TABLE 10. Bone Surface Radioactivity from Direct Measurements of Pu in Dogs

Compartment	Relative take-up of Pu[a]	Bone mass, g	Total bone activity, cpm	% of Whole body activity
I				
Skull	0.318[b]	720	229	
Radii, ulnae, and hands	0.306[b]	370	113	
Humerii	1.46	300	438	
5 Cervical vertebrae	1.07	50	53	
Subtotal		1440	933	19.0
II				
Clavicle, scapula	1.40	180	252	
Sternum	3.32	20	66	
Ribs	1.64	580	951	
2 Cervical vertebrae	1.07	20	21	
12 Thoracic vertebrae	2.72	215	585	
3 Lumbar vertebrae	1.84	105	193	
Subtotal		1120	2068	42.2
III				
2 Lumbar vertebrae	1.84	70	129	
Pelvis	1.50[b]	500	750	
Head and neck of femur	2.00[c]	160	320	
Subtotal		730	1199	24.5
IV				
Femur, shaft	0.50[c]	640	320	
Tibiae	0.48	620	298	
Fibulae	0.32	100	32	
Feet	0.34	150	51	
Subtotal		1510	701	14.3

[a] From Reference 9, p. 122.
[b] Weighted mean; original data divides this into two or more subdivisions.
[c] Adjusted mean.

Before comparing these predictions with the experimentally-observed distribution, the latter were corrected for counting efficiency as determined primarily by two variables: (1) source-detector geometry and (2) absorption of the 60-keV radiation by intervening body tissues. The first can be determined to the required precision by analytic means or by experimentation. The second factor is much more difficult to evaluate since it depends upon the detailed distribution of the source within the body—as yet undetermined. Intuition tells us that the attenuation in such disparate structures as the cranium, the rib cage and spine, or the long bones of the extremities will differ greatly. Since the calculations are based upon relative values, errors introduced by neglecting absorption losses altogether (as we have done) are proportional to the *difference* in mean attenuation of

0016532

TABLE 11. COMPARISON OF PREDICTED AND OBSERVED DISTRIBUTION OF RADIOACTIVITY IN THE BODY

Compartment	Mean count rate, cpm	Relative efficiency	Corrected count rate, cpm	Relative count rate	Predicted distribution of radioactivity	
					Method 1[a]	Method 2[b]
I. Head	305 ± 10	0.50	610 ± 20	19.5 ± 0.7	23.2	23.3
II. Chest	1370 ± 20	1.0	1370 ± 20	44 ± 0.85	32.8	40.2
III. Pelvis	470 ± 10	0.90	520 ± 10	16.6 ± 0.4	17.8	25.5
IV. Legs	315 ± 10	0.50	630 ± 20	20 ± 0.7	26.2	14.0
Total	2460 ± 26		3130 ± 40			

[a] Derived from Table 9.
[b] Derived from Table 10.

the various compartments, estimated to be of the order of 20–25% at most. The use of counting rates averaged over prone and supine body positions provides partial compensation.

With these qualifications in mind, the predicted and observed distributions are listed for comparison in Table 11.

DISCUSSION

While there is no a priori reason to expect that the calculations of bone area lead to better prediction of relative counting rates, they in fact do show a significantly closer correlation with the observed data. The greatest discrepancy occurs for the chest region, and this is consistent with the presence of additional deposits in the lung. If there is no contribution from soft tissues elsewhere, comparison of the relative count rates observed and predicted leads to an approximate allocation of activity as follows:

I. Head and arms	19.5% bone	none from soft tissue
II. Chest	28.0% bone	16% from soft tissue
III. Pelvis	16.6% bone	none from soft tissue
IV. Legs, feet	20.0% bone	none from soft tissue

In order to make an estimate of the total body activity, four Am^{241} sources of known (5%) activity were placed in a 21-cm thick* phantom composed of polyethylene sheets 56 cm by 34 cm in size, with a central region of reduced density. The location of the sources was varied until the ratio of net counting rates observed under the standard conditions as the phantom was turned over, to simulate the prone and supine positions of the patient, matched the corresponding ratio in the human subject. By direct proportion, the total body burden was estimated to be 1.8 μCi. Since this phantom in no way simulates the highly probable skeletal and lung distribution postulated nor the attenuation properties of bone and other intervening tissues, the figure is subject to considerable error. It does indicate a total burden in this individual approaching one hundred times the recommended MPL.[10] Reduction of the body burden by chelation therapy was promptly instituted elsewhere, and at the time of this report has been in progress for nearly a year. It is anticipated that similar whole-body measurements on the individual will be repeated soon.

* Equivalent to average thickness of patient's trunk.

Elizabeth Lloyd and John Marshall were of great assistance in frequent discussion and evaluation of the data on radioelement uptake and of bone calculations. The continued interest and encouragement of L. D. Marinelli is also deeply appreciated.

REFERENCES

1. Durbin, P. W. Metabolic characteristics within a chemical family. *Health Phys.* **2,** 225–238 (1960).
2. Kyker, G. C. and Anderson, E. B., Eds. Rare Earths in Biochemical and Medical Research: A conference sponsored by the Medical Division, Oak Ridge Institute of Nuclear Studies, October 1955. Report ORINS-12 (1956), unpublished.
3. Herring, G. M., Vaughan, J., and Williamson, M. Preliminary report on the site of localization and possible binding agent for yttrium, americium, and plutonium in cortical bone. *Health Phys.* **8,** 717–724 (1962).
4. Ramsden, E. N. A review of experimental work on radioyttrium. *Intern. J. Radiation Biol.* **3,** 399–410 (1961).
5. Mechanik, N. *Z. Ges. Anat.* **79,** 58–99 (1926).
6. Trotter, M. and Peterson, R. R. The relationship of ash weight and organic weight of human skeletons. *J. Bone Joint Surg.* **44A,** 669–681 (1962).
7. Spiers, F. W. and Burch, P. R. J. Measurements of body radioactivity in a radium worker. *Brit. J. Radiol.*, Proc. Conf. on Measurement of Body Radioactivity, Leeds, April 1956. **Suppl. 7,** 81–89 (1957).
8. Johnson, L. Morphologic analysis in pathology: The kinetics of disease and general biology of bone. *Bone Biodynamics*, Ed. Harold Frost. Little, Brown & Co., Boston, 1963, Ch. 29, pp. 543–654.
9. Atherton, D. R., Mays, C. W., and Stover, B. J. Radionuclide distribution in adult beagle bones. University of Utah Report COO-217 (September 1958), unpublished.
10. Report of Committee II on Permissible Dose for Internal Radiation (1959). *Health Phys.* **3,** 1–380 (June 1960).

0016533