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RADIOLOGICAL AND ENVIRONMENTAL RESEARCH DIVISION ANNUAL REPORT

Center for Human Radiobiology

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FOREWORD

This is the tenth Annual Report of the Center for Human Radiobiology. The Center now has two sizable populations under study: (1) radium dial painters and other persons with industrial or medical exposure to intake of radium, and (2) the former employees of a thorium-processing plant. Each population comprises about 4000 persons exposed to highly concentrated natural radioactivity. Although thorium has much lower specific activity than radium, the scientific purposes are the same for both studies: to determine the effects and relevant radiation doses of internally deposited alpha emitters in humans. However, there are great differences between the two studies.

Two serious effects of radium have been known for many years: bone cancer since the 1920's and carcinomas of head sinuses since the 1930's. Early recognition was aided by the low natural incidence of these malignancies and their relative insensitivity to other environmental factors. Since then, and especially since the early 1950's, much effort has been devoted to study of humans exposed to radium and to animal models, chiefly because of the hope of deriving valid risk estimates for the induction of bone cancer by those bêtes noires in fission-energy technology, ^{90}Sr and ^{239}Pu. This hope cannot be realized simply by counting numbers of bone sarcomas, because dogs are not humans, plutonium is not radium, alpha particles are not beta particles, and a few high-level radium cases are not millions of people exposed to low levels of radioactive materials. Hence, numerous reports appear in these annual reports relative to characterization of the cells at risk and radiation doses on a microscopic scale. As has been true of cancer research in general, there have been no sudden breakthroughs; progress sometimes consists mostly of greater recognition of complexities. Thus one has to compare work such as that in papers 5—12 with work being done 10 years ago to realize the progress that has been made toward characterizing the basic mechanisms, determining the relevant doses, and developing a theory that will indeed provide the means to extrapolate from bone cancer induced by high levels of radium to low doses of other radionuclides.

0019422

The immediate objective of the thorium studies is to help assess the health and safety impacts of possible increased use of thorium in fission energy technology (cf. paper 20). Although injected Thorotrast (ThO_2) is well known to cause liver cancer, leukemia, and possibly bone cancer, no serious health effects of industrial exposure to thorium have been reported. In thorium milling, the principal radiation exposures are due to airborne thorium and thoron and external radiation from thorium ores in the work place. Thus the metabolism of inhaled thorium (and daughter products), respiratory diseases, leukemia, and soft-tissue cancers are of greatest interest for study. The difficulties of separating thorium effects from the effects of other potential environmental hazards are great.

Up to the present, much of the effort in the thorium program has been devoted to acquisition and study of employment records, plant records of radiation exposure, and reports of regulatory compliance inspections. However, mortality and morbidity studies are underway, and almost 200 of the former thorium workers have been examined at Argonne. Surveys of immune status and chromosome changes are reported in papers 21 and 22.

Papers 1—4 refer to new work of a different nature. Observations of unusually large amounts of radon and radon daughters in some control subjects at the Center have led to a program of investigating airborne radioactivity in residential buildings. The purposes are to determine the existing average concentrations of radon and its daughters in houses, to evaluate the effects of energy conservation measures to reduce ingress of outside air during the heating and cooling seasons, and to determine the levels in new energy-efficient houses, such as those built underground, or those which use solar-heating systems. Evaluation of the risk to man of radon and its daughters is the underlying purpose.

0019423

TABLE OF CONTENTS

Center for Human Radiobiology

Foreword

1. Some Measurements of Radon and Its Daughters in Houses 1
   J. RUNDO, F. MARKUN, H. A. MAY, N. J. PLONDKE, AND D. J. KEEFE

2. Radon Daughters in the Occupants of a House with an Unusually High Concentration of Airborne Radon 13
   J. RUNDO, J. Y. SHA, AND R. E. TOOHEY

3. Radioactivity in the Furnace Air-Cleaning Filter from a House with an Unusually High Level of Airborne Radon 21
   J. RUNDO, M. A. ESSLING, AND P. W. URNEZIS

4. Elevated Radon Levels in Buildings: An Investigation of Potential Sources in Revion V Arising from Energy-Conserving Practices 31
   H. A. MAY

5. Cells at Risk for the Production of Bone Tumors in Man— An Electron Microscope Study of the Endosteal Surface of Control Bone and Bone from a Human Radium Case 39
   E. L. LLOYD AND C. B. HENNING

6. Inhibition In Vitro of the Growth of Cells Transformed by Alpha Radiation 54
   E. L. LLOYD AND M. A. GEMMELL

7. Suppression of Growth of Two Human Osteosarcoma Cell Lines by Normal Human Fibroblasts 55
   E. L. LLOYD AND M. A. GEMMELL

8. Suppression of B.P.-Transformed Foci in C3H 10T1/2 Cells by Retinyl Acetate 65
   E. L. LLOYD AND M. A. GEMMELL

9. Stripping of the Alpha Spectrum from a Bone with a Non-Uniform Depth Distribution of Radium 66
   L. F. MAUSNER AND R. A. SCHLENKER

10. Quantitative Histology of Mastoid Air Cell Epithelium 70
    M. J. HARRIS AND R. A. SCHLENKER

0019424

11. Dosimetry of Head Carcinomas in Radium Cases 76
R. A. SCHLENKER AND M. J. HARRIS

12. Toward a Theory of the Initiation of Cancer by Alpha, Beta, and X-Radiation: The Twin Doublet Model 85
JOHN H. MARSHALL AND ANTONIO PAGNAMENTA

13. Distribution of Plutonium Amongst and Within Selected Bones from an Injection Case 115
R. P. LARSEN, R. D. OLDHAM, AND S. M. MARTIN

14. The Concentration of Plutonium in Hair Following Intravenous Injection 120
R. E. TOOHEY, C. G. CACIC, R. D. OLDHAM, AND R. P. LARSEN

15. Urinary Excretion Rate of ^{241}Am Produced In Vivo 122
R. E. TOOHEY, R. E. WALKUP, R. P. LARSEN, AND R. D. OLDHAM

16. Progress Report on the Gastrointestinal Absorption of Plutonium in Mice 124
R. P. LARSEN, R. D. OLDHAM, M. H. BHATTACHARYYA, E. S. MORETTI, D. J. AUSTIN, AND C. G. CACIC

17. Plutonium in Drinking Water: Effects of Chlorination on Its Maximum Permissible Concentration 130
R. P. LARSEN AND R. D. OLDHAM

18. Application of Radiolead to Metabolic Studies 131
RICHARD B. HOLTZMAN

19. The Radioactive Properties and Biological Behavior of ^{224}Ra (ThX) and Its Daughters 133
J. RUNDO

20. A Study of Radioactivity and Health Status of Former Thorium Workers: Preliminary Report 135
J. RUNDO, A. P. POLEDNAK, A. M. BRUES, H. F. LUCAS, JR. B. C. PATTEN, R. E. ROWLAND, AND A. F. STEHNEY

21. Mitogenic Stimulation of Peripheral Lymphocytes of Thorium Workers 136
C. S. SERIO, C. B. HENNING, AND E. L. LLOYD

22. Chromosome Breakage in Peripheral Lymphocytes of Thorium Workers 143
S. F. HOEGERMAN AND H. T. CUMMINS

0019425

Appendix A. Exposure Data for Radium Patients 147

Appendix B. Radium-Induced Malignancies 213

Publications 220

0019426

RADON DAUGHTERS IN THE OCCUPANTS OF A HOUSE WITH AN UNUSUALLY HIGH CONCENTRATION OF AIRBORNE RADON*

J. Rundo, J. Y. Sha, and R. E. Toohey

The residents of a house which has high radon concentrations contains easily measurable levels of radon daughters (typically ∿ 400 Bq of ^{214}Bi). The results of the measurements are reported and some calculations of absorbed dose to lung, and of the growth of ^{210}Pb in vivo are presented. The relevance of the findings to the problem of assay of plutonium in lung is also discussed.

Introduction

The discovery that an employee of ANL (CHR case number 50-026) lives in a house where the concentration of radon commonly reaches or exceeds 750 Bq m^{-3} raised the question as to whether the occupants of the house might contain sufficient amounts of the short-lived daughters of radon to be detectable by external counting. The house is situated about 20 km from the Laboratory and it takes the employee about 35 minutes to drive to work. Although the daughters of radon have half-lives which are less than this, the activity due to ^{214}Bi decreases more slowly because its decay is countered by production from the decay of ^{214}Pb and (to a lesser extent) of ^{218}Po, which were present initially. The content of ^{214}Bi in a mixture consisting initially of equal activities of the three, is reduced to half in about 1 hr. It is the 1.76 MeV gamma ray from ^{214}Bi which affords our most sensitive measure of the body content of radon daughters. On separate occasions we have made body radioactivity measurements of the three residents of the house, i.e., the employee, her spouse, and their baby.

Procedure

On arrival at ANL each of the adult subjects went immediately to the underground vault, quickly changed into clean clothing and was seated in the tilted chair. For both subjects, gamma-ray counting was started less than 1 hr after

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0019427

they had left the house. Several consecutive 10-min observations were followed by consecutive 20-min ones, with one- or two-min intervals for data readout, for about 1.5 hr. Less frequent 20-min observations were continued for the next 6 to 7 hr. Background measurements were taken at intervals during the day. On one occasion two series of measurements were made with a 180-mm diameter proportional counter presented alternately to the chest and hair of subject 50-026, mainly to determine the influence of radon daughters in the thorax on the determination of plutonium in vivo.

The baby (male, age 25 days, weight 3.6 kg) was taken to the vault, changed into clean clothes that had been stored at ANL for some days, and placed on a bed between two 29-cm diameter detectors, one 15.5 cm above the baby's chest and the other 10 cm below the bed. Gamma-ray counting was started 34 min after the infant left the house, and five consecutive 10-min observations were made.

Results

The gamma-ray spectra from the adults showed easily measurable levels of ^{214}Bi. Counts observed above background in the energy band covering the 1.76 MeV peak were used to estimate the body content of ^{214}Bi. We used the average calibration factor that is used to calculate the skeletal content of this nuclide in radium-exposed subjects. This procedure gave values for ^{214}Bi in soft tissues and blood which were approximately correct, but over-estimated the possible thoracic content by a factor of 1.6.

The estimates of the body content of subject 50-026 are plotted on a logarithmic scale as a function of time on a linear scale in Figure 1. An initial rapid decline was followed by a much slower one. We interpret this as a consequence of the decay of radon daughters unsupported by ^{222}Rn, followed by the decay of daughters at a rate controlled by exhalation of radon transported to the lung from a supply stored in body fat and fluids. During the intervals between the later body radioactivity measurements the rate of exhalation of radon was determined. In the period 150 to 265 min after leaving the house, the mean exhalation rate was 1.93 mBq s^{-1} (range 1.73-2.16 mBq s^{-2}), and this declined to

0019428

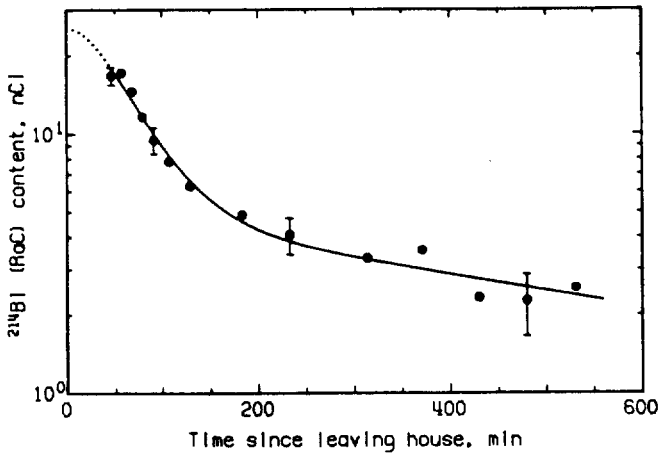

FIG. 1.--Approximate ^{214}Bi content of subject 50-026 on May 17, 1978 as a function of time since leaving a house containing a high level of radon, showing the decay of radon daughters unsupported by radon, and a residual level due to radon dissolved in body fat and fluids.

a mean value of 1.46 Bq s^{-1} (range 1.23-1.60 mBq s^{-1}) in the period 450 to 545 min after leaving the house. If the decrease was exponential it corresponded to a biological half-life of 760 ± 190 min, and an exhalation rate of 1.46 mBq s^{-1} would then imply a body content of 96 Bq (2.6 nCi) of radon at about 500 min. This is in reasonable agreement with the mean of the last three observations of the body content of ^{214}Bi of 88 Bq (2.38 nCi).

We now make the further assumptions that the unsupported daughters were all in the lung, and that at the time the subject left the house they were present at equal activities. An equation describing the behaviour of the activity of ^{214}Bi in a mixture consisting initially of equal activities of the three short-lived daughters, plus a single exponential term, was fitted to the data plotted in Figure 1 by the method of least squares.

The fitted equation is represented by the smooth continuous curve drawn through the data points in Figure 1, and the extrapolation back to the time of leaving the house is shown as a dotted curve. The intercept at that time was 940 ± 90 Bq (25.4 nCi) of which 750 ± 80 Bq represented the level of unsupported daughters and 190 ± 40 Bq was the intercept of a component with an assumed exponential decline at a rate corresponding to a half-life of 483 ± 207 min. This is somewhat lower than, but still reasonably consistent (in view of the large uncertainties) with, the value of 760 ± 190 min derived from the exhalation rate of radon over the same period.

As noted above, the value of 750 Bq for the activity of the unsupported daughters was an over-estimate if they were confined to the thoracic region, and

a more likely value was 470 Bq.

Before the data plotted in Figure 1 were collected, two other series of much less complete measruements were made. On May 9, a content of 360 Bq was observed at 54 min after leaving the house; this was about 56% of what was observed on May 17 at about the same time. On May 10, a content of 280 Bq was observed at 133 min, and this was 20% higher than was observed on May 17 at the same time. Thus this subject has been found to contain easily measurable amounts of ^{214}Bi on three occasions during a period of eight days. It may be noted that a measurement of her body radioactivity was made in January 1976 before she moved into her present residence. On that occasion the ^{214}Bi content was found to be 33 ± 19 Bq, and the rate of exhalation of radon was in the range 0.09 ± 0.12 mBq s^{-1}. These results demonstrated that the subject had no significant radium content and the present results can be attributed entirely to the high radon content of her house.

The measurements with the proportional counter were also of considerable interest; the results of the chest measurements are plotted in Figure 2. A single exponential function of time was fitted to the data by the method of least squares, and a half-time of 46 ± 14 min was obtained, as represented by the straight line. This is quite typical for the apparent half-life of the activity of radon daughters. On the other hand, the counting rates observed from the hair decreased faster, and the half-time obtained by least squares analysis was 26 ± 4 min. This is because under these conditions of minimum photon scattering, the proportional

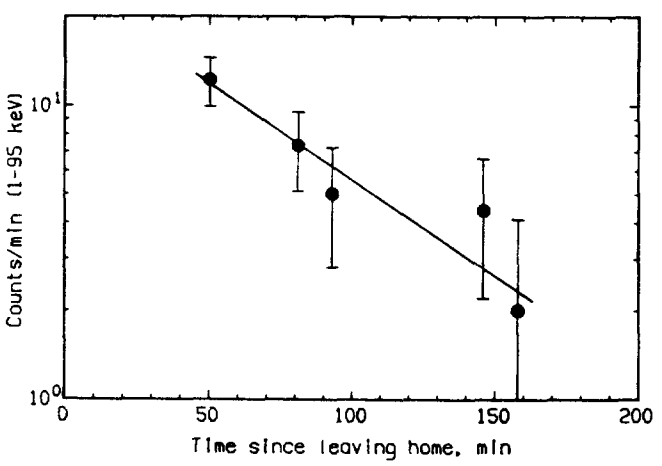

FIG. 2.--Decrease of counting rate from the chest of subject 50-026 as observed with a proportional counter.

0019430

counter responded primarily to the L x rays of bismuth emitted following the decay of ^{214}Pb (half-life 26.8 min) and hardly at all to the gamma rays emitted by ^{214}Pb and ^{214}Bi. On the other hand, when presented to the chest, the counter was detecting multiply-scattered gamma rays from these nuclides. The important point to note is that the initial counting rate from the chest might have been interpreted as due to ten times the maximum permissible lung burden of ^{239}Pu if the subject had been under investigation for that nuclide. It should also be noted that even two hours after leaving home, the counting rate was still about 33% of the rate at 50 min, corresponding to the response from three times the maximum permissible lung burden of ^{239}Pu.

Body radioactivity measurements of the employee's husband (CHR case number 50-150) were made in August 1978 and again in January 1979. The two sets of results were almost identical. A least squares fit of the equation used for case 50-026 to the first set of data yielded an intercept of 640 ± 85 Bq ^{214}Bi, of which 560 ± 80 Bq were unsupported by radon. The long-term component was at a lower level than was the case for his wife (80 ± 30 Bq initially) and the half-life was apparently much longer (and indeterminate). For reasons already explained, the result for the activity of the unsupported ^{214}Bi was too high if the daughter-products were confined to the thoracic region, and a more likely value is 350 Bq. We showed that the radium content of this subject was also not significant by making a measurement of his body radioactivity on his return from a short vacation and before he had been home.

The results of the measurements of radioactivity in the baby were not as unequivocal as those for the adults in demonstrating the presence of radon daughters. The principal gamma-ray emitter seen in the spectra was ^{40}K, but there were strong indications of ^{214}Bi. Thus, the net counting rate in the region of the 1.76-MeV total absorption peak was highly significant (12.7 ± 2.6 min^{-1} in the first measurement), and it decreased with time. Furthermore, a peak at 0.6 MeV could be clearly seen in the spectra and the counting rate in the region 535 to 695 keV decreased from 49.6 ± 4.9 min^{-1} at 39 min to 36.6 ± 4.8 min^{-1} at 86 min after leaving the house. It was estimated that no more than 23 counts min^{-1} could be attributed to ^{40}K (and perhaps also to fallout ^{137}Cs). No calibration

measurements were made, as the investigation was considered to be an exploratory one.

Discussion

Apparently the two adults maintain a considerable body content of radon daughters while they are in the house during heating or air conditioning when the windows are closed. With the assumption that the daughters which are not supported by radon are deposited uniformly throughout the lung, we have calculated the rate of absorbed dose. In making the calculation, we used values of 1000 g and 800 g for the masses of the lungs for the male and female subjects, respectively. For subject 50-026 (female) the dose-rate due to the alpha particles from equal activities of the three short-lived daughters maintained at 470 Bq each (from the measurements on May 17, 1978), was 40 mGy yr^{-1} (4 rad/yr); for subject 50-150 (male), the dose-rate was 24 mGy yr^{-1} (2.4 rad/yr) due to a lung content of 350 Bq of each of the daughters (from the measurements on August 30, 1978). These dose rates are an order of magnitude higher than those deduced from the observations with the Environmental Working Level Monitor, reported in the preceding contribution to this report. No data on the airborne concentrations of the radon daughters in the house are available for the dates when the measurements in vivo were made, and only a small amount of data on the radon concentrations. The radon concentration at 9:15 p.m. on May 16, 1978 was only 200 Bq m^{-3} (5.4 pCi/ℓ); if this was constant until subject 50-026 left the house on the morning of May 17, and if the equilibrium factor, F, was 0.1 (as discussed in the preceding contribution) then the concentration of the daughters must have been about 0.005 WL. The corresponding dose rate would have been 0.13 rad/yr (1.3 mGy/yr), from the equivalence of 0.5 rad (5 mGy) per WLM (i.e., exposure to 1 WL for 170 hr).[1] For the case of subject 50-150 (measured on August 30, 1978), radon concentrations of 500 Bq m^{-3} and 650 Bq m^{-3} were deduced at 9:30 p.m. on August 29 and at 7:15 a.m. on August 30, respectively. With the same assumptions as above, the latter value would indicate a radon daughter concentration of 0.018 WL, and a dose rate of 0.47 rad/yr (4.7 mGy yr^{-1}). This discrepancy is not completely understood, although it should

0019432

be noted that the dose derived from the Working Levels is to the bronchial epithelium, whereas our values are calculated as average doses to the whole lung. Furthermore, although the BEIR Committee adopted a value of 0.5 rad per WLM, they commented[1] that estimates have ranged from less than 0.1 rad to as much as 20 rad per WLM. All these estimates are based on lung models of varying degrees of complexity and hardly at all on experimental data, but it seems to be generally accepted that the alveolar content of radon daughters is substantially greater than the deposit on the bronchial epithelium.

It must be emphasized that the dose rates that we have calculated have to be reduced by the occupancy factor and perhaps also by the fraction of the total time that the windows are kept closed. The two subjects made realistic estimates of their occupancy factors and these were 0.50 and 0.37 for 50-026 and 50-150, respectively. The dose-rates would then become 20 mGy yr^{-1} (2 rad/yr) and 8.9 mGy yr^{-1} (0.89 rad/yr) for the two subjects, respectively.

Another consequence of the maintenance of $\sim$ 400 Bq ^{214}Bi in vivo is the production of 20.4-year ^{210}Pb. The rate of production of this nuclide from 400 Bq ^{214}Bi is 37 mBq day^{-1}; this may be compared with the "normal" average intake to the blood of about 6 mBq day^{-1} (derived from data in Ref. 2, pp. 91 and 94). This is only one-sixth of the production in subjects 50-026 and 50-150 although it must be noted that the value of 37 mBq day^{-1} has to be multiplied by the appropriate occupancy factor to obtain the true production rate in vivo. Nevertheless, it is clear that the body content of ^{210}Po will increase in each of these subjects. After residence in this house for twenty years it is estimated that their contents will approach 160 Bq and 80 Bq (for 50-026 and 50-150, respectively) in addition to the normal content of about 20 Bq (derived from data in Ref. 2, pp. 98-100).

Finally, we note that the detection of the scattered gamma rays from ^{214}Pb and ^{214}Bi by the proportional counter, and the possible interpretation as uranium L x rays from a lung burden of plutonium, may be the explanation for some of the poor reproducibility in assaying small but significant amounts of plutonium in vivo, and for some of the variability in the determination of "subject background" in those studies (see various papers in Ref. 3).

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0019434

DISTRIBUTION OF PLUTONIUM AMONGST AND WITHIN SELECTED BONES FROM AN INJECTION CASE

R. P. Larsen, R. D. Oldham, and S. M. Martin*

The burden and macrodistribution of plutonium in the skeleton of a man who received 0.38 μCi of ^{239}Pu by injection are being determined. The relative concentrations in the bones that have been analyzed are quite comparable to those obtained in a study of another subject. The concentrations in the trabecular and cortical portions of these bones have been determined; the concentration ratio ranged from 1.9 to 4.7. The data show that (1) within a bone the plutonium concentration in the trabecular portion is always higher than it is in the cortical portion, but (2) within a group of bones plutonium concentration is not correlated with degree of trabecularity.

Introduction

The burden and macrodistribution of plutonium are being determined in the skeletal remains of a 65-year-old man who received 0.38 nCi of ^{239}Pu by injection in 1945. At the time of the injection he was suffering from muscular atrophy and dermatitis; the cause of death (1947) was bronchopneumonia. The plutonium he received was tetravalent and in a citrate buffer. The subject, referred to in the early literature as HP 9, has been assigned the CHR case number 40-015. The major thrust of this investigation is the same as was carried out on the skeletal remains of Case 40-010, the results of which have been reported.[1] The similarities of the two cases are that the injected plutonium was in the same chemical form and both individuals succumbed about 500 days after the time of injection. The dissimilarities are age, sex, and the nature of illness. Case 40-010 was a 20-year-old female suffering from Cushing's syndrome.

The skeleton was intact at the time of exhumation and was free of soft tissue. It was slightly moist owing to a thin dark coating of a material that appeared to be organic in nature. Some of the highly trabecular bones had begun to soften and decay. The radiograms showed that there was some post-

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0019435

mortem lysis in these bones.

Sampling and Analytical Methodology

The sampling was comparable to that used for 40-010, the principal exception being that some additional samples were taken in this case to establish the symmetry of plutonium deposition. The same analytical methodology is being used to determine the plutonium concentrations, and the same quality control system is being employed to ensure the accuracy of the data.

For the study of the relationship between plutonium concentration and bone type, the trabecular and cortical portions of a number of bones were separated and these portions were analyzed separately. After the bones had been ashed at 600°C, they were divided into a trabecular portion (material that could be removed from the inner surface of a bone with a minimum of force) and a cortical portion (material which remained after the inner surface had been forcefully scraped).

Results and Discussion

Distribution in the Skeleton

The distribution of plutonium in those bones of 40-015 that have been analyzed is given in Table 1 and it is compared with that in the same bones of 40-010. The concentrations are relative to those in the respective humerus midshafts.

It is apparent from the data in Table 1 that the distributions are very similar. In the earlier discussion of the data for 40-010,[1] it was noted that the observed distribution might not be representative because of her illness and/or her age. Cushing's syndrome is characterized in part by osteoporosis, but at age 21 her skeleton could still have been metabolically very active. These factors do not appear to have affected the distribution of plutonium significantly.

Distribution within Bones

A general conclusion drawn from the data obtained in the analysis of 40-010 was that the highest concentrations of plutonium were in those bones that are primarily trabecular.[1] It has since been noted that there were several

0019436

Table 1. Skeletal Distribution of Injected Plutonium

Bone	Relative concentration, bone/humerus midshaft	
	Case 40-010	Case 40-015
Humerus - distal end	0.93	1.09
Femur - head	2.75, 6.40[a]	2.11
Rib - 2	10.2	10.2
Rib - 8	11.2	8.9
Sternum	26.0	18.0
Vertebrae, cervical, body	19.2	19.3
Vertebrae, thoracic, body	23.6	29.8
Tarsals	1.42	1.09

[a]These values are for the right and left femur heads, respectively.

exceptions. The average concentration in the ribs, which are primarily cortical bone, was a factor of 2.1 higher than the average concentration for the entire skeleton and was the same as that of the femur head, which is primarily trabecular bone. In contrast, the concentration in the tarsals, which are primarily trabecular bone, was a factor of about 4 lower than the skeletal average, and was about the same as in the femur diaphysis, which is primarily cortical bone.

The distribution of plutonium in the trabecular and cortical portions of selected bones of 40-015 is given in Table 2. The concentrations are relative to that in the cortical bone from the femur midshaft.

It is evident from these data that the generalization cited above (relating plutonium concentration to degree of trabecularity) is valid only when comparing one part of a bone with another. The distribution of plutonium within the skeleton would appear to be related to the general metabolic activity level of individual bones, not bone-to-bone differences in composition. For those bones which are the most active metabolically, the concentrations of plutonium in both the trabecular and cortical portions are high relative to the skeletal average.

A comparison has been made of the trabecular to cortical plutonium concentration ratios for the femur head and thoracic vertebrae with the corresponding surface area ratios, which Lloyd and Hodges[2] measured using computer analysis

0019437

Table 2. Plutonium Concentrations in the Cortical and Trabecular Portions of Selected Bones from Case 40-015.

Bone[b]		Relative concentration[a] Trabecular	Cortical	Concentration ratio, trabecular/cortical
Femur shaft	(<10)	4.5	(1.00)	4.5
Femur head	(>75)	2.7	1.19	2.3
Humerus shaft	(<10)	2.9	1.12	2.6
Tarsal	(>90)	1.19	c	
Clavicle	(50)	3.3	0.92	3.6
Rib 2	(<10)	25	7.1	3.5
Rib 8	(<10)	29	6.1	4.8
Sternum	(>75)	39	9.1	4.3
Vertebral body				
Cervical	(>75)	31	13.3	2.3
Thoracic	(>75)	43	22.2	1.9

[a]Concentrations are relative to that in the femur shaft.

[b]Parenthetic values are the percentages of trabecular bone.

[c]No cortical portion could be isolated.

of microradiographs. This shows that there is a relationship between the extent of plutonium deposition and area for a particular bone. The trabecular to cortical concentration ratios for the femur head and thoracic vertebrae were 2.3 and 1.9, respectively, while the corresponding area ratios were 3.3 and 4.0. For the 10 bones we analyzed, the mean of the concentration ratios was 3.3 ± 1.0; for the 4 bones analyzed by Lloyd and Hodges the mean of the area ratios was 4 ± 1.

There are two general conclusions that can be drawn from this part of the study of plutonium distribution: (1) the concentration of plutonium will always be higher in the trabecular portion of a particular bone than it is in the cortical portion because the surface area of the former is always higher than that of the latter, and (2) the highest concentrations of plutonium in both trabecular and cortical bone are in those bones that have the highest metabolic activity levels.

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0019438

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A STUDY OF RADIOACTIVITY AND HEALTH STATUS OF FORMER THORIUM WORKERS: PRELIMINARY REPORT*

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Studies have been started of the late effects of thorium on the health of a population of 4316 persons (80% males) who worked at a thorium refinery during the period from the mid-1930's to 1973. The population and the study are described briefly, as are the methods and results to date of measurements of radioactivity in vivo and in the exhaled breath. Of 46 men examined so far, all but three had readily measurable amounts of thoron in the breath, but there was little correlation between the amount of activity in the breath and the ^{212}Bi content of the chest.

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