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RADIOLOGICAL AND ENVIRONMENTAL RESEARCH DIVISION ANNUAL REPORT

Center for Human Radiobiology

July 1971–June 1972

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FOREWORD

This is the third Annual Report to be published by the Center for Human Radiobiology.

The many articles dealing with current studies on radium patients reflect the fact that our contact and examination procedures for living patients became fully operational at Argonne National Laboratory during the past year.

Regularly scheduled examinations of radium patients were started at Argonne in July of 1971, and 159 patients had been examined by 30 June, 1972. Each examination consisted of a comprehensive physical examination, gamma-ray and radon-breath measurements of body radioactivity, measurements of bone mineral mass for evidence of accelerated bone resorption, and examination of chromosomes from peripheral blood leukocytes. The case load was increased during the year from 2 persons per week to 6 per week.

Examinations of radium patients living in the eastern part of the United States continued at the Radioactivity Center of the Massachusetts Institute of Technology, and 83 persons were examined at that laboratory from 1 July, 1971 to 30 June, 1972.

Facilities were also developed and personnel acquired during the past year for dissection of human remains and preparation of bone sections. Work is now under way on examination of exhumed or willed bodies obtained with the help of personnel at the Radioactivity Center. This program is directed toward estimating body burdens for radium patients who died prior to measurement or adding to previous information by medical, chemical, and microscopic analyses of tissues. Examination of bone tumors is made for each case.

The reader's attention is directed to three important articles: "A Preliminary Comparison of the Carcinogenesis of ^{226}Ra and ^{228}Ra in Man" by R. E. Rowland, A. T. Keane, and H. F. Lucas, Jr.; "The Effect of

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Remodeling of Bone upon the Relative Toxicities of Radium and Plutonium in Man and Dog" by J. H. Marshall and E. L. Lloyd; and "Alkaline Earth Metabolism in Adult Man" by J. H. Marshall et al. Only abstracts of these papers are given in this report because the complete papers are being published elsewhere in the scientific literature.

Tables of "Exposure Data for Radium Patients" to 31 December, 1971, are given at the back of this report.

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THE RATE OF BONE LOSS WITH INCREASING AGE: A COMPARISON OF RADIUM CASES WITH UNEXPOSED INDIVIDUALS

Robert A. Schlenker, Billie G. Oltman, and Barbara A. Roth*

Bone scan data on female radium cases and on normal women, who have had no exposure to radium, have been analyzed and compared. For women over 40 years of age, it was found that the rate at which the mineral mass per unit area in compact bone diminishes with age is significantly greater in radium cases than in normals. Furthermore, the rate in radium cases is significantly higher when the ^{226}Ra body burden is greater than 0.1 μCi, than when it is less than 0.1 μCi.

Introduction

As part of the radium study, bone scan measurements have been routinely made since July 1971 on radium cases who come to CHR for examination. Through June 30, 1972, 10 male and 144 female cases were measured. Analysis of the data collected through June 30, 1972 is reported here. The emphasis of the analysis is on the comparison between the rate of demineralization with age in radium cases and in individuals who have had no exposure to radium ("normals"). The measure of mineralization employed is the mineral mass per unit area of bone. Only data on female radium cases have been analyzed, the measurements on male cases having been too few to permit analysis in depth.

Three sets of data on normals were used in the analyses: those of J. R. Cameron,[1] J. O. Johnston and N. F. Goldsmith,[2] and C. C. Johnston.[3] Only data on normal females were used, and all of the normals and radium cases were Caucasian.

Bone Scan Measurements

The bone scan apparatus is of the type described by Cameron and Sorenson.[4] It measures the transmission of a narrow, collimated, nearly monochromatic beam of x rays as a function of position across a bone. The

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transmission data are currently analyzed to provide two bone parameters, the mineral mass per unit length of bone often just called mineral mass, and the bone width. The mineral mass in the x-ray beam pathway is computed by comparing the measured beam attenuation with the attenuation in a calibration bone of known mineral mass. The bone width is the distance between the points at which the x-ray beam is first attenuated by and ceases to be attenuated by the bone. The analysis of the data taken at CHR has been automated and the relevant computer programs have been described by Strash.[5]

Mineral mass and width in compact bone are strongly correlated with a linear correlation coefficient of approximately 0.9.[6] Within a sample of people of similar ages on whom scans of compact bone have been made, the coefficient of variation in the ratio of mineral mass to width, i.e., in the mineral mass per unit area, is often smaller than the coefficient of variation in mineral mass.[7] (Hereafter, mineral mass per unit area is called sigma.) In cancellous bone, the coefficient of variation of sigma is approximately equal to the coefficient of variation in mineral mass. Because of the greater sensitivity of sigma in compact bone to departures from normal, sigma is adopted as the measure of bone mineralization in both compact and cancellous bone for the analyses of data in this report.

Bone scan measurements have been made at three skeletal sites in the radium subjects: (a) across the right arm, one-third of the distance from the styloid process of the ulna to the proximal end of the ulna; (b) across the right arm, 2 cm proximal to the tip of the styloid process of the ulna; (c) across the distal end of the proximal phalanx of the middle finger of the right hand at the third major skin wrinkle proximal from the interphalangeal joint. The scans at sites (a) and (b) provide sigma values for both the radius and ulna. Only data on the radius are analyzed here. For ease of reference, sites (a), (b), and (c) are dubbed radial midshaft, distal radius, and phalanx, respectively. The radial midshaft site provides a scan across compact bone, while the distal radius and phalanx sites provide scans

across cancellous bone. Both compact and cancellous sites were scanned to determine whether or not differences existed in the rate of demineralization with age in these two types of bone.

Not all radium cases have been scanned at both compact and cancellous sites, and when a scan of cancellous bone has been made, it has been made at only one of the two cancellous sites. A summary of the number of cases scanned at the three sites is presented below:

Sex	Radial midshaft	Distal radius	Phalanx
Female	144	37	66
Male	10	2	5

Sigma values for normals were available for right and left radii at scan locations similar to the radial midshaft and distal radius locations. No data on normals were available for comparison with the phalanx scan. The specific scan locations for the normals, quoted from the investigators who report normal data are:

1. Radial midshaft
   (a) J. R. Cameron—left arm, "one-third of the distance from the distal to the proximal end of the radius"[6]
   (b) C. C. Johnston—right arm "8 cm from the distal end" of the radius.[8]
2. Distal radius
   (a) J. O. Johnston and N. F. Goldsmith—left arm, "two centimeters proximal to the tip of the styloid process of the ulna"[9] and
   (b) C. C. Johnston—right arm, "3 cm from the styloid process" of the radius.[8]

In order to compare the position of the radial midshaft and distal radius scans in radium cases with those in normals, the scan locations were translated into centimeter distances measured along the arm from the tip of the styloid process of the ulna. The distal radius scan location in radium cases is already referenced to the styloid process of the ulna; i.e., it lies

2 cm proximal to it. The radial midshaft location in radium cases may be determined from the length of the ulna. The maximum lengths of ulnae in a group of deceased female radium cases averaged 24.0 ± 1.4 cm.[10] One-third of this distance is 8.0 ± 0.5 cm. The uncertainty in these numbers represents one standard deviation so that if the distribution of ulna lengths is the same in living and deceased radium cases, and the lengths are normally distributed, about 95% of all radial midshaft scans are made at locations which fall between 7 cm and 9 cm from the styloid process of the ulna. The location for J. R. Cameron's normals may be determined from the length of the radius and the relative placement of the radius and ulna. The lengths of radii in a group of deceased females averaged 22.2 ± 1.2 cm.[11] One-third of this distance is 7.4 ± 0.4 cm. If the lengths of radii in the deceased females and in Cameron's female subjects were similarly distributed, his scan location lay 7.4 ± 0.4 cm from the styloid process of the radius. The radius is offset distally with respect to the ulna by about 1 cm so that Cameron's scan location was 6.4 ± 0.4 cm from the styloid process of the ulna. The uncertainty in this number is one standard deviation so that 95% of Cameron's scans were between 5.6 cm and 7.2 cm from the styloid process of the ulna. The distal radius location described by J. O. Johnston and N. F. Goldsmith is already referenced to the styloid process of the ulna; i.e., it lies 2 cm proximal to it. C. C. Johnston's scan locations are both referenced to the radius rather than the ulna. If it is assumed that the radius and ulna are offset by 1 cm, Johnston's distal radius and radial midshaft locations are 2 cm and 7 cm from the styloid process of the ulna. A summary of scan locations, referenced to the tip of the styloid process of the ulna for the radial midshaft and distal radius positions, is presented on the following page for ease of comparison.

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	Radium cases	Normals: Cameron[1]	Normals: Johnston, Goldsmith[2]	Normals: Johnston[3]
Radial midshaft	7-9 cm	5.6-7.2 cm	--	7 cm
Distal radius	2 cm	--	2 cm	2 cm
Phalanx	3rd major skin wrinkle proximal to interphalangeal joint	--	--	--

This summary makes it clear that the radial midshaft scans for the two groups of normals and for the radium cases were made at substantially different locations. In order to compare radial midshaft demineralization in radium cases with that in normals, it is necessary to estimate how sigma varies with scan location. Figure 1 presents data on the variations of sigma with position in the right radius of a 30-year-old normal white male. The vertical axis gives the value of sigma normalized to 0.896. The latter value was observed in the scan made at one-third the distance from the styloid process to the proximal end of the ulna. The horizontal axis gives distance either side of the one-third point. The one-third point lies 9.7 cm from the styloid process of the ulna so that the graph represents the range 7.7 to 13.7 cm from the styloid process of the ulna. The error bars represent one standard deviation. The differences between pairs of sigma values are two standard deviations or less. Thus, the differences are, at worst, on the borderline of statistical significance at the 5% level. The data are consistent with the hypothesis that sigma is constant over the range of scan locations presented on the graph. Unfortunately, no information is available for this report on scan locations less than 7.7 cm from the styloid process of the ulna. The fact that these scan data refer to a 30-year-old male and the radium subjects and normals for which comparisons are made in the data analyses which follow are females between about 40 and 90 years of age weakens any conclusions which may be drawn from these data. However, with these data offered in support, it is assumed that the value of sigma at the radial midshaft of an individual is constant over the range

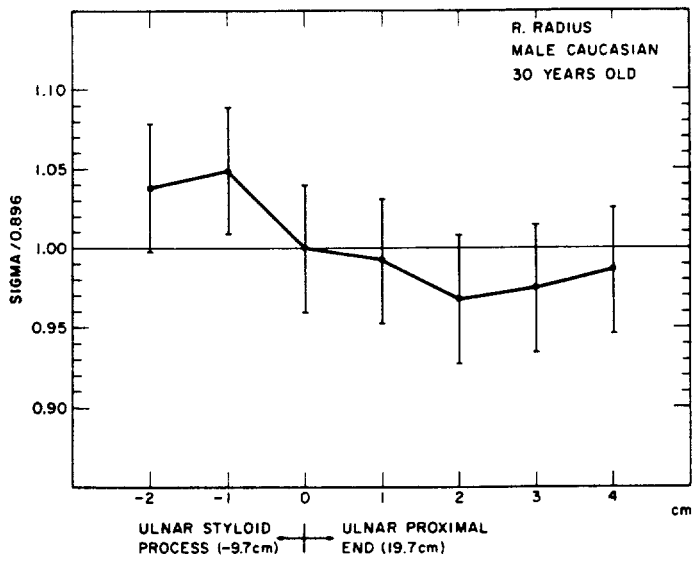

FIG. 1.--This graph presents sigma/0.896 as a function of distance along the radius of a 30-year-old Caucasian male. The zero on the horizontal axis marks the point on the radius which lies 1/3 the distance from the styloid process of the ulna to the proximal end of the ulna. For this person the point lies 9.7 cm from the styloid process.

of scan locations encountered in radium cases and normals.

Data Analysis

Gradual demineralization of the skeleton, beginning in middle age and continuing to the end of life, is a well documented phenomenon in humans. The main purpose of the data analysis is to determine whether or not female radium cases and female normals differ in the rate of demineralization with age. The measure of demineralization employed was sigma measured at the scan locations discussed previously. The rate of decrease of sigma with age was determined by least square fitting straight lines of the form sigma = A x (years age) + B, to sigma values of radium cases and normals who were 40 years of age or older. The slope of the straight line gave the rate of change of sigma with age.

Tabulations of normal data give the mean values of sigma and age for decade age groupings, e.g., 40 to 49. In order that treatment of the data be the same as for normals, the radium cases were grouped into identical intervals and average values of sigma and age were computed for each decade. Five decades, 40 to 49 through 80 to 89, were represented in the radium case data; six decades, 40 to 49 through 90 to 99 in the data of J. R. Cameron and of J. O. Johnston and N. F. Goldsmith, and 41 to 50 through 91 to 100

in the data of C. C. Johnston. Lines were fitted to the mean sigma as a function of mean age; standard errors of mean sigma values were used as weights in the fitting procedures. The values of the straight line coefficients are presented in Table 1 for each of the scan locations in radium subjects and normals. Data for each of the scan locations are plotted in Figures 2-4.

TABLE 1. Coefficients of Least Square Fit Straight Line, Sigma = Ax (years of age) + B, for Each Location

	Normals		Radium cases (CHR)	
Location	A, g/cm^2 per year x 10^{-3}	B, g/cm^2	A, g/cm^2 per year x 10^{-3}	B, g/cm^2
Radial midshaft	-6.09 ± 0.31 (1) -5.69 ± 0.46 (3)	1.03 ± 0.02 (1) 1.00 ± 0.03 (3)	-7.87 ± 0.82	1.14 ± 0.04
Distal radius	-3.58 ± 0.28 (2) -2.96 ± 0.41 (3)	0.63 ± 0.01 (2) 0.63 ± 0.03 (3)	-4.02 ± 1.64	0.67 ± 0.09
Phalanx			-5.00 ± 0.87	0.68 ± 0.05

The least square fit lines for radium cases and normals are plotted on these graphs for the age ranges over which least square fitting was carried out. More significant digits were retained in the straight line parameters for the purposes of plotting the graph than are quoted in Table 1. Thus, slight differences may exist between the plotted lines and the lines of Table 1. The goodness of fit of the straight lines was evaluated using the χ^2 test. The χ^2 test failed to reveal any discrepancies between the data and the best fit lines, which were statistically significant at the 5% level. An estimate of the error introduced by grouping the data was made by fitting lines to the ungrouped radium case data. Line slopes differed by 1 to 2% between the grouped and ungrouped data, and standard errors in the line slopes differed by 3 to 5%.

Pairs of line slopes were compared with one another to determine

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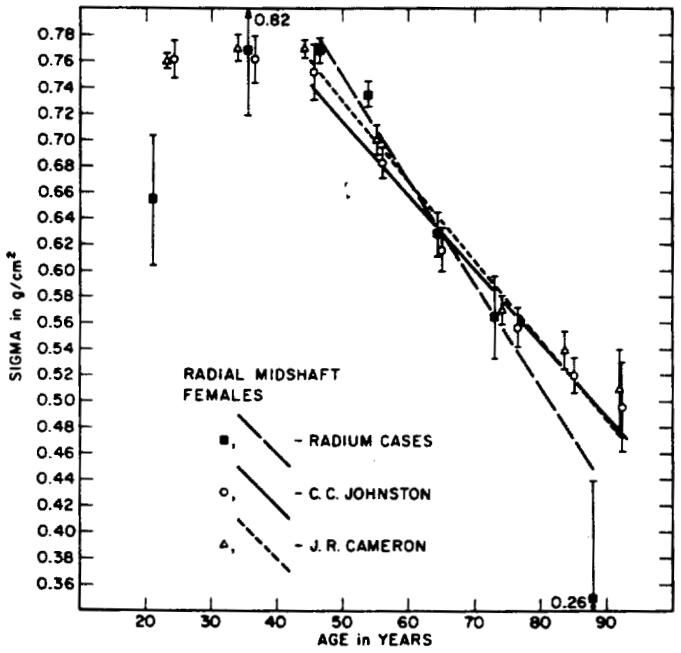

FIG. 2.--This graph presents sigma vs. age at the radial midshaft in female radium cases and normals. Mean values of sigma for ten-year age groups are plotted against mean values of age for the group members. Each error bar represents one standard error in the mean sigma value. Straight lines, least square fit to radium case data and to normal data are shown plotted over the age ranges to which they apply.

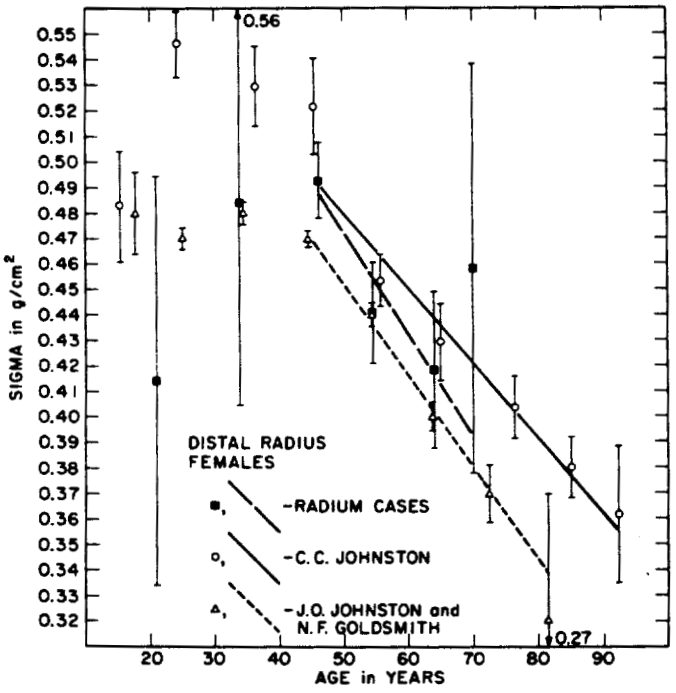

FIG. 3.--This graph presents sigma vs. age at the distal radius in female radium cases and normals. Mean values of sigma for ten-year age groups are plotted against mean values of age for the group members. Each error bar represents one standard error in the mean sigma value. Straight lines, least square fit to radium case data are shown plotted over the age ranges to which they apply.

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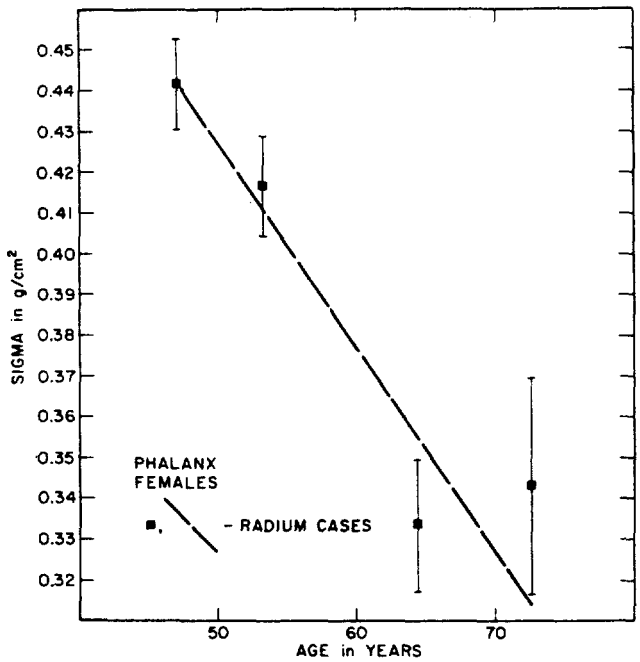

FIG. 4.--This graph presents sigma vs. age at the phalanx in female radium cases. Mean values of sigma for ten-year age groups are plotted against mean values of age for the group members. Each error bar represents one standard error in the mean sigma value. A straight line, least square fit to the data, is shown plotted over the age range to which it applies.

whether or not they were statistically different at the 5% level. For the radial midshaft and distal radius the line slopes for the two sets of normal data were not statistically different. For both the distal radius and radial midshaft data, one group of normals was scanned on the right arm and one was scanned on the left arm. Thus, right and left arms show symmetry with respect to rate of decrease of sigma. For the radial midshaft, the magnitude of the line slope for radium cases was significantly greater than the magnitude of the slope for either group of normals, indicating that radium cases suffer demineralization at a greater rate than do normals. No significant difference was noted between normals and radium cases at the distal radius location. However, the parameters of the fitted line for radium cases at the distal radius are much less precise than those at the radial midshaft, because scan data are much less abundant for the distal radius. Data on the phalanx and distal radius were compared to determine whether or not demineralization of the two types of cancellous bone takes place at different rates. No significant difference in line slopes was found between the phalanx and the distal radius for radium cases and for one group of normals; a significant difference was found in comparison with the second group of normals. These

results fail to give a conclusive indication of whether or not phalanx and distal radius demineralization rates are different.

The loss rate for sigma was significantly greater in radium cases than in normals at the radial midshaft. Radial midshaft data were analyzed to determine whether or not the loss rate was a function of body burden. X-ray score data on radium cases show that for body burdens below about 0.1 μCi ^{226}Ra pure radium equivalent 30 to 50 years after initial uptake, the average x-ray score is not statistically different from the average for unexposed controls.[12] With this information as guidance, the radium case radial midshaft data were separated into two body-burden classes: 1) < 0.1 μCi containing 134 members distributed among 4 decades, 40 to 49 through 70 to 79, and 2) ≥ 0.1 μCi containing 10 members distributed among 3 decades, 60 to 69 through 80 to 89. Straight lines were least square fit to the data in each class; the parameters of the straight lines obtained at the radial midshaft in radium cases are summarized below.

Body burden, μCi	A, g/cm^2 per year x 10^{-3}	B, g/cm^2
< 0.1	- 8.21 ± 0.84	1.16 ± 0.05
≥ 0.1	-18.3 ± 4.4	1.89 ± 0.32

The line slopes are significantly different from one another and from the line slopes for normals at the 5% level. Plots of the radial midshaft data and the least square fit lines for radium cases are presented in Figure 5. More significant digits were retained in the straight line coefficients for the purposes of plotting the straight lines than are quoted above. Thus, slight differences may exist between the plotted lines and lines based on the coefficients above.

Discussion

Analysis of the data reveals a significant difference between the rate at which sigma diminishes with age in radium cases and in normals in

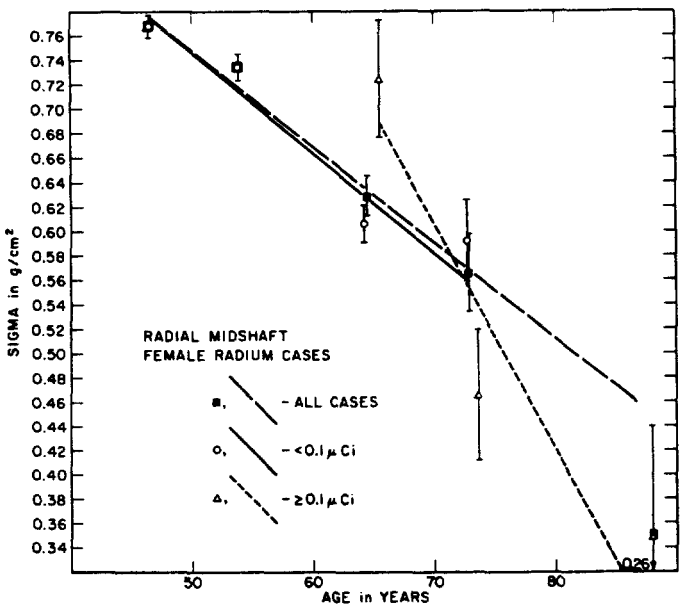

FIG. 5.--This graph presents sigma vs. age at the radial midshaft in female radium cases. Three data classifications are presented: 1) all cases, 2) cases with ^{226}Ra body burden < 0.1 μCi, and 3) cases with ^{226}Ra body burden ≥ 0.1 μCi. Mean values of sigma for ten-year age groups are plotted against mean values of age for the group members. Each error bar represents one standard error in the mean sigma value. Straight lines, least square fit to the points in each data classification are shown.

the radial midshaft. The most obvious explanation of this is that the difference results from exposure to radium, and support is lent to such a conclusion by the fact that the rate of decrease is greater in radium cases who have a body burden greater than 0.1 μCi than in radium cases whose body burdens are less than 0.1 μCi. However, factors other than radium burden, which tend to accelerate demineralization, may be present at higher levels in the radium case population than in the normal population. The normal population is composed of people who have no evidence or history of bone disease other than the natural tendency to bone loss which accompanies increasing age. Among the radium cases, four persons were identified who had suffered bone loss from causes other than radium. However, no systematic search of the radium case records was made to identify all persons who might be suffering from bone disease unrelated to radium. Until such a search is made, radium cannot be accepted conclusively as the cause of the difference in the rate of demineralization in the radial midshaft between radium cases and normals.

There are six radium cases with radial midshaft scans between 60 and 69 years of age who have body burdens ≥ 0.1 μCi. As may be seen from

examination of Figure 5, the average value of sigma in these six cases is significantly greater than the average value predicted by the line fitted to the radium cases with body burdens < 0.1 μCi. This may be due to hypermineralization resulting from radium damage to the bone. Radiographs of the right radius and ulna in each of these cases show areas of hypermineralization in only one of the cases. In the others, areas of resorption are noted or no changes are noted at all. If the one case which shows hypermineralization were removed from the 60 to 69 year group and the average for the remaining five cases were computed, it would be greater than for the original six. Thus, the hypothesis that the large value of sigma is due to hypermineralization cannot be verified by radiographic evidence.

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CYTOGENETIC STUDIES IN PATIENTS WITH A CHRONIC RADIUM BURDEN

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We describe an ongoing study at the Center for Human Radiobiology designed to detect chromosomal changes in a large population of radium patients. The long-term goal is to determine the nature of the dose-response relationship between alpha radiation and the frequency with which certain chromosomal aberrations are observed in cultures of circulating lymphocytes.

A new program at the Center for Human Radiobiology is attempting to correlate both the patients' current body burdens and their total estimated internal radiation doses with the level of chromosomal damage in their peripheral lymphocytes.

Papers reviewed in an earlier publication of the Center by Simmons and Cummins[1] have indicated that the amount of cytogenetic damage resulting from a variety of types of radiation can serve as a biological dosimeter of radiation dose. We desire to test this hypothesis for patients with long-term chronic doses.

Methods and Materials

For our cultures of peripheral lymphocytes, we culture blood by a modified Moorehead technique.[2] The details of this technique were presented earlier.[1] Briefly, it involves removing the plasma-leucocyte fraction of heparinized blood and culturing this fraction in a growth medium supplemented by phytohemaggutinin to stimulate cell division. After 48 hours, Colcemid is added to arrest mitosis, further contract the chromosomes and increase the number of mitotic figures available for later analysis. The cells are transferred to hypotonic saline to swell the cells, thereby improving chromosome spreading. The materials are then fixed, spread on slides, and stained. To date, material from 160 patients has been prepared.

The slides are then scanned under the microscope and the chromosomes are scored for the presence of aberrations. We are emphasizing the scoring of dicentric and centric ring chromosomes. Although these aberrations are somewhat unstable, reliance is usually placed on them in the

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scoring of human cytogenetic damage since they are relatively easy to observe compared to pericentric inversions, small deletions and translocations. As recently recommended,[3] we will also take note of all other aberrations observed. (The reader who is not familiar with the nature of chromosomal aberrations is referred to two excellent reviews[3,4] which discuss the scoring of chromosomal damage in irradiated persons.)

In a parallel study, we are examining the lymphocytes of a normal person after in vitro exposure of the plasma-leucocyte faction to known doses of short-term radiation. This study will serve as a standard of comparison for the material from the radium patients. We are also using samples from unirradiated persons as controls.

A sample of at least 100 cells per patient is scored. Larger samples will be obtained from patients whose first sample shows unexpectedly high or low aberration frequencies.

Results to Date

Scoring is carried out on coded slides to avoid subjective bias, and decoding will not be done until the remainder of the slides on hand has been scored. Therefore, we will not present full data at this time.

We can, however, report on one case with a high body burden. Slides from this patient have been used to standardize scoring techniques between scorers. Case 10-010 has a current body burden of 8.72 μCi ^{226}Ra. Her total dose cannot be calculated since the time of radium administration is not known, but it is assumed to be at least on the order of 10,000 rads.

One hundred and three cells from this patient have been scored, and four cells with centric rings or dicentrics were seen. Figure 1 shows a cell from this patient with dicentrics and other abnormalities. The cell is near tetraploid and has two identically appearing dicentrics, indicating that doubling of the chromosome number probably followed dicentric formation. At least three fragments and a chromatid minute are also present.

Twenty-one cells with other aberrations were observed. These cells had fragments, minutes, chromatid gaps, or were hyperdiploid. The total

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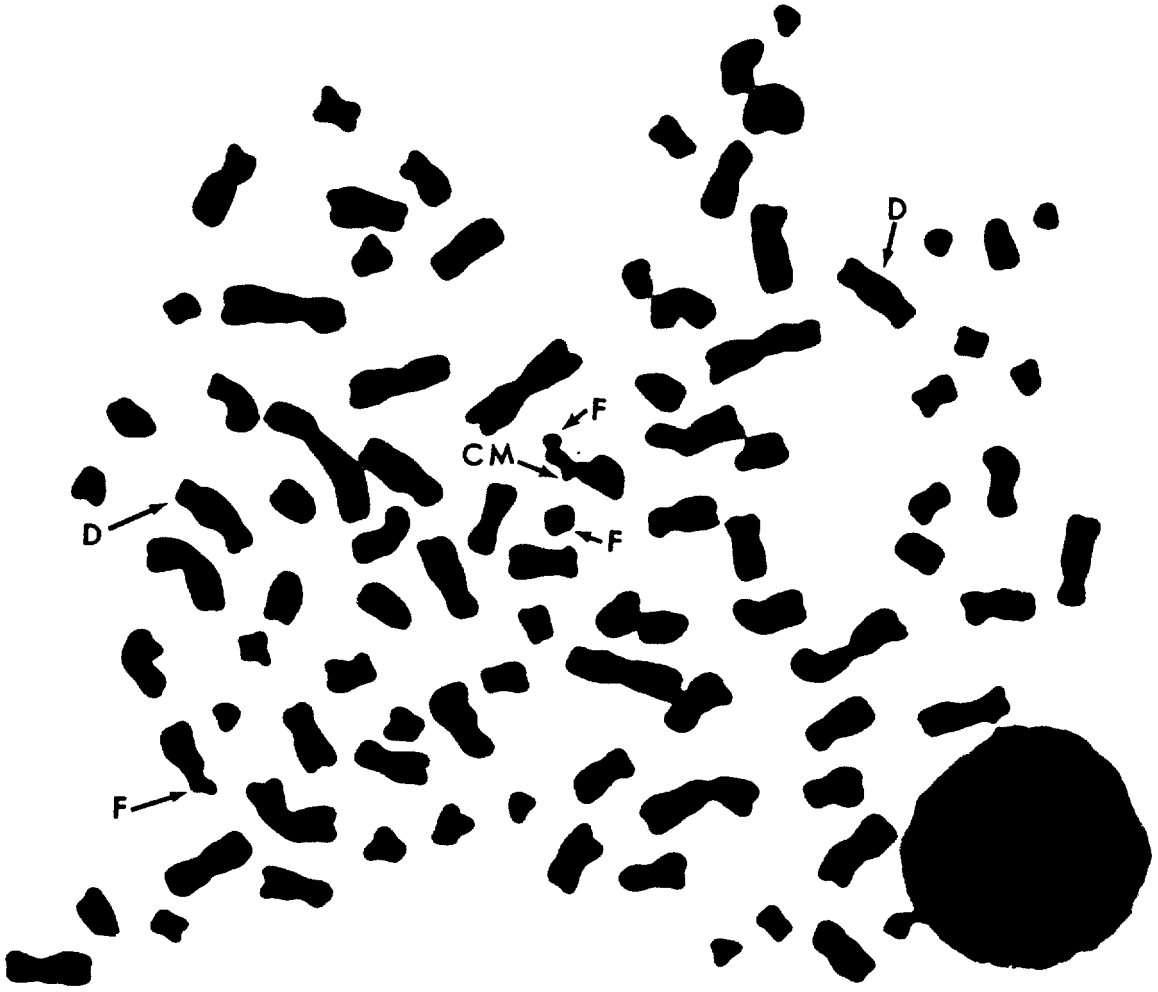

FIG. 1.--A near tetraploid cell with dicentric chromosomes and other abnormalities. D, dicentric; F, fragment; CM, chromatid minute. Case 10-010. X 1700.

abnormality rate for this case is, therefore, 24.3%, expressed as the percent of cells with abnormalities.

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RADON BREATH MEASUREMENTS: ESTIMATES OF CORRECTIONS FOR PREVIOUSLY INHALED RADON

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In measurements of the ^{226}Ra content of living humans by the radon breath method, a correction must be made for the background radon which is absorbed into the body from natural environmental sources and released when samples of exhaled air are taken for analysis. This correction would be equivalent to about 20 nCi of emanating radium in the body for typical concentrations of 0.3 pCi of $^{222}Rn/l$ of room air, so a preliminary period of breathing air of low radon content is employed when body burdens of less than 1 μCi ^{226}Ra are to be measured. The breath content of previously absorbed radon is reduced by a factor of about 10 to 20 during the 1- to 2-hr period which is available (including time during which gamma-ray measurements are made). Even so, corrections accurate to the equivalent of a few tenths of a nanocurie of body radium are difficult to achieve for each person because of the wide range of radon content possible for air previously breathed.[1-3] Also, the rate of release of absorbed radon appears to depend on the individual's physiology and anatomical factors such as the amount of body fat.[4-6] Since longer periods of deemanation or accurate determinations of radon blanks for each individual are not feasible, a group of controls (natural levels of approximately 0.04 nCi expected[7]) was studied in order to determine standard blank corrections. Parameters were adjusted to provide an apparent body burden of about 0.03 nCi as the average for the group and a realistic estimate of the error of individual results.

The first step was to obtain an equation for the elimination of previously absorbed radon as a function of time of breathing radon-free air. In the manner of Harley et al.,[4] observed ratios of the concentration of radon in exhaled air to the concentration in air previously breathed were expressed as the sum of five exponential terms. In our case, a single set of parameters was fitted to measurements on 2 volunteers after uptake of air with 100 pCi $^{222}Rn/l$ for 72 hr[6] and measurements on 5 persons after exposure to natural

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environmental levels of radon.

$$R(t) = \sum_{1}^{5} C_i \exp(-k_i t) \qquad (1)$$

Coefficients and half-time values for the 5 terms of Equation (1) are shown in Table 1. The indicated rate of elimination of radon is somewhat less than would be expected from extrapolation to saturation uptake[8] of the Harley data.

TABLE 1. Coefficients and Half-Time Values for Equation (1)

Term	Coefficient	Half-time, min
1	0.641	0.38
2	0.175	4.47
3	0.139	41.2
4	0.0399	120.
5	0.00485	1100.

Applying Equation (1) to measured concentrations of radon in air exhaled 1 to 3 hr after start of breathing radon-free air, an average value of 0.4 pCi ^{222}Rn/l in previously breathed air was estimated from preliminary tests on four subjects. Setting the standard deviation of the blank at 67% of the value of the blank provided a root-mean-square error of the mean equal to that calculated from the dispersion of the individual values of apparent radium content. It should be noted that these values refer to exposure to unknown levels of radon in air in various places during periods of many hours prior to the measurements of exhaled air. In general, measuring room air at the time and place of the tests would not provide the information needed about previous exposure to radon. The above results (Equation (1), 0.4 pCi/l, and standard deviation of 67%) were used for estimating blank corrections for all radium cases analyzed during the period of June 1, 1971 to July 31, 1972.

Measurements of the apparent body content of emanating ^{226}Ra for 9 control subjects tested during the past year are summarized in Table 2. Two different room air values were used. The first results were obtained using the 0.4 pCi/l room air value estimated from the first 4 control subjects. Evidently too large a correction was subtracted, because a positive result was obtained for only one of the nine subjects and the mean value of apparent body content was -0.36 nCi ^{226}Ra. When an average room air content of 0.27 pCi/l was used, nearly equal numbers of positive and negative results were obtained. The difference in the means for these two conditions is about 0.4 nCi, and suggests that values of emanating ^{226}Ra reported for subjects measured prior to 31 July 1972 were too low by this amount. A similar suggestion has been made by J. Rundo in another section of this report.

The standard deviation of the blank was determined from the data in Table 2 by an iterative procedure. A value of 75% of the blank was chosen because the internal and external standard errors of the mean were nearly equal when that value was tried. The equations of Jaffey[9] were used, as indicated below.

The weighted mean, $\bar{x}$, was calculated from the individual values

$$\bar{x} = \frac{\sum^{k} W_j X_j}{\sum^{k} W_j} , \qquad (2)$$

where $W_j = 1/\sigma_j^2$.

The standard error in the mean, $\sigma(\bar{x})$, (internal error) was calculated from the individual estimate of standard errors:

$$\sigma(\bar{x}) = \frac{1}{N} \left(\sum^{k} \sigma_j^2 \right)^{1/2} . \qquad (3)$$

The standard error in the mean, $S(\bar{x})$, (external error) was calculated from the dispersion in the results:

TABLE 2. Apparent Emanating ^{226}Ra in Control Subjects as Estimated for Two Different Sets of Conditions

Subject No.	Old conditions,[a] nCi ± σ	New conditions,[b] nCi ± σ
50-013	-0.730 ± 0.603	-0.387 ± 0.496
50-001	-0.209 ± 0.702	+0.326 ± 0.555
50-003	+0.616 ± 0.461	+0.935 ± 0.379
50-007	-0.560 ± 0.596	-0.129 ± 0.483
50-008	-0.197 ± 0.519	+0.166 ± 0.425
50-009	-0.377 ± 0.803	+0.189 ± 0.656
50-010	-0.769 ± 0.546	-0.384 ± 0.446
50-011	-0.887 ± 0.577	-0.478 ± 0.471
50-012	-0.762 ± 0.599	-0.335 ± 0.486
Weighted mean $\bar{x}$	-0.365	+0.032
$\sigma(\bar{x})$	0.202	0.165
$S(\bar{x})$	0.180	0.172

[a] The concentration of radon in previously breathed air was assumed to be 0.4 pCi/l, and the standard deviation of each blank was 67% of the blank.

[b] The concentration of radon in previously breathed air was assumed to be 0.27 pCi/l, and the standard deviation of each blank was 75% of the blank.

$$S(\bar{x}) = \left(\sum^{k} \frac{(x_j - \bar{x})^2}{N(N-1)} \right)^{1/2} . \qquad (4)$$

The new estimate of the average room air radon content of 0.27 pCi/l and the uncertainty of 75% of the blank will be used for all radium cases measured after August 1, 1972. Additional control subjects will be run as time permits.

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CALIBRATION OF COUNTING EQUIPMENT FOR MEASUREMENTS OF RADIUM IN VIVO

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The "arc" method for determining the amount of a radionuclide in the body by gamma-ray spectrometry was used to estimate the radium C burdens of 11 patients. Gamma-ray measurements of these persons in the tilted chair yielded a mean calibration factor for this geometry, which is used to determine low levels of radium C in the body. Various parameters affecting the counting efficiency for the chair geometry are discussed.

Introduction

The "meter-arc" method for determining, in vivo, the amount of a gamma-ray emitting radionuclide in the body was developed by Evans[1] many years ago, and the validity of the method has been demonstrated in several experiments.[2-4] It has been used by us during the past year to estimate the radium C (^{214}Bi) body content of 11 patients. These persons were then counted in the tilted (Argonne) chair to calibrate this more efficient geometry. The application of the arc method to the radium studies is discussed below.

The Basic Equation

The equation derived by Evans is (using his notation):

$$R = A(S_3 S_4 N_1 N_2 / N_3 N_4)^{1/2} \quad , \qquad (1)$$

where

$$A \equiv \left(1 - \frac{d}{u} + \frac{x(d-x)}{u^2}\right)^{n/2} \doteq 1 - \frac{nd}{2u}$$

and R is the amount of the radionuclide in the body at an unknown effective distance x from one side of a body of thickness d. The subject lies on an arc having radius of curvature u, with the detector at the center of curvature of the arc. R has the units of S_3 and S_4, which are the strengths of two point sources, one of which is placed behind (under) the body at distance u and gives counting rate N_3, and one of which is counted bare at distance u

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and gives counting rate N_4. In practice, the standard source is counted at several positions under the body, and N_3 is the mean counting rate for the several measurements. Note that when $S_3 = S_4$, $N_3 = N_4 \cdot e^{-\mu d'}$, where μ is the appropriate linear attenuation coefficient for the body and d' is the effective body thickness. N_1 and N_2 are counting rates for the subject facing the detector, and with his back to the detector, respectively. The n of Eq. (1) is determined for a particular source, detector, and counting room, where the relationship between the γ-ray intensity I_r at r cm from a source, and the intensity I_0 at 1 cm is given by $I_r = I_0 r^{-n}$. In this way allowance can be made for possible deviations from strict adherence to the inverse square law ($n = 2$), which may result from the detection of scattered radiation.

The detector employed in this work is the "log" crystal, a cylindrical crystal of NaI(Tl) 6" in diameter and 8" in length. The arc bed, made of 1/4" Teflon on a tubular steel frame and covered with a 1" thick polyurethane foam mattress, has a radius of curvature u of 150 cm. The counting room has been described previously.[5] The method of determination of each of the parameters of Eq. (1) is described below. All counting rates are for the energy band from 1605 to 1875 keV.

Determination of n

The large value of u and the selection of the high energy γ-ray peak are favorable conditions for obtaining a value of $n = 2$. The counting rates N_i from a radium source were determined at several distances r_i from the cylindrical surface of the detector to determine the effective center of the detector in terms of its depth from the detector surface. A plot of $N_i^{-0.5}$ as a function of r_i was a straight line which gave an "r intercept" at a value of about -7.5 cm, which is very close to the geometric center of the detector.

Thus, n has a value of 2 for our measurements, with the crystal positioned so that its surface is 142.5 cm above the arc bed mattress.

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Determination of Mean Transmission Factor and Thickness d

Measurements with relatively few subjects have shown that the mean of the counting rates given by the radium standard under the chest, lower back, and one knee, with the knee position value weighted twice, is within a few percent of the mean of the counting rates for the standard under many different locations along the subject. Therefore, N_3 is the mean for the three positions of the standard under the subject (knee value weighted twice), and the mean transmission factor is $(N_3/N_4)^{1/2} = e^{-\mu d'/2}$, when $S_3 = S_4$.

It should be noted that, although we use the single-depth attenuation correction term $e^{\mu d'/2}$, the correction term for a uniformly distributed source is given by the inverse of the transmission factor averaged over the distance d', and is $\mu d'/(1-e^{-\mu d'})$. This term averages about 2% lower in value for subjects than the single-depth term, for $\mu = 0.047$ cm^{-1}. Of course, the distribution of radium in the human skeleton is neither of these extremes, but it may average somewhere in between.

Elsewhere in this report,[6] Rundo et al. present data which show that an "average body thickness" can be predicted from the weight and height of a subject. This quantity is $d'\rho$, where ρ is the density of the body in g cm^{-3}.

A mass attenuation curve for the 1.76-MeV γ-ray energy band of radium C was determined in a similar manner to that described by Rundo et al. for 1.275-MeV γ rays from sodium-22. This curve gave a value for the mass attenuation coefficient μ/ρ of 0.047 cm^2/g. Using their expression relating $\sqrt{\text{weight/height}}$ ($\sqrt{W/H}$) and thickness, the product $\frac{\mu}{\rho} \cdot d' = -\ln (N_3/N_4)$ can be determined without recourse to the time-consuming measurements of standards under the subject. An error of ± 1 cm in the estimate of $d'\rho$ will result in a $\pm 2.3\%$ error in $e^{\mu d'/2}$.

The parameter d which appears in the inverse square correction term A of Eq. (1) has a value close to, but probably not the same as, the value of $d'\rho$, even if ρ is assumed to be 1 g cm^{-3}. If the body is assumed to be represented by an elliptical cylinder of uniform density with, say,

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eccentricity = 2, d is the length of the minor axis, while the length of d' is probably between d and the length of a shorter line passing through the centroid of the semi-ellipse, parallel to the minor axis. In this model, d' is about 10% shorter than d for a uniformly distributed source. In practice, however, d is taken to be the value of d'ρ. An error of ± 1 cm in the estimate of d will result in a ∓ 0.7% error in A. An error of ± 1 cm in the estimate of d'ρ, therefore, will result in an error of ± 1.6% in the product $A \cdot e^{\mu d'/2}$, and also in R.

Determination of S_3 and S_4

A radium solution sealed in a glass ampoule encased in a Lucite cylinder of 3-cm outside diameter and 2.2-cm inside diameter, dated 1952 and labeled 8.72 μCi, was used as the reference source in all the arc measurements. The transmission factor for the 1.76-MeV radium C γ ray was provisionally estimated to be 0.95. Recently, this standard was compared with both a 1.00-μg and a 0.99-μg NBS standard (1967 series), and the 1972 radium content is estimated to be 8.5 ± 0.1 μg. This value is within about 1% of the labeled value when the 20-year decay correction is made. Careful measurements place the transmission factor at a value of 0.94 ± 0.01, so that the relative calibration error in the "point-source" value of the standard is about ± 1.7%. In our use of Eq. (1), then, $S_3 = S_4 = 8.5\ \mu g \times 0.94 = 8.0\ \mu g$ (or μCi).

Determination of N_1 and N_2

N_1 and N_2 are determined from two measurements of the activity, usually of about 20 minutes each. When the subject is facing the detector the body does not conform to the arc in the region of the knees, so a small inverse-square correction averaging -2% is applied to N_1 to account for this.

Modified Form of Eq. (1)

Most of the arc results reported in the next section were determined using Eq. (1), but our present modified form of the equation is:

$$R = A \cdot S_4 \, (N_1 \cdot N_2 \cdot e^{\mu d'})^{1/2} / N_4 \, , \qquad (2)$$

where

$$\mu d' = \frac{\mu}{\rho} \cdot d'\rho = 0.047 \cdot 22.5 \sqrt{W/H} = 1.06 \sqrt{W/H}$$

and

$$A = 1 - \frac{d}{u} \quad (d = d', \; u = 150 \text{ cm}) \, .$$

Estimated Standard Error in R

The standard error $[\sigma_{\bar{x}} = \sigma/\sqrt{n}]$ of R can be determined from the estimated uncertainties in each of the parameters of Eqs. (1) or (2). From the data in Ref. 6, a reasonable estimate of ± 1 cm for a standard deviation of the value of d' leads to a ± 1.6% relative standard error $(100 \, \sigma_{\bar{x}} / \bar{x})$ of the product $A \cdot e^{\mu d'/2}$. The relative standard error of N_4 for a 10-minute count of the reference source is ± 0.7%. Normally, two 20-minute counting periods determine $(N_1 N_2)^{1/2}$, and for radium C burdens ≥ 50 nCi, relative standard errors of counting after background subtraction will be ≤ 8%. Thus, in practice, the overall relative standard error ($n = 1$) in R will be about ± 2 to ± 8%, depending on the value of R.

The total uncertainty in R includes, in addition to the overall standard error, the ± 1.7% error in the strength S_4 of the radium standard. A systematic error of ± 5% in the use of $\sqrt{W/H}$ or N_3/N_4 in the estimation of $\mu d'$ is arbitrarily assigned, leading to an additional average error of about ± 1% in R. In addition, there is the possibility that use of the single-depth attenuation correction term of Eqs. (1) or (2) will overestimate the true burden by as much as 2%, and the use of d' for d no more than an additional 1%.

Results and Discussion

Measurements in Arc Geometry

Eleven people with old radium burdens have been measured in the arc in the past year. Their physical characteristics and the results of the measurements are given in Table 1. Seven of the burdens were calculated using Eq. (1); for cases 03-416 and 03-206, Eq. (2) was used. Average body thicknesses for cases 03-505 and 03-727 were estimated from the thickness vs. $\sqrt{W/H}$ data of the seven radium cases whose thicknesses had been calculated from determination of N_3 and N_4, using the radium standard. The listed error for each RaC content is the overall relative standard error discussed in the previous section.

TABLE 1. Physical Characteristics and Results of Measurements in the Arc Geometry for Eleven Radium Cases

Case No.	Sex	Height, cm	Weight, kg	RaC content, nCi
03-505	F	155	50.8	48 ± 8.0%
01-144	F	153	50.5	232 ± 4.4%
03-402	F	155	53.2	396 ± 2.9%
01-017	F	155	54.5	421 ± 3.0%
03-404	F	150	53.1	262 ± 4.5%
03-416	F	155	62.7	385 ± 2.9%
01-431	F	163	68.4	297 ± 3.0%
03-540	F	161	73.5	623 ± 2.8%
03-727	F	163	74.5	63 ± 7.9%
01-208	M	170	82.6	282 ± 4.2%
03-206	M	178	91.3	1090 ± 3.0%

Measurements in the Tilted Chair

Each of the eleven subjects was counted in the tilted chair with the

6" dia. x 8" long "log" crystal positioned above it; the crystal surface was 66 cm above the right-angle "vee" of the chair, and approximately equidistant from seat and backrest. Counting times varied from 10 to 20 minutes, with a few of the higher level cases counted for several 5-minute periods to check reproducibility and to determine the effects of arm and leg repositioning. No significant difference in counting efficiency was observed between measurements made with the arms on the armrests and with the arms on the lap. A study of the effect of movement of the legs on the counting efficiency was limited to two cases (01-017 and 03-206). The efficiency for a position with the knees apart was 0.965 ± 0.013 of that for the normal position (knees together).

Distribution Studies

The relative distribution of radium C along the length of five of the subjects was determined by scanning-type measurements, using the seven-crystal position technique developed by C. E. Miller.[7] The subject lies supine on a steel mesh bed which is covered by a 1" thick mattress. An 11-1/2" dia. x 4" thick NaI crystal is positioned with its surface 30 cm above the steel mesh, and a similar crystal is positioned with its surface 10 cm below the steel mesh. Measurements are made with the crystal above and below the subject at his midlength (position 4), and at each of three positions 24.5 cm apart toward the subject's head (positions 3 to 1) and each of three positions 24.5 cm apart toward his feet (positions 5 to 7). The results of these measurements are summarized in Figure 1.

The ratio of the counting rates observed from the front and the back of the subject in the arc measurements also gives information on relative distribution, since an effective depth from the body surface for the radium deposits [x in Eq. (1)] can be calculated from this ratio for each of the subjects. These calculations were carried out using an expression derived by Rundo[8] and are discussed below.

Counting Efficiencies for Chair and Arc Geometries

The counting rates observed for each subject in the arc and in the chair were divided by the radium C burden determined from the arc measure-

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ments to yield calibration factors for these geometries. Effective fractional depths of the radium deposits from the front surface of the body were calculated as the ratio x/d. For this purpose, d was calculated as 22.5 $\sqrt{W/H}$ for each case. These data appear in Table 2; the eleven cases are listed in order of increasing value of $\sqrt{W/H}$. The quoted standard errors in the efficiency values for the arc geometry are calculated from the overall relative standard errors in the values of R. A point-source radium standard is counted with an efficiency of 0.244 cpm/nCi at 150 cm.

The standard errors in the efficiency values for the chair geometry are calculated from the relative standard errors in the values of R combined

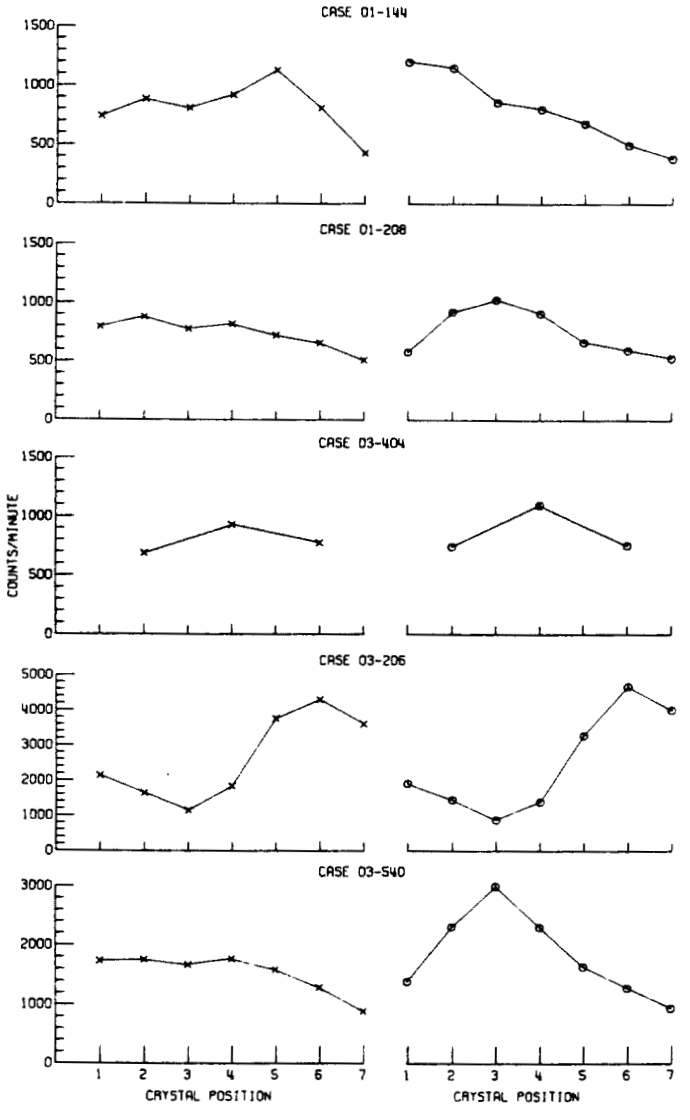

FIG. 1.--Seven-position scan profiles for five radium cases, measured in the supine position with two 11-1/2" x 4" crystals. Crystal position 1 is near the head and crystal position 7 is near the feet. Case 01-144 was counted with a small pillow under one knee. All five subjects were counted with a small pillow under the head, except for the counts taken at positions 1 and 2. X, crystal above subject; ⊙, crystal below subject.

TABLE 2. Counting Efficiencies for Arc and Chair Geometries

Case No.	$\sqrt{W/H}$, $kg/cm^{1/2}$	Facing/back arc ratio, N_1/N_2	Fractional depth, x/d	$(N_1N_2)^{1/2}/R$ 150-cm arc, cpm/nCi	Tilted chair, cpm/nCi
03-505	0.572	1.10 ± 0.17	0.44 ± 0.09	0.205 ± 0.016	1.49 ± 0.13
01-144	0.575	1.13 ± 0.09	0.42 ± 0.05	0.196 ± 0.009	1.68 ± 0.08
03-402	0.586	0.97 ± 0.04	0.52 ± 0.02	0.195 ± 0.006	1.44 ± 0.05
01-017	0.593	0.85 ± 0.04	0.60 ± 0.02	0.198 ± 0.006	1.43 ± 0.05
03-404	0.596	0.95 ± 0.08	0.53 ± 0.04	0.188 ± 0.008	1.44 ± 0.07
03-416	0.636	1.05 ± 0.06	0.47 ± 0.03	0.191 ± 0.006	1.45 ± 0.04
01-431	0.648	0.88 ± 0.04[b]	0.57 ± 0.02	0.190 ± 0.006	1.21 ± 0.04
03-727	0.676	0.91 ± 0.13	0.55 ± 0.07	0.182 ± 0.014	1.42 ± 0.12
03-540	0.676	0.87 ± 0.03	0.57 ± 0.01	0.179 ± 0.005	1.27 ± 0.04[a]
01-208	0.697	0.95 ± 0.07	0.53 ± 0.03	0.179 ± 0.008	1.29 ± 0.06
03-206	0.716	1.18 ± 0.05	0.40 ± 0.03	0.188 ± 0.006	1.35 ± 0.05
$\bar{x} \pm \sigma_{\bar{x}}$	0.633	0.94 ± 0.03	0.54 ± 0.02	0.189 ± 0.002	1.37 ± 0.036

[a] Subject had right leg extended in chair measurement.
[b] Subject had small pillow under abdomen for arc measurement with back to crystal.

with the relative standard errors of counting in the chair geometry.

Radium sources varying in strength from 0.1 μg to 8.5 μg were measured at several different geometries, and the counting efficiency in the radium C band was found to be invariant with source strength, with negligible counting errors.

Efficiency for Arc Geometry vs. $\sqrt{W/H}$

A plot of the efficiency for the arc geometry as a function of $\sqrt{W/H}$ is shown in Figure 2, together with a calculated curve derived from Eq. (2). Error bars are not shown since the only replication error which applies here is the ±0.7% counting error for the radium standard measured at 150 cm. If the burdens of all the cases had been calculated from Eq. (2) (i.e., $\mu d' = 1.06 \sqrt{W/H}$), then all eleven points would adhere closely to the line reflecting only the ±0.7% counting error in N_4. Figure 2 illustrates that for

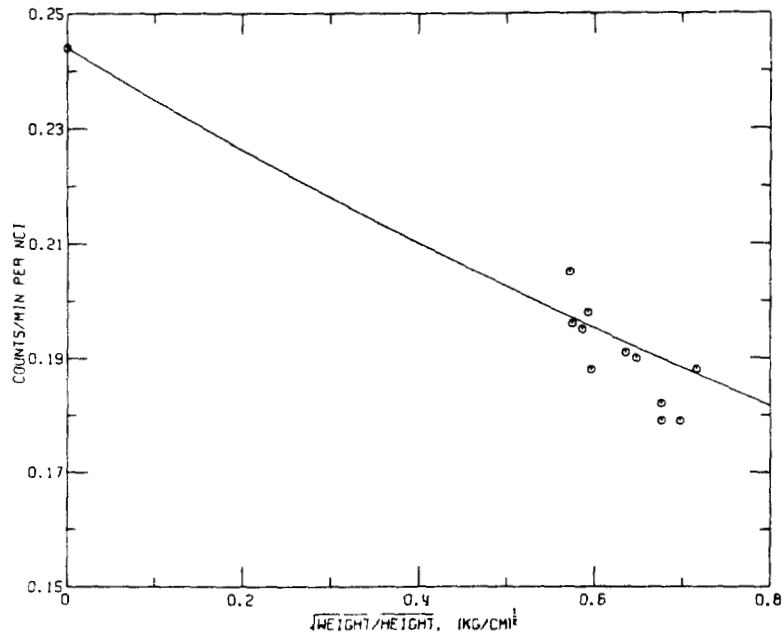

FIG. 2.--Counting efficiency for the arc geometry as a function of $\sqrt{\text{weight/height}}$. The curve is calculated from the equation $(N_1 N_2)^{1/2} \cdot R^{-1} = 0.244\, e^{-\mu d'/2} (1 - \frac{d'}{u})^{-1}$ cpm/nCi, where 0.244 is the reference standard efficiency at 150 cm, $\mu = 0.047\ \text{cm}^{-1}$, $d' = 22.5 \sqrt{W/H}$ (from Ref. 6), and $u = 150$ cm.

the three cases with the largest $\sqrt{W/H}$ values among those whose $\mu d'$ values were calculated from N_3/N_4, larger radium C values have been calculated than if $\mu d'$ were determined by the method of Ref. 6, i.e., if R were determined with Eq. (2). These differences in predicted thickness are discussed in Ref. 6, and demonstrate the desirability of repeating the experiment in that reference using a nearly "massless" radium source.

Efficiency for Chair Geometry vs. $\sqrt{W/H}$

In Figure 3, the counting efficiency for the chair geometry is plotted as a function of $\sqrt{W\ H}$, together with a calculated curve derived in a manner similar to that for the curve in Figure 2. The mean ratio of the counting rate in the chair to the counting rate in the arc (subject facing the detector) for the eleven subjects indicates that the effective radius of curvature for the

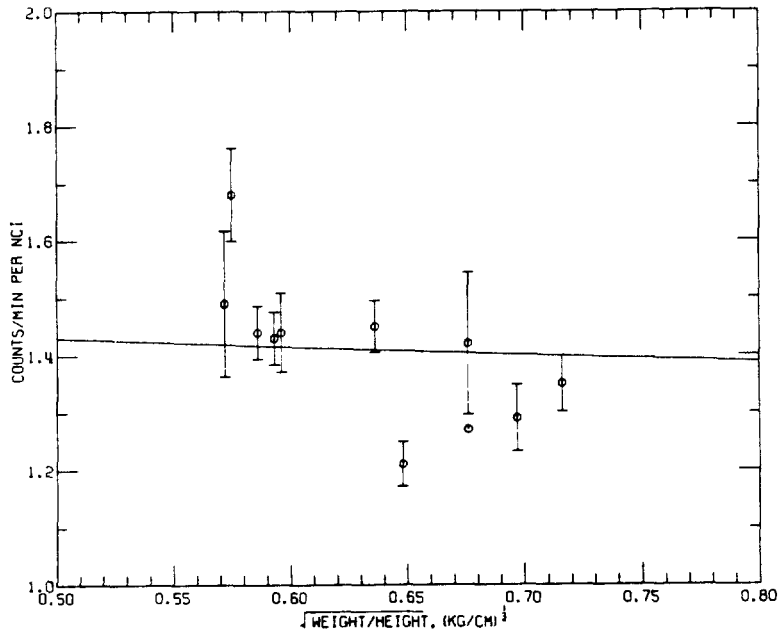

FIG. 3.--Counting efficiency for the chair geometry as a function of $\sqrt{\text{weight/height}}$. The curve is calculated from the equation $N \cdot R^{-1} = 0.244\ (150/60)^2 \cdot e^{-\mu d'/2}\ (1 - \frac{0.5d'}{60})^{-2}$ cpm/nCi, where $\mu = 0.047\ \text{cm}^{-1}$ and $d' = 22.5\ \sqrt{W/H}$ (from Ref. 6).

chair geometry is about 60 cm. Thus, the curve in Figure 3 is calculated with u = 60 cm and $A = (1 - \frac{0.5d}{60})^2$. The value of N_4 is determined by an inverse-square calculation from the observed counting rate for the radium standard at 150 cm or at 72 cm (i.e., in the "vee" of the chair). The curve passes above the weighted mean efficiency value of 1.37 cpm/nCi at the mean $\sqrt{W/H}$ value of 0.633 because an effective fractional depth of 0.5 is used in the equation for the curve.

From Tables 1 and 2 it can be seen that the values of $\sqrt{W/H}$ and height for these eleven cases are fairly well correlated, so that we are prevented from observing the separate effects of these two parameters. The use of $\sqrt{W/H}$ in Eq. (2) implies a single parameter, that of "average body thickness," and the effect of height as a separate parameter is implicitly

considered to be constant for all subjects. This is not a problem for the measurements in the arc, but for the chair geometry, and, indeed, for the scan measurements described previously, decreased efficiency with increasing height can be anticipated, other things being equal. In addition, the same comment that was made for Figure 2 regarding the determination of $\mu d'$ applies here.

Efficiency for Chair Geometry vs. Effective Depth

Differences in the effective depth at which the radium C is buried would be expected to be reflected in the counting efficiency data for the chair geometry. Figure 4 presents a plot of the efficiency in the chair as a function of effective fractional depth (x/d) for the eleven cases. Included in the figure is a curve calculated from Eq. (2), with $u = 60$ cm and $A = [(46 + 14x)/60]^2$. The calculated curve assumes a thickness of 14 cm, close to the average observed for the eleven cases. Since the curve of Figure 3 predicts < 2% variation in chair efficiency over the observed range of $\sqrt{W/H}$, differences in absolute depth (x) for the same value of fractional depth (x/d) should produce only minimal deviations from the calculated curve (< 4%).

Chair Efficiency vs. Skeletal Distribution

We may note that the effective depth values for many of the cases have very large fractional errors, so there is perhaps a reasonable fit to the calculated curve for most of the points in Figure 4. For those points farthest away from the line, fortunately there are scan data available which may help to explain the deviations. It is interesting to note that the points of the four cases which are farthest from the curve of Figure 3 (01-144, 01-431, 03-540, and 01-208) are much closer to the curve of Figure 4. Conversely, two cases (03-206 and 01-017) have points with ordinate values considerably farther from the curve of Figure 4 than from the curve of Figure 3. Scan data for these two cases show that their radium C distributions are quite unlike the distributions for other cases similarly scanned.

The scan profile for case 03-206 appears in Figure 1. Case 01-017 was not scanned at CHR, but a seven-position supine and prone scan with

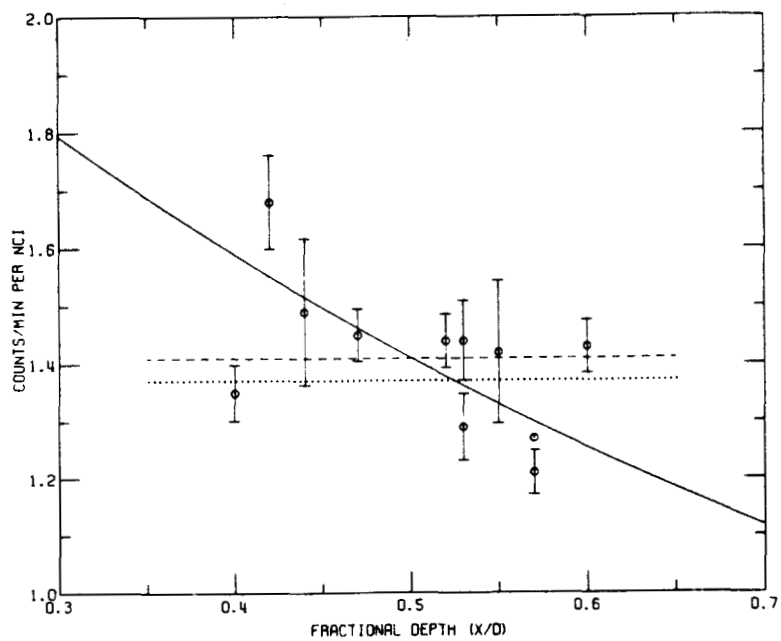

FIG. 4.--Counting efficiency for the chair geometry as a function of fractional depth (x/d). The solid curve is calculated from the equation $N \cdot R^{-1} = 0.244\ (150/60)^2 \cdot e^{-\mu \cdot x}\ [(46+x)/60]^{-2}$, where $\mu = 0.047\ cm^{-1}$, and x is the effective depth of burial of the radium C from the front surface of a body of thickness d = 14 cm. The dashed line is plotted at the arithmetic mean counting efficiency of 1.41 cpm/nCi, and the dotted line is plotted at the weighted mean efficiency of 1.37 cpm/nCi.

a similar 11-1/2" x 4" crystal was made at M.I.T. on this case.[9] Denoting by C_i the counting rate in position i for the crystal above the body, we note that the value of the ratio

$$\frac{C_6 + C_7}{\sum_{i=1}^{i=7} C_i}$$

lies between 0.20 and 0.23 for two other cases reported in Ref. 9 and for three of the cases in Figure 1. The value of this ratio is 0.43 for case 03-206 and 0.12 for case 01-017. Thus the variable deposits in the lower legs, which are counted relatively inefficiently in the chair geometry, might explain at least in part the deviations from the curve of Figure 4 for these two cases. The highest point in Figure 4, that for case 01-144, has an

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x-coordinate that is scarcely more than 1σ from the curve, and the scan data for this case, discussed later, are consistent with the observation of a higher than average counting rate in the chair geometry.

Unfortunately, scan data were not obtained for six of the eleven cases, so that the comparison of efficiency for the chair geometry with skeletal distribution of radium C is limited to five cases.

Mean Counting Efficiency and Standard Error Estimates for Chair Geometry

In Table 2, column 6, the weighted mean efficiency for the chair geometry is given as 1.37 cpm/nCi, with a standard error of ± 0.036. The predicted standard error, calculated as $\sqrt{1/\Sigma\sigma^{-2}}$ is ± 0.016. The difference between the observed and predicted variances leads to a calculated standard error of ± 0.032 from sources other than those for which errors have previously been estimated. The unweighted mean is 1.41 cpm/nCi, with an observed standard error similar to that for the weighted mean. The excess relative standard deviation ($100 \quad \sigma/\bar{x}$) is, therefore, estimated to be 7.6%, owing to a combination of additional sources of error. These other sources are assumed to be the differences in mean depths, in $\sqrt{W/H}$ values, and especially in skeletal distribution of radium C.

The excess relative standard deviation of ± 7.6% is combined with the relative standard error (± 1.2%) of the mean counting efficiency predicted from the standard errors of the arc and chair measurement data, to give an overall relative standard error of ± 7.7% for the mean efficiency value when it is used as the best estimate of the true counting efficiency for an individual. Thus, the standard error of a radium C burden of 50 nCi is the same (± 4 nCi), whether it is determined from two 20-minute observations in the chair geometry with application of the average calibration factor, or from two 20-minute observations in the arc geometry.

The precision of a radium C burden based on measurements in the chair geometry is determined by the standard error of counting, or, if larger, the observed replication error (two 20-minute measurements are usually made). A repositioning error has not yet been precisely determined, but it

is probably ± < 2%.

Therefore, the ± 4-nCi standard error in the 50-nCi burden discussed above might be thought of in a limited sense as an error of accuracy for the result of the measurements in the chair geometry (the standard error of replication would be only about ± 1 nCi), whereas in the case of measurements in the arc geometry it is a standard error of replication. However, the use of the standard error of replication to determine the significance of observed changes in the burden of an individual measured in the chair geometry at long intervals would require the assumption that changes in the distribution of radium C in the skeleton of the individual had not affected the calibration.

The relative systematic errors discussed for the arc measurements (± 1.7% in the strength of the reference standard, and ± 1% in R owing to methods of estimating γ-ray transmission), and the possible overestimation of burden by as much as 3% because of the use of the single-depth attenuation correction and the use of d' for d also apply to radium C burdens determined from measurements in the chair geometry.

Counting Efficiency for Seven-Position Scan Geometry

The average counting rate for scan positions 2, 4, and 6 combined was 103% (range = 98% to 108%) of the average for all seven positions, both for the crystal above and below the subject, for four subjects whose data are plotted in Figure 1. The fifth subject, case 03-404, was only scanned over positions 2, 4, and 6, and we will assume the average data for these positions adequately represent those of all seven positions for this case.

Table 3 summarizes the scan data. Note that there is a high degree of correlation for these few cases between $\sqrt{W/H}$ and the scan counting efficiency averaged for the two detectors. The ratio of the counting rate from the front of the subject to that from the back in the scan is also well correlated with the same ratio for the arc. In the scan, however, a subject of average build is closer to the detector below the scan bed than to the one above, so that the scan and arc ratios are not directly comparable. The ratios of the counting rates for the detector above the subjects in the scan

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TABLE 3. Counting Efficiency for Seven-Position Scan Geometry

Case No.	$\sqrt{W/H}$	Facing/back scan ratio, cpm/cpm	Mean scan, cpm/nCi	Facing scan/chair, cpm/cpm
01-144	0.575	1.03	3.48	2.10
03-404	0.596	0.92	3.17	2.11
03-540	0.676	0.82	2.87	2.04
01-208	0.697	0.99	2.63	2.02
03-206	0.716	1.05	2.36	1.79
$\bar{x}$	0.652	0.96	2.90	2.01

to the counting rates observed for the subjects in the chair are fairly constant except for the low ratio for case 03-206. Perhaps this low value is related to the height of this subject. The average ratio of 2.01 implies that the scan efficiency would be similar in value to the chair efficiency, were the same size detector used for both geometries.

Counting Efficiency for Thorium C Burdens

An estimated counting efficiency in the chair geometry for the 2.61-MeV γ ray of thorium C" was determined from measurements of a ^{228}Th NBS standard, some ThO_2 capsules, and the 8.5-μCi ^{226}Ra standard counted separately in a hardboard phantom placed in the tilted chair. The ratio of the counting efficiency in the thorium C band (2515-2725 keV) for the ^{228}Th source to the counting efficiency in the radium C band (1605-1875 keV) for the ^{226}Ra source was estimated to be ~ 1.1. The thorium C counting efficiency for subjects in the chair geometry is then 1.37 cpm/nCi x 1.1 = 1.5 cpm/nCi, and the relative systematic error in the value is estimated to be $\pm 5\%$. The minimum relative standard error for a thorium C determination from measurements in the chair is assumed to be the same ($\pm 8\%$) as for radium C determinations.

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The spectra of the ^{228}Th standard and the ThO_2 capsules were compared in order to determine the counting efficiency in the radium C band for mesothorium γ rays as well as for ^{228}Th daughter γ rays. It has been estimated that the ratio of ^{228}Th activity to mesothorium activity for old skeletal mesothorium burdens is 1.6. For this situation, the counting efficiency in the radium C band for mesothorium and ^{228}Th γ rays was determined from the phantom measurements to be equal to 37% of the counting efficiency in the thorium C band.

Counting Efficiency for Radium C in the Thorium C Band

The counting efficiency in the thorium C band for radium C γ rays was determined from measurements of radium sources bare and in phantoms, and is about 1% of the counting efficiency in the radium C band. However, it was found to be a slowly varying function of counting rate and of counting geometry. The observed variations in relative efficiency in the two bands are undoubtedly due to a combination of true and random coincidences, and, at the higher counting rates, pulse pileup.

For cases measured during the past year at CHR, the counting efficiency for radium C in the thorium C band, relative to the counting efficiency in the radium C band, was determined from the measurement of the reference standard in the "vee" of the chair which precedes each whole-body measurement. A ± 20% relative error is assigned to the observed relative counting efficiency for radium C in the thorium C band; this is about the magnitude of the change in the counting efficiency that would occur if there were an undetected shift of ± 1 channel in the analyzer zero intercept. This arbitrary uncertainty of ± 20% can be reduced when more precise information is obtained on the relationship between the relative efficiency in the thorium C band for radium in subjects and for the high-count-rate radium standard in the "vee" of the chair. Perhaps the selection of a narrower thorium C band would also improve the situation.

The counting rate from thorium C in the thorium C band is considerably less than 5% of the counting rate from radium C in the radium C band for our subjects, so that in practice, the radium C contribution to the thorium C band

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is taken to be about 0.01 ± 0.002 of the observed counting rate in the radium C band.

Calculation of Mesothorium Burdens

The mesothorium burden is taken to be the thorium C burden divided by 1.6. So far, only two cases measured at CHR were found to have significant mesothorium burdens. Both had ingested the nostrum Radithor more than 40 years ago. The ratio of the measured mesothorium burden to the radium burden for each of nine women who worked at the Radium Dial Co. plant in Ottawa, Illinois in the early 1920's was not significantly different from zero. However, the average ratio was significantly different from zero and was estimated to be 0.001 at the time of measurement. The radium C burdens of the nine ranged from 30 to 620 nCi.

Control Subjects and Water Phantom Measurements in the Chair Geometry

Several control subjects have been counted in the chair geometry, and data have been taken for a phantom composed of several polyethylene bottles filled with distilled water and placed in the chair to simulate a person. These data have not been fully evaluated yet, and more control subject measurements have to be made, but a preliminary analysis indicates that the average counting rate from a control subject is equivalent to a burden of not more than 0.3 nCi of radium C, and close to 0 nCi for thorium C. Control values of 0 cpm for both radium C and thorium C bands have been used for all the body burden determinations to date.

The room background for the radium C band averages 13.5 cpm, and for the thorium C band, 5 cpm. The minimum detectable radium C burden from a 40-minute measurement of the subject in the chair and a 100-minute measurement of room background is estimated to be 1 nCi (2σ), and the minimum detectable mesothorium burden is estimated to range from 0.3 nCi when no radium C is present to about 1 nCi in the presence of 1 μCi of ^{226}Ra.

Interlaboratory Comparison of Body Burden Determinations for Six Radium Cases

Six persons whose radium burdens were determined at the Radioactivity Center, M.I.T., subsequently were counted in the whole-body counter and

had breath radon determinations at CHR. The results of the measurements made at the two laboratories are presented in Table 4. The errors quoted reflect counting errors and scatter in replicate measurements, and do not include errors of calibration in reference sources or the effects of biological variation, which are probably similar at both laboratories.

The radius of curvature of the arc at M.I.T. is 125 cm, while the chair geometry is similar at both laboratories. The measurements at M.I.T. were made with an 11-1/2" x 4" NaI crystal. No interlaboratory comparison of radium standards has been carried out.

Apart from the data for case 01-431, the radium C burden ratios are within 2σ of a value of unity, and the average ratios for all three of the burden parameters are closer to unity than the standard errors would seem to warrant. The observed standard errors in the weighted means of the three parameters are 2 to 3 times the predicted errors.

The breath radon comparisons are also good, except for case 01-403, where the M.I.T. result for radon is somewhat high, and the CHR result somewhat low, relative to the respective radium C results. The mesothorium burden comparisons seem also satisfactory. Cases 01-017 and 01-403 are the two Radithor drinkers discussed in a previous section.

For purposes of comparison, the CHR radium C burdens determined in the chair geometry were calculated with the use of the mean counting efficiency of 1.37 cpm/nCi, although the results of the measurements in the arc are considered to give the better estimates of the burdens for these cases. The unweighted mean fractional radon retention ($RaC/^{226}Ra$) for the 6 cases is 0.36 at both laboratories.

The CHR breath measurements were made by the Radiochemistry Group under the direction of H. F. Lucas, Jr. and R. B. Holtzman. The measurements at M.I.T. were carried out under the direction of Prof. R. D. Evans.

Summary and Conclusions

Counting efficiency data for eleven persons with radium C burdens ranging from 48 nCi to 1090 nCi were compared with "average body thickness,"

TABLE 4. Interlaboratory Comparison of Radium Body Burden Measurements

Case	Laboratory	Date measured	RaC (γ), μCi	Rn (breath), μCi	^{226}Ra burden, μCi	RaC/^{226}Ra, μCi/μCi	MsTh (γ), nCi
01-431	MIT	8,9 June '71	0.301 ± 0.009 (chair)	0.464 ± 0.030	0.765 ± 0.031	0.39	0.1 ± 0.1
	CHR	14 June '71	0.262 ± 0.003 (chair)	not done			0.5 ± 0.5
	CHR	15 June '71	0.297 ± 0.007 (arc)	--	0.80 ± 0.02	assumed to be 0.37	
	Ratio	MIT/CHR	1.15 ± 0.037 (chair)				
01-144	MIT	20 July '71	0.283 ± 0.001 (chair)	0.464 ± 0.014	0.747 ± 0.014	0.38	0.3 ± 0.2
	CHR	2 Aug. '71	0.285 ± 0.006 (chair)	--			-0.3 ± 0.4
	CHR	2 Aug. '71	0.232 ± 0.009 (arc)	0.462 ± 0.004	0.694 ± 0.011	0.33	
	Ratio	MIT/CHR	0.993 ± 0.020 (chair)	1.00 ± 0.031			
01-208	MIT	3 Aug. '71	0.289 ± 0.012 (arc)	0.515 ± 0.088	0.80 ± 0.09	0.36	0.3 ± 0.2
	CHR	9 Aug. '71	0.294 ± 0.011 (arc)	0.636 ± 0.014	0.93 ± 0.02	0.32	-0.7 ± 0.5
	Ratio	MIT/CHR	0.983 ± 0.055	0.810 ± 0.140	0.86 ± 0.10		
01-017	MIT	12 Aug. '71	0.403 ± 0.016 (arc)	0.930 ± 0.020	1.33 ± 0.03	0.30	5.9 ± 0.9
	CHR	24 Aug. '71	0.421 ± 0.011 (arc)	0.790 ± 0.064	1.21 ± 0.065	0.35	3.7 ± 0.7
	Ratio	MIT/CHR	0.957 ± 0.045	1.18 ± 0.10	1.10 ± 0.06		
03-727	MIT	12 Jan. '72	0.061 ± 0.001 (chair)	0.090 ± 0.012	0.151 ± 0.012		-0.1 ± 0.2
	MIT	27 Jan. '72	0.073 ± 0.006 (arc)	0.091 ± 0.004	0.164 ± 0.007	0.45	
	CHR	31 Jan. '72	0.065 ± 0.002 (chair)				0.0 ± 0.3
	CHR	31 Jan. '72	0.063 ± 0.005 (arc)	0.101 ± 0.003	0.164 ± 0.006	0.38	
	Ratio	MIT/CHR	1.16 ± 0.13 (arc)	0.90 ± 0.046	1.00 ± 0.05		
01-403	MIT	1,2 July '70	0.010 ± 0.0011 (chair)	0.027 ± 0.001	0.037 ± 0.002	0.26	-0.1 ± 0.3
	CHR	30 Sept. '71	0.012 ± 0.0014 (chair)	0.016 ± 0.001	0.028 ± 0.002	0.43	0.5 ± 0.2
	Ratio	MIT/CHR	0.82 ± 0.13	1.7 ± 0.2	1.3 ± 0.1		
Weighted mean	Ratio	$\bar{x} \pm \sigma_{\bar{x}}$	1.02 ± 0.03	0.99 ± 0.06	1.04 ± 0.06		

represented by $\sqrt{W/H}$, with effective depth of burial of radium C, and with the distribution of radium C in the body.

Data were compared with theoretical curves which assumed that the burden could be represented by a point source in the body. For the range of observed values of $\sqrt{W/H}$, this assumption leads to theoretical values which differ by no more than a few percent from values calculated with the assumption of uniform distribution of the activity in the body of mean thickness d.

The counting efficiency for the chair geometry seemed to be correlated with $\sqrt{W/H}$ to a higher degree than the theoretical curve would predict. This was found to be at least partly fortuitous, since some of the deviations could be explained by differences in effective fractional depth of burial of the activity, as determined from the ratio of the counting rates observed from the front and from the back of the subject in the arc configuration. Effects of gross nonuniformity in skeletal distribution of the activity, as determined from scanning studies, were also evident in the counting efficiency data.

A minimum relative standard error of about ± 8% in a radium C burden determined from measurements in the tilted chair, using a single value for counting efficiency, has been estimated from the data on the eleven cases. This error probably cannot be reduced unless scanning studies are done on the subject, but not enough scanning data are yet available to determine this.

The results of an interlaboratory comparison of body burden determinations on six cases are essentially satisfactory. All radium C burdens determined at CHR from measurements of subjects in the chair geometry have a possible bias of as much as +0.3 nCi, but a good estimate of this value awaits the completion of control subject studies.

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MEASUREMENTS OF ^{226}Ra IN HUMAN BONES

M. A. Essling, A. T. Keane, and D. R. Huff

Bones and bone parts from 20 exhumed skeletons were assayed for ^{226}Ra by γ-ray measurement. Results for four cases are tabulated. A correlation is shown between the trabecular/cortical nature of the bone sample and its radium concentration.

Total body burdens of radium can be estimated by measuring the radioactivity of parts of the skeleton. Work of this type has been done at M.I.T. on whole bones.[1,2] At this laboratory, the study has been expanded to measure the radium distribution in various bone parts in order to obtain information on the partition of radium between cortical and trabecular bone. Such data should also enable a more accurate prediction of body burdens from small pieces of bone, since frequently only parts of bone are available for counting.

The ^{226}Ra is assayed through γ-ray counting of ^{214}Bi(RaC) in equilibrium with ^{222}Rn, the 3.82-day decay product of ^{226}Ra. Radon is a gas and readily emanates from bone. Since the nuclide actually counted is a daughter of radon, any loss of radon will result in a low value for radium content. To prevent loss of radon, the bone is sealed in a copper can.[3] Several counts are made on the sealed can, usually with one count near the time of sealing and another when the radon is nearly in equilibrium. In this way it is possible to estimate the fraction of radon retained in the bone and to check for loss of radon from the can.

Figure 1 depicts the radon growth in a typical sealed can. The radon growth factor, x, plotted on the abscissa is

$$x = 1 - \exp(-\lambda T),$$

where λ is the decay constant for ^{222}Rn and T is the time between sealing and counting.

The amount of RaC present at the time of sealing (x = 0) relative to the amount present at equilibrium (x = 1) is the fractional retention of radon

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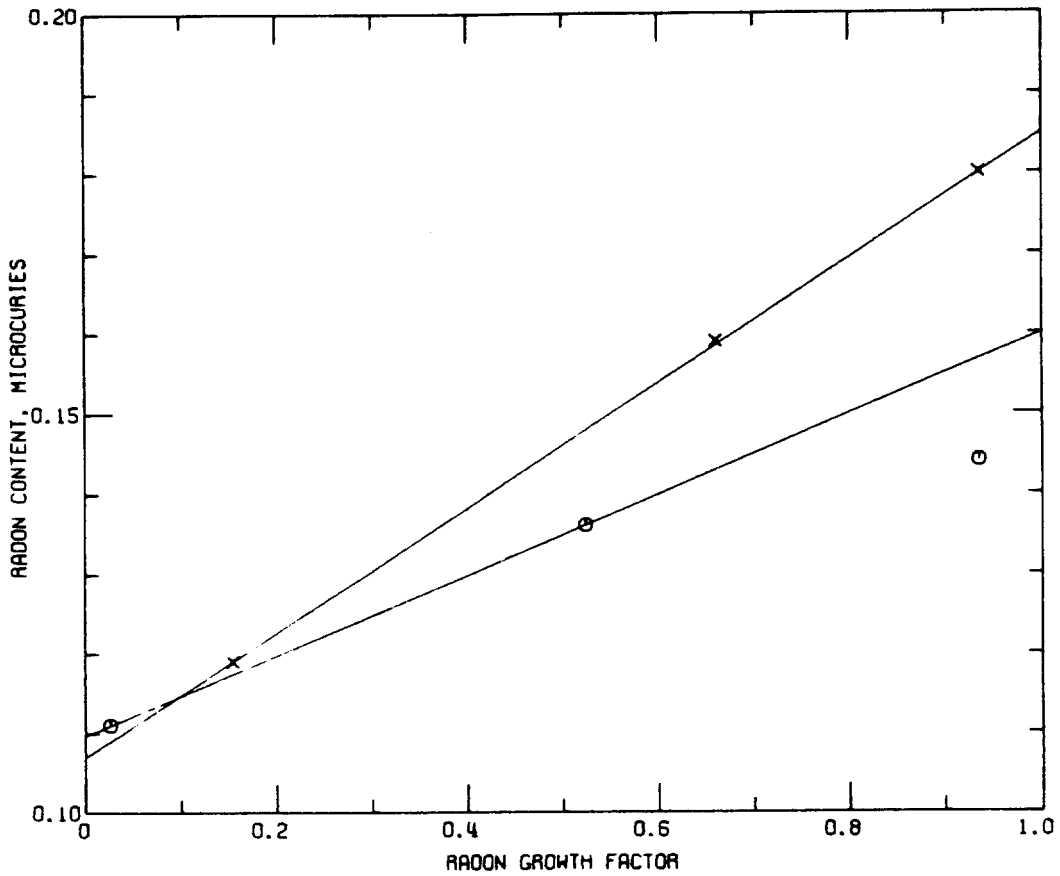

FIG. 1.-- Growth of radon in two groups of bones from case 00-008, showing how integrity of the sealing can be established. X, midshafts of long bones; O, ends of long bones, can leaking.

by the bone. To get an accurate radon retention value, the bone must be well ventilated before sealing. Also, the first count should be delayed a few hours to permit the decay of any RaB or RaC deposited on the bone surface by emanated radon.

Since the equilibrium value of radon is independent of the amount of RaC present when the bone is sealed, it may be determined by a single measurement several weeks after sealing. Nevertheless, a series of at least two measurements permits the determination of the radon retention, and a series of at least three measurements permits a check for radon leakage. When plotted as in Figure 1, the data for a leak-free can (crosses) will follow a straight line. It must be stressed that the

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determination of the true equilibrium radon value requires that the sealed container remain leak-free until the counting is completed. The circles in Figure 1 represent the radon growth in a can which appraently developed a leak sometime before the third count was made. The third data point lies well below the line drawn through the first two points. Since the validity of the first two counts could not be assumed, the sample was recanned and recounted. In fact, the radon equilibrium value obtained for the sample was the same as that indicated in Figure 1, demonstrating that the can did not leak until after the second observation.

After counting is completed, all cans are tested again for radon leakage by an independent method, namely, monitoring the air in a steel drum in which the cans themselves have been sealed.

The radioactivity of the bones was measured using either two 11.5-in diameter by 4-in thick NaI(Tl) crystals 83 cm apart or a single cylindrical ("log") NaI(Tl) crystal, 8 in long and 6 in in diameter, 1 meter from a Styrofoam table. The source-crystal distance is large enough in either configuration to reduce to negligible proportions any difference in counter efficiency for RaC deposited on the inside of the sealed can compared to that deposited inside the bone. The Styrofoam material was used for the can support in both configurations to minimize backscatter (150 to 200-keV region). If ^{226}Ra could be measured directly via its weak 186-keV γ-ray, the problems associated with sealing and counting bones in cans would be circumvented. The spectra collected in the course of this work included the peak from the 186-keV γ-ray and were thus suitable in principle for the direct determination of the radium-226 content. The Styrofoam support was used in anticipation of such an analysis of the data.

The necessary calibrations (allowance for different can sizes, bone attenuations, crystal distances, counting efficiencies, etc.) are now completed. Measurements have been made of radium in bones or complete skeletal collections from 20 cases; the results for four are shown in Table 1. The relative radium concentration nCi/g / nCi/g is the ratio of the

TABLE 1. Fractional Radon Retention and Relative Radium Concentration for Four Cases

Bone	Case No. 01-001 (a)	01-001 (b)	00-008 (a)	00-008 (b)	01-613 (a)	01-613 (b)	01-739 (a)	01-739 (b)
Skull, mandible	0.58	0.78	0.72	0.62	0.42	0.80	0.38	0.95
Cervical + lumbar vertebrae	0.84	1.63	0.67[c]	1.00[c]	0.45[c]	1.16[c]	0.41	1.90
Thoracic vertebrae	0.88	1.71	0.56	0.92	0.46	1.15	0.56[c]	1.63[c]
Sternum, manubrium	0.92	2.64	0.68	1.18	0.43	0.54	0.29	1.74
Iliac crests			0.61	2.70	0.43[c]	1.37[c]	0.48	1.07
Ribs	0.70	1.07	0.55[c]	1.67[c]	0.39[c]	1.08[c]	0.42[c]	1.03[c]
L. humerus	0.56	0.79	0.59	0.64	0.39	0.92	0.41	0.71
L. ulna			0.68	0.81	0.42	0.61	0.68[c]	0.47[c]
L. radium	0.67	0.55	0.62	1.34	0.43	0.75	0.31	0.55
L. tibia			0.58	0.58	0.39	0.87	0.52	0.60
L. fibula	0.41	0.59	0.53	0.85	0.62	0.64	0.17[c]	0.58[c]
Proximal + distal ends[d]			0.73	1.79	0.42	1.08	0.34	1.02
Midshafts[d]			0.57	1.00	0.50	0.44	0.39	0.53
R. femur, prox. end			0.73	1.79	0.40	1.42	0.33	1.11
R. femur, distal end			0.62	0.69	0.37	1.57	0.51	0.85
R. femur, prox. metaphysis	0.54	0.95	0.47	1.13	0.49	0.91	0.30	0.73
R. femur, distal metaphysis			0.41	0.55	0.21	0.81	0.25	0.61
R. femur, midshaft			0.42	0.81	0.64	0.72	0.33	0.64
L. femur	0.51	0.93			0.45	1.08	0.43	0.56
Misc. bones[e]			0.59	1.29	0.39[c]	1.30[c]	0.36[c]	1.32[c]

(a) Fractional radon retention (± 5% — see text for definition).
(b) Relative radium concentration (± 5% — see text for definition).
(c) Partial sample.
(d) Right humerus, ulna, radius, tibia and fibula.
(e) Innominates, hands, feet, clavicles, scapulae, sacrum, patellae.

radium concentration in the bone sample to the average concentration in the entire available skeleton.

The average and range of the relative radium concentration for each bone group are graphed in Figure 2. All four cases may not be represented in each category, since only those which were measured comparably were included in that grouping.

A correlation can be seen between the trabecular/cortical character of a bone and the average relative radium concentration. The mainly

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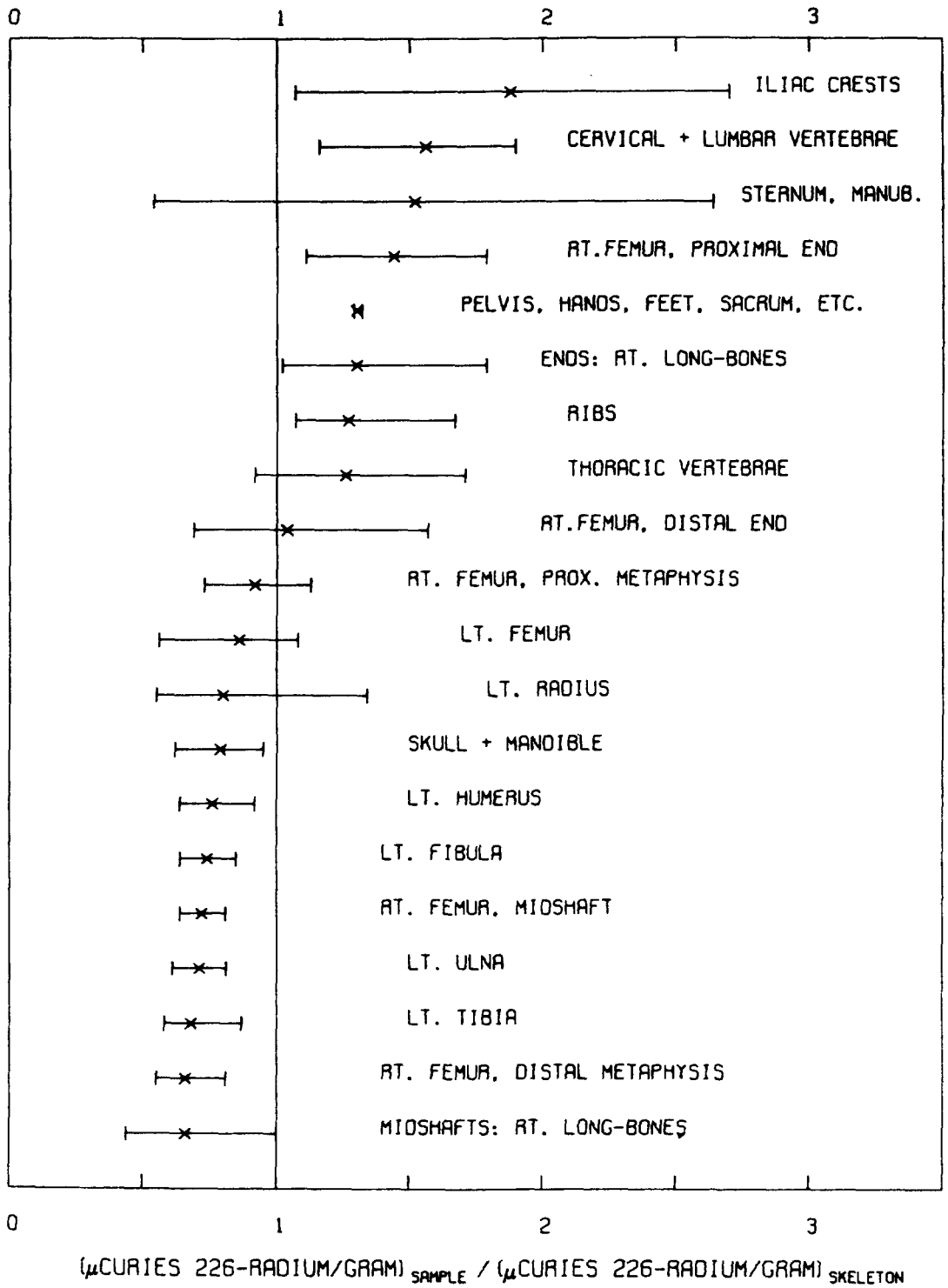

FIG. 2.--Radium concentration in various groups of bones from four cases relative to the concentration for the whole available skeleton, showing mean values and ranges (not errors).

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trabecular bones have values greater than 1, while those of a more cortical composition have values less than 1. This is observed for both whole bones and bone parts.

The ^{226}Ra contents estimated by skeletal measurement are shown in Table 2 for the four cases.

TABLE 2. Estimated ^{226}Ra Content of Four Exhumed Skeletons

Case	Year of death	Estimated fraction of total skeleton	Total skeletal content, μCi
00-008	1938	0.87	3.10 ± 0.18
01-001	1949	1.0	15.6 ± 0.7
01-613	1936	0.91	0.69 ± 0.04
01-739	1928	0.82	11.4 ± 0.9

The need to seal each bone or group of bones, to make repeated γ-ray measurements on each can and to check that the can is sealed is an extremely time-consuming procedure. If it should prove possible to assay the radium content of the bone via its 186-keV line, the procedure would be greatly simplified. This possibility is under investigation.

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MEASUREMENTS OF ^{210}Pb AND $^{210}Pb/^{226}Ra$ RATIOS IN HUMAN BONE IN VITRO

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Measurements are reported of the concentration of ^{210}Pb and of the $^{210}Pb/^{226}Ra$ ratio in human bone using the techniques of gamma-ray spectroscopy. The methodology for the determination of the $^{210}Pb/^{226}Ra$ ratio without direct evaluation of the corresponding activities is outlined, and the implication of this procedure is discussed.

Introduction

Detailed information on the distribution of ^{226}Ra and its decay product ^{210}Pb (half-life 22.3 years) in individuals with long-term body burdens of ^{226}Ra could confirm some of the metabolic parameters of ^{226}Ra and its gaseous daughter ^{222}Rn. Such information would not only be valuable in improving radium dosimetry (the ^{210}Pb-^{210}Bi-^{210}Po series may yield 10% or more of the effective dose from radium),[1] but it would also add to our knowledge concerning the metabolism of both ^{210}Pb and stable lead in man.

Presented in this paper are determinations of the concentration of ^{210}Pb in human bone in vitro and the $^{210}Pb/^{226}Ra$ ratio using gamma-ray spectroscopy. Lead-210 emits at 46.5-keV photon following 4.1% of the disintegrations, and we shall show that the low energy region of the gamma-ray spectrum contains all the information needed to determine the activity ratio without direct evaluation of the corresponding activities.

Experimental

Gamma-ray measurements were made on exhumed skeletal remains (skull bones) from four cases of radium poisoning. Information concerning the subjects is listed in Table 1. Figures 1 and 2 illustrate the low energy gamma-ray spectra observed with a 0.78 cm^2 x 1 cm Ge(Li) detector. With the exception of subject 01-010, the presence of ^{212}Pb (ThB) was indicated

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TABLE 1. Data on Radium Subjects

Case No.	Terminal body burden, μCi	Time radium burden carried, yr	Source of radium [a]	Storage time, yr
01-010	5.2	30	M.E.	15
01-014	2.24	33	D.P.	22
01-054	2.1	13	D.P.	32
01-390	7.4	6	R.D.	40

[a] M.E., Medical Exposure; D.P., Dial Painter; R. D., Drank Radithor.

in all subjects. Lead-212, which is present as a result of ^{228}Ra (mesothorium) exposure, emits a 238.6-keV photon which is partially resolved from the 242.0-keV line of ^{214}Pb (RaB). To ensure that the presence of ^{228}Ra and its daughters would not interfere with the ^{210}Pb measurements, spectra were taken of a thorium ore sample and a source of ^{228}Th. In addition to indicating no structure in the 46.5-keV region, these spectra confirm the identification of ^{212}Pb in the spectra from the radium subjects.

Calibration measurements were made with a source of ^{210}Pb distributed uniformly throughout a 5-mm thick slab of plaster of Paris at a concentration of 20.6 pCi/g; in the peak at 46.5 keV, a counting rate of 2.32 ± 0.09 min^{-1} was observed from this source. In calculating the ^{210}Pb concentrations shown in Table 2, the assumption was made that the dry skeletal weight was 40% of the fresh weight.[2]

The ^{210}Pb concentration cannot be corrected for the sample storage time (the time elapsed since death) without information concerning the concentration of the parent ^{226}Ra. Thus, a determination of the ^{210}Pb to ^{226}Ra activity ratio is of interest. The activity of radium could be obtained from the 186-keV line of ^{226}Ra observed in the spectra. However, the 53.2- and 242-keV lines of ^{214}Pb provide a means of determining the ^{210}Pb to

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and the 242-keV line of ^{214}Pb in bone are predominantly due to the Compton effect, which is a slowly varying function of energy. The mass attenuation coefficients in bone for the 186- and 242-keV quanta are 0.137 and 0.124 cm^2/g, respectively.[3] Since the samples approach infinite thickness with respect to the 46.5- and 53.2-keV radiations, a first order self-absorption correction could be applied, namely, the ratio of the mass attenuation coefficients, 0.395 and 0.321 cm^2/g in bone at 46.5 and 53.2 keV, respectively. The abundances of the various photons in the decay schemes can be obtained from the literature. Unfortunately, the abundance of the 53.2-keV line of ^{214}Pb is not accurately know,[4,5] and thus the factor, F, relating the ratio of the 46.5- to 53.2-keV counting rates to the activity ratio had to be determined empirically using the data of case 01-390. This case was selected since the initial intake of ^{210}Pb and the biological loss during the six years the radium burden was carried could be assumed to be negligible with respect to the ingrowth of ^{210}Pb during the forty-year storage period. It was assumed that the average retention of radon over the forty-year storage period was not significantly different from the recent retention experienced by the sample. With these assumptions the $^{210}Pb/^{214}Pb$ activity ratio, R_1, would be given by $1 - e^{-\lambda t}$, where λ is the ^{210}Pb decay constant, and t the storage plus burden time of 46 years; thus, $R_1 = 0.76$. A ^{210}Pb to ^{214}Pb counting rate ratio of 2.03 ± 0.23 was determined for case 01-390, from which we deduce a value for F of 0.37 ± 0.04. Included in this factor are the relative detection efficiencies, the abundances of the gamma rays, and the self-absorption corrections.

The observed counting rates in the various peaks (46.5 keV from ^{210}Pb, 53.2 and 243 keV from ^{214}Pb, and 186 keV from ^{226}Ra) are listed in Table 3. To illustrate the calculation of the ^{210}Pb to ^{226}Ra ratio, consider case 01-010. From Table 3, ^{210}Pb to ^{214}Pb and ^{214}Pb to ^{226}Ra counting-rate ratios of 2.59 ± 0.25 and 0.530 ± 0.017 were calculated. The activity ratio R_1 is then

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TABLE 3. Net Counting Rates (Counts/Minute) in the Four Gamma-Ray Peaks

Case No.	Gamma-ray energy			
	46.5 keV (^{210}Pb)	53.2 keV (^{214}Pb)	186 keV (^{226}Ra)	242 keV (^{214}Pb)
01-010	16.47±0.70	6.35±0.56	20.41±0.46	10.83±0.24
01-014	5.22±0.13	1.30±0.07	6.62±0.10	2.54±0.03
01-054	8.90±0.21	2.98±0.16	12.04±0.17	5.78±0.81
01-390	18.20±0.99	8.95±0.87	23.98±0.79	14.51±0.45

$$R_1 = (2.59 \pm 0.25) \times F$$
$$= 0.958 \pm 0.143 \ .$$

The high energy activity ratio R_2 was computed without applying any self-absorption corrections or source detector geometry factors. The abundances of the 186- and 242-keV lines are 3.3% and 7.4%, respectively, with relative detection efficiencies of 3.91 and 2.25, respectively. Thus, the ^{214}Pb to ^{226}Ra activity ratio for case 01-010 was calculated as

$$R_2 = (0.521 \pm 0.017) \times \frac{0.033}{0.074} \times \frac{3.91}{2.25}$$

$$= 0.411 \pm 0.013.$$

The measured ^{210}Pb to ^{226}Ra activity ratio, R_m, was given by

$$R_m = R_1 \times R_2$$
$$= (0.958 \pm 0.143)\ (0.411 \pm 0.013)$$
$$= 0.394 \pm 0.060 \ .$$

The low energy activity ratio, the high energy activity ratio, and the ^{210}Pb to ^{226}Ra ratio for the various subjects are shown in Table 4.

Also listed in Table 4 are the terminal ^{210}Pb to ^{226}Ra ratios. These values, corrected for the growth of ^{210}Pb from the ^{226}Ra during the period the specimens were stored, were calculated from the expression

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TABLE 4. Activity Ratios and Terminal $^{210}Pb/^{226}Ra$ Ratios

Subject	$^{210}Pb/^{214}Pb$	$^{214}Pb/^{226}Ra$	$(^{210}Pb/^{228}Ra)$, R_m	$(^{210}Pb/^{226}Ra)$, R_0
01-010	0.958 ± 0.143	0.411 ± 0.013	0.394 ± 0.060	0.356 ± 0.054
01-014	1.486 ± 0.183	0.2973 ± 0.0057	0.422 ± 0.055	0.424 ± 0.053
01-054	1.11 ± 0.14	0.3720 ± 0.0052	0.412 ± 0.052	0.329 ± 0.042
01-390	0.752 ± 0.117	0.469 ± 0.021	0.353 ± 0.057	0.088 ± 0.014

Unweighted mean radon retention = 0.387 ± 0.072.

$$R_0 = [R_m - f(1 - e^{-\lambda t})]\, e^{\lambda t},$$

where f is the fraction of radon retained during storage, λ is the ^{210}Pb decay constant, and t is the storage time. Because of the long storage periods, the values of R are sensitive to the assumed radon retention value. The standard deviation for the spread of the values of the ^{214}Pb to ^{226}Ra ratio (mean 0.387) was ± 0.072, or about 19%, which may indicate the variation one could expect in the average retention of the radon in bones from various subjects over the storage period. For two cases, 01-014 and 01-054, a ^{210}Pb to ^{214}Pb activity ratio greater than unity was observed. These subjects also exhibited the lowest recent radon retention. The unsupported ^{210}Pb was considered to be unsupported only in terms of retained ^{214}Pb, not ^{226}Ra, implying that these bones had experienced storage periods of considerably higher radon retention than reflected in the recent retention. The terminal ratios were calculated with the assumption that the fraction of radon retained during storage was the same as the recent retention of radon in the skull of case 01-390, i.e., 0.469.

Discussion

The terminal ^{210}Pb to ^{226}Ra ratios are shown in Figure 3 as a function of burden time. The points shown as crosses are the data of

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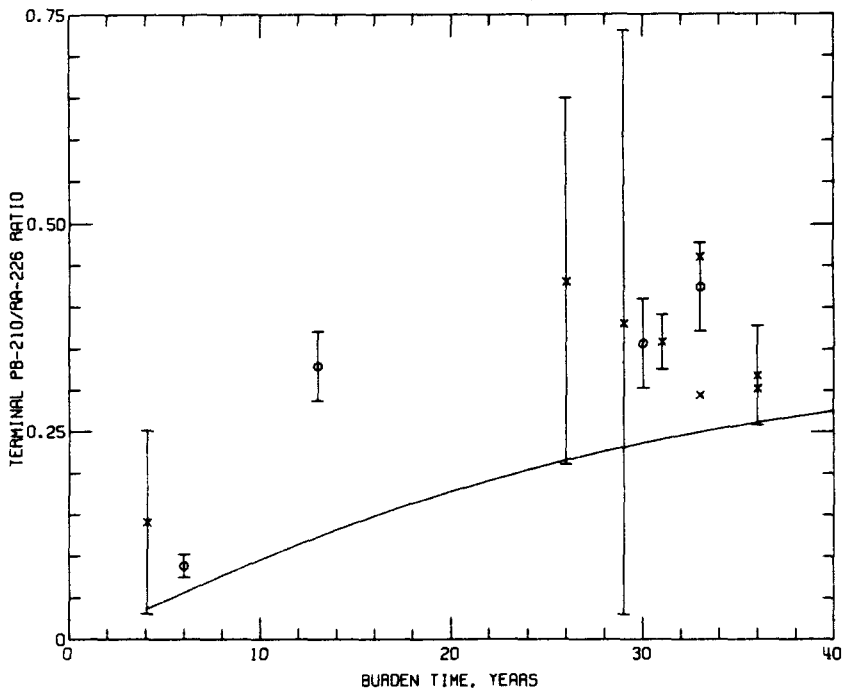

FIG. 3.--Experimental values of the terminal ratio activities of ^{210}Pb and ^{226}Ra as a function of time after intake, as determined by chemical analysis (x, Ref. 1) and by gamma-ray spectrometry (O, this work), compared with the theoretical curve (Ref. 1).

Holtzman[1] as measured by the chemical extraction of ^{210}Pb. The ^{226}Ra activity was determined by the radon emanation method of Lucas.[6]

The smooth curve of Figure 3 is a theoretical representation of the change in the ^{210}Pb to ^{226}Ra ratio with burden time in vivo calculated from the radon retention curve of Norris et al.,[7]

$$R_{Rn} = 0.021\ t^{-0.28},$$

their radium retention equation,

$$R_{Ra} = 0.54\ t^{-0.52},$$

and an exponential loss of ^{210}Pb with an effective half-life of 15 years. The expression for the curve is

$$R_0 = \frac{0.021\,\lambda e^{-\lambda_e T}}{0.54\,T^{-0.52}} \int_1^T e^{\lambda_e T}\, t^{-b}\, dt\,,$$

where T is the burden time, λ is the physical decay constant of ^{210}Pb ($0.693/22.3\ y^{-1}$), and λ_e is the effective decay constant of ^{210}Pb ($0.693/15\ y^{-1}$). The values of this function at various burden times were determined by Holtzman using numerical integration.

In general, the terminal ratios determined by gamma-ray spectroscopy appear to conform to the nature of the theoretical curve. As pointed out by Holtzman,[1] slight changes in the empirical constants in the above expressions will change the location of the curve to give an apparent better fit to the data. The error bars (standing error) associated with the gamma-ray spectroscopy data are based solely on the counting statistics. The standard errors of the data of Holtzman are based on the variation observed between duplicate samples. Uncertainties introduced in the computation of the terminal ratios for the long storage times of the subjects measured here far exceed the uncertainties arising from the counting statistics. The former uncertainties are, of course, common to both analytical techniques and are reduced considerably for samples of recent origin.

The ^{226}Ra activity concentrations for the various cases were calculated from the measured ^{210}Pb to ^{226}Ra ratios, R_m, and the ^{210}Pb activity concentrations in Table 2. The results are shown in Table 5, together with the reported activity concentrations of the skull bones of the various subjects. Our values are somewhat lower, but additional measurements would be required to confirm a systematic error. The activities calculated here depend not only on the assumptions used in the calculation, but also on the ^{210}Pb calibration and the assumed 40% wet-to-dry skeleton conversion applied in calculation of the ^{210}Pb concentration.

Conclusions

The above analysis was carried out to indicate the potential of the Ge(Li) spectrometer in studies of radium poisoning. The determination of

TABLE 5. Radium Concentrations in the Four Skull Bones

Subject	^{210}Pb, pCi/g	R_m	^{226}Ra, pCi/g	Reported ^{226}Ra, pCi/g
01-010	366 ± 21	0.394 ± 0.060	929 ± 153	1180 ± 120
01-014	115.8 ± 5.3	0.442 ± 0.055	262 ± 35	374 ± 37
01-054	197.6 ± 9.0	0.412 ± 0.052	480 ± 64	590 ± 59
01-390	404 ± 27	0.353 ± 0.057	1140 ± 199	1810 ± 180

the ^{210}Pb to ^{226}Ra ratio without the establishment of the corresponding calibration factors is not just an interesting use of the information content of the spectra, but a highly desirable feature with implications for further applications. For example, the nondestructive nature of the assay and the freedom from calibration factors, which in gamma-ray spectroscopy are notorious functions of sample geometry, would permit a study of a macroscopic $^{210}Pb/^{226}Ra$ distribution in the various bones of the skeleton to be performed with relative ease. Such a study could establish the validity of in vivo radon retention measurements; this parameter is of fundamental importance in the radium studies as it is used in the calculation of radiation doses.

As noted above, two samples exhibited a $^{210}Pb/^{214}Pb$ ratio greater than unity. It is of interest to note that these cases (01-014 and 01-054) were both dial painters. Storage of the samples at a high radon retention was assumed. However, inhalation of radon daughter products in the working environment of the dial painter is an additional possibility. Further measurements on other former dial painters would be of interest to confirm these observations.

The determination of the $^{210}Pb/^{226}Ra$ ratio by gamma-ray spectroscopy as outlined above could only be accomplished with a Ge(Li) spectrometer. To resolve the 46.5- and 53.2-keV lines, a resolution better than

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2.3 keV is required, if the signal window for each line is three times the FWHM. Ge(Li) detectors of 10- and 15-cm^2 area have recently become commercially available, with resolutions a factor of three to four times better than the above requirement. With such systems an order of magnitude reduction of the counting times would be possible, thus implying that ^{210}Pb to ^{226}Ra determinations in vivo would be feasible, for high body burden cases.

We wish to thank M. A. Wahlgren for use of the Ge(Li) detector. This work formed part of a thesis submitted by K.F.E. to the Graduate School of Northwestern University in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

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EXPOSURE DATA FOR RADIUM PATIENTS

Table 1 presents a summary of exposure data known as of 31 December 1971 for 1,032 radium cases under study at the Center for Human Radiobiology. It includes all persons measured since the start of the Center in 1969 and all persons for whom we have known measurements of radium from earlier work at the Radioactivity Center of the Massachusetts Institute of Technology, the Argonne Radium Studies at Argonne National Laboratory, and the Argonne Cancer Research Hospital. Some of the cases were first studied by the Radium Research Project of the New Jersey Department of Health, and additional persons from that project will be added to the list after study of records recently made available. Exposure histories and body burdens for 65 persons measured in the period 1 July, 1971 to 31 December, 1971 are given in Tables 2 and 3, and data for these persons are included in the first table.

The cases are listed in order of identification number in Table 1. Column 7 gives the amount of initially acquired radium expressed as microcuries per kilogram of bone, a quantity[1] calculated by applying the Norris retention function[2] for body burdens which were measured long after the initial uptake in almost every instance. The values shown differ slightly (less than 0.5 percent) from those in our first list of exposure data[3] because of small changes in the method of calculation and rounding off to fewer places. The cumulative rads listed in column 8 refer to the average ionization dose to the skeleton[4] calculated to death or to 1971 for the living subjects. Since the previous list carried the calculation only to 1969, doses calculated to 1971 show increases of about 3 to 15 percent, depending on the dates of intake of radium. Standard skeletal masses of 5 kg for women and 7 kg for men were utilized to obtain the results of columns 7 and 8. For purposes of comparison, it may be noted that a residual body burden of 0.2 μCi Ra, 40 years after intake, corresponds to initial uptake of about 10 μCi/kg and a dose of about 600 rads accumulated in 40 years to a 5-kg

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skeleton.

Since the data of Table 1 were obtained from examination of past histories and from radioactivity measurements of various kinds, some changes may be expected when new information or improved measurements of body burdens are obtained. Comparison with radioactivity values for our first list of 955 radium cases in the 1971 Annual Report shows appreciable changes for 22 of the 128 persons remeasured in the interval from 1969 to 31 December, 1971. Thirteen of those (numbers 01-142, 151, 176, 262, 278, 301, 368, 475, 599, 626, 05-631, 905, and 09-115) had previously been assayed only by radon breath or by analysis of a small bone specimen, and nine had previously been measured by whole-body techniques (numbers 01-035, 070, 075, 240, 403, 435, 03-470, 548, and 05-413). In addition, errors in data listing were corrected for 8 cases (numbers 01-485, 574, 03-222, 228, 238, 406, 583, and 03-678).

In the present format, data in Tables 1, 2, and 3 serve primarily to indicate the nature of the population under study. Future listings will be expanded to include significant medical data and estimates of error.

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