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Argonne National Laboratory

RADIOLOGICAL PHYSICS DIVISION SEMIANNUAL REPORT

July through December, 1960

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RADIOLOGICAL PHYSICS DIVISION SEMIANNUAL REPORT

July through December, 1960

J. E. Rose, Division Director
L. D. Marinelli, Associate Division Director

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BONE STUDIES ON AN EXHUMED RADIUM PATIENT

R. E. Rowland

Exhumation of radium poisoning victims has long been considered as a means of studying the early effects of radium in human bone. However, although many of the cases who died a few years after acquiring this isotope undoubtedly could be found, it is questionable whether the metabolic and dosimetric information obtained from such studies would be sufficiently precise unless the quantity of radium and the route of administration were known.

In this respect the Elgin State Hospital patients who were given Ra^{226} under controlled conditions in 1931[1] are ideal. After 20 or more years retention studies have been made on these cases,[2,3] and the distribution of radium in their bones has been studied by quantitative autoradiographic techniques.[4] In addition, a clinical investigation of these patients was reported in 1955[5] and is continuing to date.[6] Hence, any additional information that could be developed about this series would be of considerable value to the over-all problem of the effects of radium on man.

A careful review of the records of these patients has recently been made,* from which eight early deaths among the cases have been found. These patients are identified by code number in Table 1, with the date and quantity of radium administered, when known, and the date of death.

Table 1

Deceased Elgin State Hospital patients injected with Ra^{226}

Patient code	Date of death	Quantity of Ra^{226} administered	Year in which Ra^{226} was given	Remarks
03132	10/4/1931	Unknown	1931	Suicide
03129	5/6/1933	Unknown	1931	
03137	9/10/1934	Unknown	1932	
03130	3/26/1936	Unknown	1931-32	
03138	3/31/1936	Unknown	1932	Cremated
03123	2/5/1937	90 μc	1931	
03140	4/26/1937	Unknown	1933	
03120	6/25/1937	110 μc	1931	

*We are greatly indebted to the superintendent, Dr. Daniel Haffron, and the Clinical Director, Dr. Werner Tuteur, of the Elgin State Hospital, for permission to study the files of these cases.

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Unfortunately, for six of these cases the quantity of radium administered was not explicitly stated. In fact, three of these patients did not even receive their radium in 1931, but later, during 1932 or 1933. Consequently, the method of administration of Ra^{226} to these latter groups may not have been the same as it was for the 1931 group when the following schedule prevailed: 10 μc Ra^{226}, I.V., once a week for periods ranging from 8 to 45 weeks.

Nevertheless, when it appeared possible to obtain a bone sample from Case 03140, an exhumation was performed. The pertinent details of this case are listed in Table 2. In the records the following notation was found:

> "This patient has been on this ward about six weeks. Has received 11 injections of radium chloride. 4/10/33."

Table 2

Description of patients

	Case 03140	Case 03139	Case 03141
Sex	Male	Male	Male
Date of birth	February 9, 1905	1908	1907
Radium acquired	Age 28, 1933	Age 25, 1933	Age 26, 1933
Physical data	129 lbs., 5'7" (12/3/31)	140 lbs. (10/23/31)	127 lbs. (5/23/30)
Date of death	April 26, 1937, age 32	Living	Living
Cause of death	Pulmonary tuberculosis		
Exhumation date	October 13, 1959		

At exhumation a sample was removed from the right femur for study. It was found that the burial vault had not been penetrated by ground water, and that the preservation of the body tissues was remarkably good, considering that 22 years had elapsed since death. The following studies have been completed on the sample:

1. Total Ra^{226} content and radon retention in a 79-g cortical bone sample, by the spectroscopic measurement of the γ-rays emitted.

2. Specific activity measurements of the hotspots and the diffuse distributions by means of quantitative alpha-track autoradiographs.

3. Bone changes by means of microradiographs of 100-micron thick bone sections.

4. The microscopic distribution of radium by means of stripping-film autoradiographs.

The results of the γ-ray and the α-ray autoradiographic measurements are summarized in Table 3. From each set of measurements an extrapolation to the total skeletal radium content has been made; the γ-rays yield 0.53 μc, and the autoradiographs yield 0.55 μc. For both measurements the extrapolations are based on the assumption that the total skeletal mass is one tenth the body weight; usually extrapolations from gamma-ray measurements of small masses of bone are poor, but it is interesting to note that both measurements yield a similar body content.

Table 3

Summary of measurements (Case 03140)

From γ-ray measurements	
Radon retention	25.9%
Ra^{226} content (79 g bone)	0.090 $\mu\mu$c/mg bone
Extrapolated body burden	0.53 μc
From autoradiographic measurements	
Maximum hotspot intensity	10.8 $\mu\mu$c/mg bone
Average diffuse concentration	0.044 $\mu\mu$c/mg bone
Ratio: hotspot to diffuse	246.
Extrapolated body burden	0.55 μc

The autoradiographic extrapolation is based on the observation that the diffuse level is 0.47 of the average skeletal content;[7] this is probably the more reliable of the two methods mentioned above.

The stripping film autoradiographs showed that the intense deposits of radium in the bone samples were in narrow rings in haversian systems, averaging less than 50 microns in thickness. This is in marked contrast to the deposits in the other Elgin cases which have been studied,[4] where completely labeled osteons were often observed. It thus appears that the radium injections did take place over only a short time interval and that the average linear growth rate in replacement haversian systems was somewhat less than 10 microns per week.

The narrow width of these bands proved to be a serious deterrent to quantitative evaluation of the specific activity levels in the "hotspots." Since the average range of the radium alpha particle is about 30 microns,

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the tracks covered an area in the autoradiograph larger than the radium deposit actually covered in the bone section; therefore, the specific activity level listed in Table 3 is probably too low.

An indication that this is the case is given by the ratio of the maximum hotspot to the average diffuse level, which was found to be 246 in this patient. Previously a ratio of 218 was found in an Elgin patient who had been injected over an 18-week interval (Case I, Reference 7). Since this ratio is independent of the magnitude of the radium injections (as long as they are equal), but dependent upon the length of time the injections are continued, we would predict a six-week period of injections to produce a hotspot to diffuse ratio of magnitude close to 500.

The microradiographs of these bone sections showed no changes characteristic of radium damage. This was to be expected, for few such changes are seen in patients with 30-year burdens of less than 1 μc,[8] and, by comparison with the radium now present in Case 03139, (see below) the 30-year burden for our exhumed case would have been a small fraction of 1 μc. Thus, from this study we could not expect to determine how early the typical radium changes actually occur in human bone; an examination of bone from a case who had received a much larger quantity of radium is required.

One of the unusual features about this case is the fact that another patient, who received radium at the same time, is still alive and has been extensively studied and measured, and had been considered to be one of the classic 1931 cases. This latter case, coded as 03139,* was of similar age and build as 03140, as can be seen from the data in Table 2.

In the hospital records the following notations for case 03139 were found:

> "Patient is receiving (radium)** chloride intravenously. 3/31/33."

> "Patient has been on this ward about six weeks and has received 11 injections of radium chloride intravenously. 4/10/33."

A third case, whom we code as 03141, was also found who had received radium in 1933. In his records the notation read:

> "Patient has received 11 injections of radium chloride intravenously. Has been on this ward about six weeks. 4/10/33."

---

*This patient was coded as Paul N. in References 2, 5, and 9.

**On the typewritten record sheet the word "radium" had been inserted in ink, above the line, with a caret (^) to indicate that it belonged before the word "chloride."

This patient is still living in a state institution, but his total body content of Ra^{226} has not yet been measured.

These three cases, then, appear to have received identical radium treatments at the same time. Although we have no indication that the treatments actually terminated on 4/10/33, they may well have, for the fact that the date of the final notation in all three cases was the same may indicate that this treatment was being summarized.

It is of interest to compare the findings in our exhumed case with observations on the living patient in regard to total body burden. We first ask: How much radium did these cases actually receive? Granted that they received 11 injections over a 6-week period (i.e., two per week), and assuming that these were equal in magnitude, how large were they? Three possible magnitudes are suggested below with the reason for considering each alternative:

1) 10 μc Ra^{226} each injection:
(the same quantity as given in the 1931 series.)

2) 5 μc Ra^{226} each injection:
(half the magnitude given in 1931, to yield the same weekly dose.)

3) 3.3 μc Ra^{226} each injection:
(three cases were found who received radium at this time; one 10 μc ampule could provide three 3.3-μc doses each time the injections were given.)

In Table 4 are tabulated the expected body burdens at several times following administration, calculated from

$$R_t = 0.54t^{-0.52} \qquad \text{(Reference 2)}$$

for the three possibilities suggested above. Included for comparison are the measured values for Case 03139. The values of the total body content reported for 1951 and 1953 are now known to be incorrect, for subsequent gamma-ray measurements with large NaI(Tl) crystals in the low background room have yielded higher values for retained RaB and RaC than were found in the earlier measurements. Fortunately, it appears that the early radon breath measurements were valid, so that they may be employed to calculate the total Ra^{226} retained in the body. In Table 4 both the published and the recalculated values are shown; for the latter the assumption was made that 70% of the radon formed in the body is exhaled. The 1960 measurement was of the retained RaB and RaC only; the total Ra^{226} content is calculated from this measurement with the same assumption, namely, that 70% of the radon is lost. (See also a summary by C. E. Miller[9] regarding the early measurements on this and other Elgin cases.)

Table 4

Power function prediction of total body content of Ra^{226} following administration during March of 1933

Date	Predicted retention for three injection levels, μc			Measured retention, μc	
	36.3	55	110	Case 03140 (exhumed)	Case 03139 (living)
April 1937 (4 years)	0.446	0.615	1.35	0.53, 0.55	-
May 1951 (18 years)	0.205	0.309	0.62	-	0.46* (0.56)**
July 1953 (20 years)	0.194	0.292	0.585	-	0.52* (0.60)**
1960 (21 years)	0.167	0.250	0.50	-	(0.56)***

* Published total retention in Reference 2.

** Values from Reference 2 recalculated from radon breath measurements.

*** Based on an unpublished measurement of 0.167 μc (RaB + RaC) by C. E. Miller.

The conclusion is reached from examination of Table 4 that the total body measurements from Case 03139 are in excellent agreement with the assumption that he received 11 injections, 10 μc Ra^{226} each, totaling 110 μc Ra^{226}. It thus seems quite logical to assume that our exhumed case received the same total dose since the medical records are identical. Why, then, do the estimates of total skeletal content differ so from the predictions based on the power law?

Let us re-examine the concepts from which we extrapolated the total skeletal content. It is quite possible that the total radium content of our 79-g sample is not representative of the total skeleton, so that little reliability is based on this estimate. However, the magnitude of the diffuse level has been a reliable guide (within a factor of two) to the total skeletal content as measured in the human cases that have carried radium for 20 years or longer. If the total burden were actually 1.35 μc at time of death, as the power law predicts, then the observed diffuse level is only

0.19 times the average skeletal concentration. No value this low has been observed as yet although ratios as low as 0.23 have been found twice[7] (once for another Elgin case who had carried radium for 24 years, Case R).

The power law itself, however, was derived from data that show a spread of a factor of two over a 22-year period.[2] Thus, the difference between the expected and the calculated total body burden at time of death could be within the normal spread to be expected.

Measurement of Case 03141, the third patient who received radium in 1933, will be of great significance in the interpretation of the differences between the retention of radium in the exhumed (03140) and the living case (03139). Until further clarification of the administered and retained radium doses has been achieved, the validity of the total body radium content extrapolated from exhumation samples must be held in doubt.

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0016576

MEASUREMENT OF K^{42} IN HUMANS

C. E. Miller

The gamma-ray spectra of 20 subjects who contained known quantities of K^{42} were obtained with several different sized NaI(Tl) crystals located at various points around the body. These spectra were analyzed to determine the variations in counting rates (per μc of K^{42} in the body) that resulted with each patient-crystal arrangement when subjects of widely differing body builds were measured in order to assess errors inherent in various "geometries."

Twelve subjects were measured both prone and supine while curved on a 175-cm arc* and while seated in the tilting chair.(1) Also, six of these subjects were measured with a 3" x 5" NaI(Tl) crystal located 12 cm from the back at a sitting height elevation of 27 cm. The same deviations from the average counting rates were noted with the 175-cm arc technique as with the tilting chair technique. The counting rates (per μc in the body) were 2 ± 2% higher than the average for those subjects who weighed between 130 and 140 pounds, 2 ± 2% lower than the average for those who weighed between 180 and 200 pounds, and within ±2% of average for those who weighed between 150 and 160 pounds. With the 3" x 5" NaI(Tl) crystal at the small of the back, the counting rates varied from 12% above the average for the smaller subjects to 15% less than the average for the larger subjects. A variation of ±5% was noted for subjects of approximately the same height and weight.

Six subjects were measured while seated in the tilting chair first with an 8" x 4" crystal and then a 9" x 4" crystal over the chair above the subject. They were then measured with a 3" x 5" crystal located under the seat of the chair. The same variations from the average counting rate were observed with both the 8" x 4" and the 9" x 4" crystals. The counting rates observed with the 3" x 5" crystal deviated ±10% for subjects who gave counting rates within 1% of the average observed with the larger crystals.

The gamma-ray spectra were obtained from six patients, lying supine and prone with a 9" x 4" crystal placed at 7 equally spaced locations 24.5 cm apart along the body. (See Figure 10.) The crystal was first placed 30 cm and then 40 cm above the surface of the bed. The variations in counting rates between subjects observed at each crystal position were calculated as well as the variations that existed if combinations of crystals were used. The same percentage deviation occurred for each subject regardless of whether one crystal was used or whether an array of two or more crystals was used front and back, or both. The percentage deviations were about three times those observed when the same subjects were counted with the tilting chair technique.

*An 8" x 4" NaI crystal was located at the center of the 175-cm arc.

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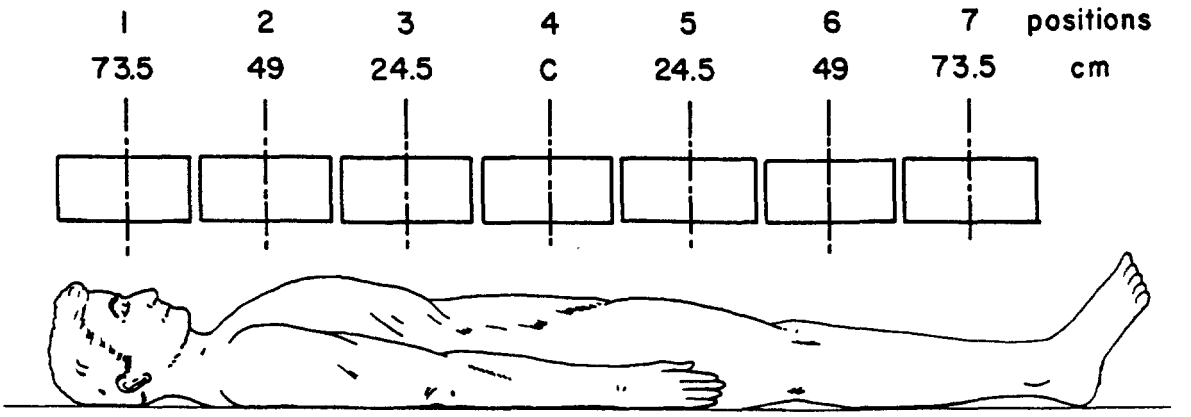

Figure 10

The counting rate observed with the crystal 40 cm above the surface of the bed was 71% of that observed with the crystal 30 cm above the bed. Thus it is not possible to eliminate or reduce the deviations from one subject to another by choosing a particular array of crystals at staggered heights from the subject. These data indicate that minimal variability is attained when patients are measured in the tilting chair.

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0016578

CESIUM-137 POTASSIUM-40 RATIOS IN EIGHT CONTROL SUBJECTS OVER A SPAN OF 30 MONTHS

C. E. Miller and J. B. Corcoran

The total-body gamma-ray spectra of 12 unexposed employees of the laboratory were measured with Human Spectrometer No. 1 on two dates during 1960. These subjects have been measured periodically, usually every 3 months, since March of 1955.

Their Cs^{137} body contents, expressed in units of $\mu\mu c$ Cs^{137} per gram of natural body potassium, have been determined, and the contents of 8 are given for 8 dates during the past 30 months in Table 5. It will be noted that the average Cs^{137} content dropped from 55 $\mu\mu c$ Cs^{137}/g K during December of 1959, to 49.6 $\mu\mu c$ Cs^{137}/g K during May of 1960, to 37.4 $\mu\mu c$ Cs^{137}/g K during December of 1960. This represents a 10% decrease in Cs^{137} content during the first 5 months of 1960 and a 25% decrease during the last 6 months. The same percentage decreases were noted for the other four subjects for these last three dates, but their data were not included in the table since they had not been measured every time on the earlier dates.

Table 5

Cesium-137 potassium-40 ratios in eight control subjects over a span of 30 months

Subject	7/31/58	11/20/58	3/19/59	6/17/59	9/14/59	12/8/59	5/24/60	12/15/60
C. L.	57	48.5	59	68.5	60	56.2	54	38.4
I. S.	51.5	52	59	54.6	54.5	47.4	49	44.6
H. M.	71.0	68.5	75.2	83.2	68.2	69.5	64.9	55.9
C. M.	43	58	76	81	64.2	62	50.5	33.9
B. O.	47	47.8	54.5	57.2	45.5	42.2	45.4	29.8
B. P.	43.5	46.2	53.5	59	54.7	53.5	48.2	31.6
R. R.	39	39.5	47	46	45.6	45.2	38.2	27.9
O. S.	48.4	51	61.8	65.5	64	63.1	46.6	37.3
Average	50	51.4	60.7	64.4	57.1	55	49.6	37.4

A group of 11 male Chicago residents, age 40 to 70 years, measured during the last 3 months of 1960 contained an average of 38.8 $\mu\mu c$ Cs^{137}/g K. This value agrees with the amount found for the controls during December of 1960. The spread in individual values was somewhat greater in that they ranged from 20 $\mu\mu c$ Cs^{137}/g K to 60 $\mu\mu c$ Cs^{137}/g K while the values of the younger controls ranged from 27.9 to 55.9--a 2 to 1 spread.

0016579

ANGULAR RESPONSE OF SCINTILLATION DETECTORS

Harold A. May

The response of a scintillation counter to gamma rays as a function of the angle of incidence may be useful for several reasons. In biological or environmental studies, extended sources must frequently be counted at small source-detector distances in order to reduce statistical uncertainties or counting time. Calibration sources of identical geometry and composition are required for greatest accuracy but may be difficult to devise; however, calculations to a precision of 25 to 50% are usually possible from a knowledge of the scattering and absorption properties of the matrix in which the radioactivity is located and the spatial response and absolute detection efficiency of the counter at the gamma ray energy to be observed.(1) Similarly, in relating the observed background counting rate within a given massive shield to the gamma rays originating in its walls, be it from cosmic-ray interactions or from its residual activity, the average response of the crystal to a uniform, omnidirectional flux must be known.

Some preliminary experiments have been performed to evaluate the response of a 7-inch diameter by 3.5-inch thick crystal, canned in a double-walled stainless steel container designed to provide a built-in one-half-inch thick mercury shield around the sides of the crystal. The mercury, which may be readily drained out, is separated from the crystal surface by 0.020-inch steel sheet and some 0.075 to 0.100-inch of MgO reflector. The walls are welded to stainless steel flanges at both ends, to which the $\frac{3}{8}$-inch quartz optical window and the 0.010-inch stainless steel gamma-ray "window" are gasketed and clamped by light steel rings. Since there is a fair amount of absorbing material near the corners of the crystal, its response is not likely to be typical of commercially packaged units.

Gamma-ray spectra of Ra^{226} + daughters, Cs^{137}, and Co^{60} were obtained as the angular position of the source, lying in a plane which included the crystal axis, was changed by 30° intervals between runs. The source distance was maintained at 70 cm; therefore, the full angular divergence of the gamma rays at the crystal was of the order of 14°. Total net counts (energy > 25 kev) and those falling in the photopeak were determined at each angular setting. Data from positions of symmetry about the axis (i.e. 30° and 330°, etc.) were averaged; in no case did the total count of either differ by more than $3\frac{1}{2}$% from their mean. The degree of collimation produced by filling the shield with mercury was also tested using Ra^{226} only. The resulting numbers, expressed as a fraction of the count obtained with the source at 0° (in front of the crystal, on axis), are tabulated in Table 6. Figures 11 to 13 present the same data in polar coordinate form.

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Table 6

Source	Cesium-137		Cobalt-60		Radium	
Energy range	25 - 910 kev (total)	410 - 910 kev (photopeak)	0.025 - 1.575 Mev (total)	0.87 - 1.575 Mev (both peaks)	0.025 - 1.57 Mev	0.025 - 1.57 Mev (Hg in housing)
Angle						
0°	1.0	1.0	1.0	1.0	1.0	1.0
30°	0.93	0.89	0.94	0.92	0.92	0.83
60°	0.81	0.72	0.84	0.76	0.80	0.45
90°	0.72	0.70	0.78	0.75	0.70	0.21
120°	0.74	0.62	0.87	0.64	0.71	0.34
150°	0.80	0.79	0.76	0.78	0.75	0.54
180°	0.65	0.60	0.73	0.58	0.61	0.46
I_{avg}		0.73		0.76	0.76	0.43

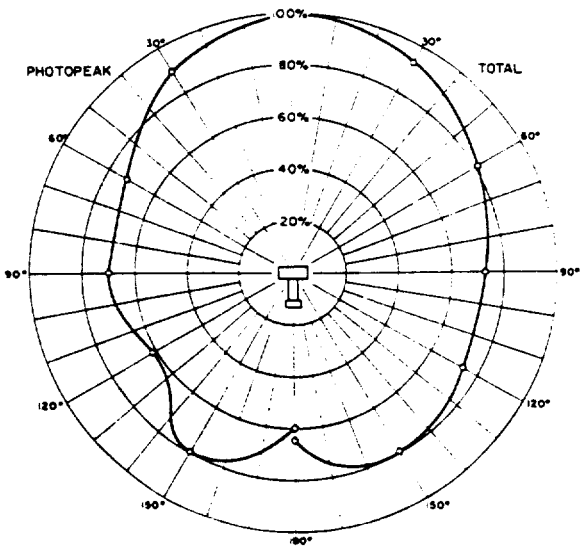

Figure 11
Angular response, Cs^{137}

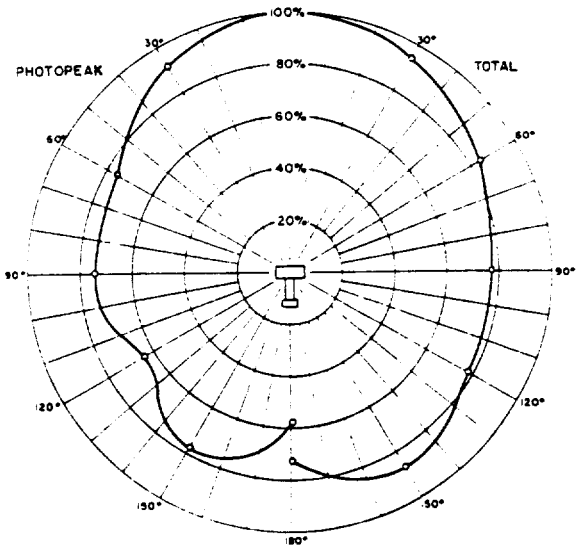

Figure 12
Angular response, Co^{60}

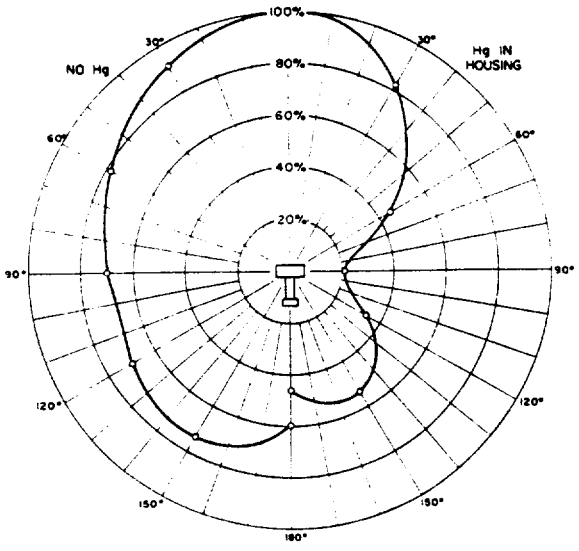

Figure 13
Angular response, Ra^{226} and daughters. Total counts

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The average response of the crystal to a uniform isotropic flux, relative to that of an isotropic detector with an efficiency equal to the maximum observed experimentally (i.e., at $\theta = 0°$), is given by:

$$I_{avg} = 1/2 \int_0^{2\pi} F(\theta) \sin\theta \, d\theta \quad .$$

Although $F(\theta)$, a continuous function having the observed values shown in the table at certain discrete angles, is unknown, an approximate value of I_{avg} may be calculated by means of graphical integration. The results of such calculations appear in the last row of Table 6.

The variation in counting rate with angle results from three separate effects. These are:

1. The solid angle subtended by the crystal, and hence the number of gamma rays striking it, is variable. This purely geometric effect may be readily evaluated for a simple right circular cylinder, but would be far more difficult to evaluate for the more complex contour of the crystal container.

2. The number of gamma rays passing through the crystal edges, and therefore traversing less than the average path length within it, also varies, with a resultant change in the interaction ratio. This also could be directly evaluated, in theory, by an extension of available computer-programmed Monte Carlo methods.

3. Interaction of gamma rays with the non-uniform mass of the steel container introduces an angular dependence of the number, energy distribution and directionality actually reaching the crystal.

While the results for cesium and cobalt sources are similar, the energy-dependent differences are readily seen. Minima in the photopeak curves, in the vicinity of 75° and 120° apparently are caused by the thicker steel of the container flanges. Further measurements at lower energies should demonstrate the same general behavior to an even greater extent.

The technical assistance of Paul Hess is gratefully acknowledged.

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0016582

CORRELATION OF THE NATURAL RADIOACTIVITY OF THE HUMAN BODY TO THAT OF ITS ENVIRONMENT: UPTAKE AND RETENTION OF Ra^{226} FROM FOOD AND WATER

Henry F. Lucas, Jr.

I. Introduction

Measurements of the natural radium content of man are of great importance in studies to evaluate the effects of small increases in the radiation dose resulting from skeletally deposited radioelements for two reasons: one, our knowledge of long-term radiation effects is largely confined to the effect of radium; two, as a natural radioelement found in our food, water and air, its uptake and retention is a product of a life-long natural environmental condition.

The relative contribution of food and of water to the whole-body content is required for a valid estimate of the body contents of the large populations known to consume water with a high Ra^{226} content, on which direct measurement is not possible. This is important since 35,000 people are known to consume water from municipal systems with from 10 to 37 $\mu\mu$g/liter Ra^{226}. An additional 800,000 people consume water with more than 1 $\mu\mu$g/liter, of which one-half or 400,000 consume water with from 4 to 10 $\mu\mu$g/liter.(1) These municipal waters were obtained from wells, some of which have been in use for 90 years. The Ra^{226} content of these waters can be compared to the currently accepted MPC for industrial workers of 100 $\mu\mu$g/liter and of large populations of 10 $\mu\mu$g/liter.(2)

To evaluate the relative contribution of food and of water on the whole-body content of humans, early measurements were made on a relatively small number of living subjects by a considerable extension of the radon breath method of analysis.(3) The living subjects measured in this work were tested in two experiments which were designed to determine: (1) the rate of Ra^{226} accretion by humans after reaching adulthood and (2) the relative accretion by two groups of youths whose drinking water contained 8 and 0.03 $\mu\mu$g Ra^{226}/liter respectively.

In the first of these, prisoners at Stateville Penitentiary, Illinois who had previously resided in Chicago or other areas with surface or low radium water sources were chosen. The body contents of new prisoners were compared to those who had been at the prison for up to 22 years, consuming water at 4 $\mu\mu$g/liter. This experiment demonstrated an increase of body content with duration of exposure. The average body contents after 4 months and 20 years were 1.0 and 2.4×10^{-10} g respectively.

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Proper interpretation of these and the following measurements requires the evaluation of the rate of radium intake. Direct measurements of radium intake were not possible. It was, however, apparent that for intake at a steady state, excretion must be within a very few percent of intake or a much higher body content would have been found. Thus, excretion rates can be considered equal to intake rates. The accretion of Ra^{226} from water for the 20-year prisoners corresponds to 17 times the daily intake or that in about 50 liters of water. However, prison dietary conditions are not entirely normal, and intake of radium from water was unusually high. The average Ra^{226} intake corresponded to that from food plus that in 3 liters of water. This results, in part, from the extremely frequent occurrence of soups, stews and boiled foods. A normal population might be expected to have a daily intake equal to that from food plus that in 1 to 2 liters of water. The 20-year uptake for this normal group would still correspond to 17 times the daily intake. The body content, however, would only be equal to that in 20 to 30 liters of water.

In the second experiment, the accretion of radium by 8 high school boys from Lockport, Illinois, (average age 17.5 years; 8 $\mu\mu g$ Ra^{226}/liter drinking water) was compared with that by 7 similar boys from Chicago (0.03 $\mu\mu g$ Ra^{226}/liter). The average value for each group was 3.7×10^{-10} g and 0.4×10^{-10} g whole body, respectively. From measurements of the rate of excretion it was determined that the whole-body content of the Chicago boys corresponds to 22 times the daily intake of radium, which necessarily came from food. Similarly, the whole-body content of the Lockport boys corresponds to that from food plus that in 40 liters of water or about 45 times the daily intake.

In this present work, measurements are being made of the Ra^{226} content of normal human bone, teeth and soft tissue to determine for a normal population the relative contribution of food and water to the whole body content.

II. Experimental Method

Ashing Procedure. For this study normal, uninvolved bone obtained during ordinary surgical procedures or at autopsy was used. The samples were shipped immediately without any preservative, thereby avoiding possible contamination from the local water source. On receipt the samples were scraped to remove soft tissue, dried at 110°C and stored at -27°C until analysis.

Samples selected for analysis were prepared in one of two ways, depending on the number and type of analyses to be performed. For those on which the Ra^{226} content only was desired, the following procedure was used. The dried, weighed sample was placed in a small uncovered platinum

crucible or dish and ashed at 600°C overnight in an electric muffle furnace lined with Crystalon* to reduce the possibility of the contamination from fire brick. The weighed ash was then transferred to 50-ml centrifuge cones and dissolved in 1.85 cc concentrated nitric acid per gram of bone ash and diluted with distilled water to give a solution having 0.2 g ash/ml. With one exception no residue was observed.

Samples on which both Ra^{226} and Th^{228} were determined were given a preliminary treatment to remove blood residues by boiling 3 times in freshly distilled ethylene diamine and 3 times in distilled water; they were dried at 110°C, then ashed and dissolved as previously described above. The thorium was extracted into dibutyl phosphate, while the aqueous solution containing the radium was transferred to an emanation flask and processed in the usual manner. No loss of radium or contamination was detectable.

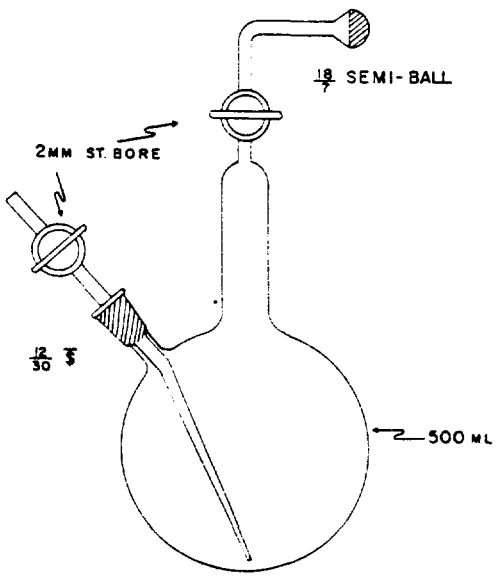

Figure 15

Emanation flask

Measurement. The bone solution was transferred to an emanation flask of the type shown in Figure 15, using 1 N HNO_3 to a final volume of 300 ml. The solution was flushed with a radon-free air, sealed and allowed to stand for at least 7 days.

The sample flask was attached to the transfer system shown in Figure 16 with water traps (B) and radon trap (C) dry-ice cooled. The system, with the exception of the sample flask, was evacuated to a pressure of ca. 1 mm Hg. Aged tank air which had been dried** and passed through a charcoal trap at dry-ice temperatures for radon removal was then used to purge the transfer system and transfer the radon from the flask to the charcoal trap. It is unnecessary to heat the flask since quantitative (99.9%) transfer of the radon was effected in 13 liters of the 30-liter flush routinely used.

The charcoal trap containing the radon sample was evacuated while still at dry ice temperatures with a mechanical pump to about 0.1 mm Hg in 7 min or less and the radon transferred to the alpha scintillation counter

*Cat. No. 204 MC Lindberg Eng. Co. E. H. Sargent and Co., 4647 West Foster Ave., Chicago, Illinois.

**Linde Molecular Sieve, Type SA, $\frac{1}{16}$" pellets, Linde Co., 230 N. Michigan Ave., Chicago, Illinois.

previously described.[4] This system has a sensitivity of 5.40 cpm/$\mu\mu$c radon and a background counting rate of less than 0.1 cpm. Using this system, it was possible for one person to process 16 to 20 radon emanations per day. All radon samples were counted for 7 to 16 hours. Repeated determinations were made if the first two failed to agree within the 0.9 statistical counting error.

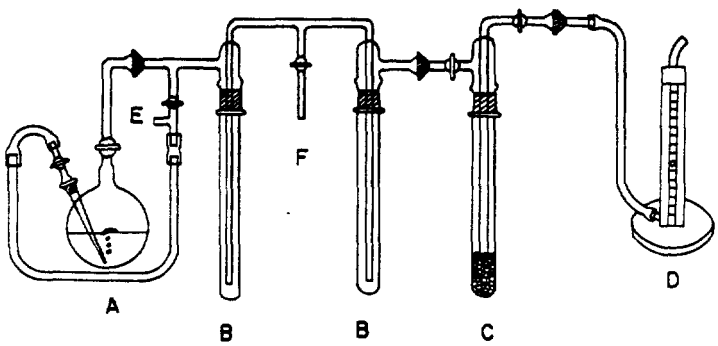

Figure 16

Radon collection system. A, Emanation flask; B, Dry-ice-cooled water freeze out traps; C, Dry-ice-cooled, charcoal-filled (7 g) radon trap; D, Flow rator, 0 to 2 l./min; E, Radon-free air input; F, Vacuum.

A system blank of 0.002 ± 0.002 $\mu\mu$c Rn was found if the rubber tubing used in the Sigma Motor pump to transfer radon from the charcoal trap to the radon counter bottle was replaced at start of sample processing each day with radon-free (aged in sealed jar) tubing. Reagent and distilled-water blanks were found, with some exceptions, to be less than 0.002 $\mu\mu$g, and no corrections have been made except for samples of less than 1 g ash.

The alpha scintillation radon counting system had been calibrated previously with an aliquot from a National Bureau of Standards certified radium solution.[3] Additional aliquots were evaporated on bone and, in eight measurements, 99 ± 2% of the radium was recovered, and thus, no correction for chemical recovery of radium from bone has been applied to the data.

III. Measurements

A. Uptake from food

To evaluate the effect of food on the content of radium in bone, subjects were selected who had lived all their lives in cities having surface water sources or wells with less than 0.1 $\mu\mu$g/liter Ra^{226}. The average concentration of Ra^{226} in bone ash from 42 individuals summarized in Table 12, is 0.0124 $\mu\mu$g/g ash. This corresponds to a whole skeletal content (2600 g ash) of 0.32×10^{-10} g. As has been found by other investigators[5,6] from a random sampling of non-occupationally exposed humans, no correlation of radium content with age, sex, or illness was observed.

Table 12

Ra^{226} content of human bone from food sources*

Sample number	Number of cities of residence	Age, years	Sex	Bone	g ash	Ra^{226}, $\mu\mu g/g$ ash ± E**
185	4	56	M	rib	1.30	0.005 ± 0.007
163	2	49	M	rib	7.47	0.005 ± 0.001
213	2	56	M	rib	3.65	0.006 ± 0.003
156	2	55	F	rib	2.85	0.006 ± 0.003
130	3	35	M	rib	6.62	0.006 ± 0.001
244	2	34	M	rib	5.51	0.007 ± 0.002
212	3	21	F	rib	2.85	0.007 ± 0.003
100	1	25	M		3.17	0.008 ± 0.003
122	1	38	M	rib	6.11	0.009 ± 0.002
126	1	46	M	rib	9.29	0.009 ± 0.001
243	1	45	M	rib	3.64	0.009 ± 0.003
246	4	58	M	rib	3.44	0.009 ± 0.003
211	3	52	M	rib	2.96	0.010 ± 0.003
215	1	70	F	joint	6.24	0.010 ± 0.002
158	2	82	F	rib	1.55	0.010 ± 0.007
338	1	70	M	tibia	5.83	0.010 ± 0.001
339	1	65	M	tibia	4.06	0.010 ± 0.002
152	1	72	F	rib	0.87	0.011 ± 0.022
286	3	61	M	tibia	7.40	0.011 ± 0.002
245	2	61	M	rib	6.47	0.011 ± 0.001
170	5	69	M	rib	4.54	0.011 ± 0 003
121	3	33	M	rib	5.48	0.011 ± 0.001
125	2	57	M	rib	7.51	0.012 ± 0.001
132	1	26	M	rib	4.99	0.012 ± 0.003
282	3	50	F	femur	9.75	0.013 ± 0.001
171	1	47	M	rib	6.68	0.013 ± 0.002
370	1	23	M	femur	7.24	0.013 ± 0.001
230	2	50	F	vertebra	6.19	0.014 ± 0.002
142	3	36	M	rib	7.40	0.014 ± 0.002
209	2	49	M	rib	6.67	0.014 ± 0.002
168	1	73	F	rib	0.98	0.015 ± 0.007
103	6	30	M	tibia	12.32	0.016 ± 0.001
160	1	54	F	rib	3.41	0.016 ± 0.003
228	1	0	F	vertebra	0.53	0.016 ± 0.016
216	1	67	F	cort.	6.16	0.016 ± 0.002
129	2	62	M	rib	8.83	0.017 ± 0.002
135	1	57	M	vertebra	2.92	0.019 ± 0.003
138	1	39	M	rib	4.84	0.019 ± 0.003
210	1	46	M	rib	3.41	0.020 ± 0.003
279	1	3	M	femur	1.68	0.021 ± 0.006
205	1	0	F	vertebra	2.29	0.022 ± 0.004
173	1	69	M	rib	1.71	0.027 ± 0.006
mean ± σ						0.0124 ± 0.005

*Ra^{226} content of drinking water equal to or less than 0.1 $\mu\mu g$/liter throughout entire lifetime.

**90% confidence interval.

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Walton[7] demonstrated that radium was distributed to within 15% of the average in each of 11 bones from 11 skeletons tested for variation. This information lends welcome validity to measurements of bone specimens as a means of estimating whole body content. However, as shown in Table 13 this is not always the case.

Table 13

Relative Ra^{226} content of human bone

Bone	Sample number					
	41A	41B	42A	42B	43A	43B
	Ra^{226}, $\mu\mu g/g$ ash ± E*					
Skull	0.070 ± 0.004	0.070 ± 0.004	0.100 ± 0.009	0.051 ± 0.002	0.028 ± 0.003	0.032 ± 0.003
Tibia	0.019 ± 0.003	0.014 ± 0.001	0.030 ± 0.002	0.025 ± 0.002	0.030 ± 0.002	0.026 ± 0.002
Jaw Bone	0.016 ± 0.002	-	0.025 ± 0.002	-	0.027 ± 0.003	-
Teeth	0.019 ± 0.003	-	0.026 ± 0.002	-	0.027 ± 0.003	-
Rib	0.012 ± 0.001	-	0.028 ± 0.002	-	0.030 ± 0.002	-
Birthplace	Nebraska		Connecticut		Florida	

*90% confidence interval.

In two of these three subjects the radium content is seen to vary significantly from bone to bone. No difficulties were encountered in analysis, and the differences in the duplicate analyses on sample numbers 41 and 42 are statistically significant. Since the radium content of individual bones has been found to vary considerably after single injections,[8] it is probable that these differences represent changes in the environment. While the city of residence during their early years could not be determined, well waters with high radium content have been found by Barker[9] in both Nebraska and Connecticut, and it may be presumed that these individuals consumed water of high radium content. These samples were provided by Dr. J. B. Hursh and while they were not a part of his earlier study,[5] they have nearly identical Ra^{226} contents.

In order to extrapolate from bone to the whole-body content, measurements of radium in soft tissue are required. As shown in Table 14, the average radium content of soft tissue ash is 0.8 that of bone ash. This is in agreement with measurements by Radley, Turner and Mayneord[10] and Hursh et al.,[11] but only about 3% of that found by Muth et al.[12] Since the skeleton constitutes 86% of the whole-body ash, only 10 to 15% of the radium will be found in the soft tissues of the body.

The 0.04×10^{-10} g found in soft tissues added to the 0.32×10^{-10} g found for the skeleton gives a whole-body content of 0.36×10^{-10} g. This is in excellent agreement with the 0.4×10^{-10} g found previously by the

radon breath technique.[3] Using the previously determined daily intake rate for humans in Chicago, Illinois, of 1.6 $\mu\mu$g/day, this whole body content corresponds to 22 times the daily intake and necessarily comes from food.

Table 14

Ra^{226} content of human soft tissue

Sample number	Bone		Soft tissue			Ra^{226} ratio
	g ash	Ra^{226}, $\mu\mu$g/g ash	g wet	g ash	Ra^{226}, $\mu\mu$g/g ash	Tissue ash / Bone ash
216	6.16	0.016 ± 0.002	101*	0.794	0.020 ± 0.008	1.25 ± 0.42
338	5.83	0.015 ± 0.0015	412**	4.06	0.0069 ± 0.0025	0.66 ± 0.24
339	8.22	0.0129 ± 0.0010	409**	4.06	0.0096 ± 0.0015	0.74 ± 0.18
Weighted average						0.81

*muscle and fat
**muscle

B. Uptake from water

Measurements have been completed on 35 bones from individuals who lived either all their lives, or not less than the last thirty years, in a city with high radium drinking water. The radium content of these samples is summarized in Table 15. The radium content varied from a low of 0.012 $\mu\mu$g/g ash for a resident of a city with 0.13 $\mu\mu$g/liter in the drinking water to a high of 0.47 $\mu\mu$g/g ash in a city with 10 $\mu\mu$g/liter of water.

The correlation of the radium content of these samples with that in the drinking water is shown in Figure 17. For all data plotted in this figure it was assumed that 0.012 $\mu\mu$g/g ash was normally obtained from food and has been subtracted from the value found in Table 12. An excellent correlation of radium in bone with that in the water supply is obtained for drinking water with more than 1 $\mu\mu$g/liter. For less than 1 $\mu\mu$g/liter, normal statistical variations from food uptake overlap the uptake from water, and the scatter of points is not statistically significant. Extrapolating to a Standard Man (3000 g ash, whole body), uptake from water equal to that in 50 liters of drinking water would seem to be indicated as a suitable average.

Variations in sources of municipal water or its treatment have significant effects on the level of radium. The value reported for each city is the average of samples from all wells in use over the past 3 or more years. In two cases even this is not a very satisfactory method of

estimating the average radium content of the drinking water. In the first, City No. 13, Table 15, a water-softening system was installed two years ago, which removed 60 to 70% of the radium. In the second, City No. 11, Table 15, all of the wells in the Northeast part of the city have 2 to 4 times the radium content of those in the rest of the city. The radium content of water samples from the distribution system correlates with that of the nearest active well and, until a better estimate can be made, the average of the four samples from the distribution system will be used.

Table 15

Ra^{226} content of human bone from cities with high radium drinking water

City number	Drinking water, $\mu\mu g/l.$	Age	Residence		Sample number	Bone	g ash	Ra^{226}, $\mu\mu g/g$ ash	Mean, $\mu\mu g/g$ ash
			Years	City code					
1	0.13	9	9	KR08-4	280	femur	3.56	0.012 ± 0.003	-
2	0.2	72	43	2GN1RG1L-1	110	rib	2.46	0.015 ± 0.004	
	"	36	35	"	109	rib	2.28	0.017 ± 0.005	0.024
	"	0	-	"	229	vertebra	0.19	0.031 ± 0.037	
	"	0	-	"	240	"	1.27	0.032 ± 0.008	
3	0.4	11	11	2N0TS2P3P-5	384	rib	6.51	0.013 ± 0.002	-
4	0.48	12	12	13R02P-1	386	humerus	1.76	0.033 ± 0.006	-
5	0.85	75	75	R2TSB2W-2	284	femur	4.44	0.033 ± 0.005	-
6	1.8	12	12	IN0T160-5	283	"	4.69	0.020 ± 0.003	-
7	2.5	33	33	5R2P-1	172	"	1.04	0.055 ± 0.010	
		87	63	"	145	"	2.38	0.082 ± 0.006	0.081
		25	25	"	118	vertebra	6.68	0.105 ± 0.002	
8	3.1	65	65	1N0N36-5	281	talus	3.75	0.052 ± 0.005	-
9	4.6	64	33	161TT5-1	249	rib	1.97	0.044 ± 0.005	
	"	68	68	"	175	"	1.30	0.058 ± 0.011	
	"	79	55	"	226	"	1.83	0.060 ± 0.007	
	"	65	30	"	275	"	2.04	0.094 ± 0.007	
	"	69	69	"	120	femur	4.88	0.100 ± 0.008	
	"	50	50	"	164	rib	2.09	0.118 ± 0.008	0.112
	"	71	40	"	293	"	1.89	0.130 ± 0.012	
	"	44	44	"	201	"	2.57	0.147 ± 0.008	
	"	66	66	"	194	"	3.20	0.170 ± 0.007	
	"	47	34	"	235	"	2.57	0.200 ± 0.088	
10	5.0	66	66	TSR5HML2-1	124	iliac	5.10	0.078 ± 0.003	-
11	5.0	82	80	1R4R51-1	111	tibia	10.98	0.042 ± 0.002	
	"	48	48	"	112	rib	2.09	0.047 ± 0.003	
	"	61	61	"	150	"	7.37	0.080 ± 0.002	0.069
	"	72	55	"	165	"	2.23	0.083 ± 0.004	
	"	53	53	"	113	"	4.52	0.095 ± 0.003	
12	8.2	44	44	KN0N3M-1	197	rib	1.60	0.056 ± 0.009	-
13	8.5	84	35	8BS2LG0-1	248	rib	1.87	0.042 ± 0.005	
	"	54	54	"	169	"	2.23	0.137 ± 0.014	0.137
	"	58	58	"	272	"	1.95	0.191 ± 0.016	
14	10.5	54	54	2LL3VN2RG-1	148	rib	4.05	0.472 ± 0.010	-

In addition, care must be taken to check as often as possible the actual level of radium consumed at work and at home. Home-type water softeners of the ion-exchange type removed approximately 98% of the radium. For instance, Subject No. 117 lived in a house in which both hot

and cold water were softened. The Ra^{226} content of 0.013 ± 0.002 $\mu\mu g/g$ ash found correlates with the 0.01 $\mu\mu g$/liter actually consumed and not with the 5 $\mu\mu g$/liter of the city supply.

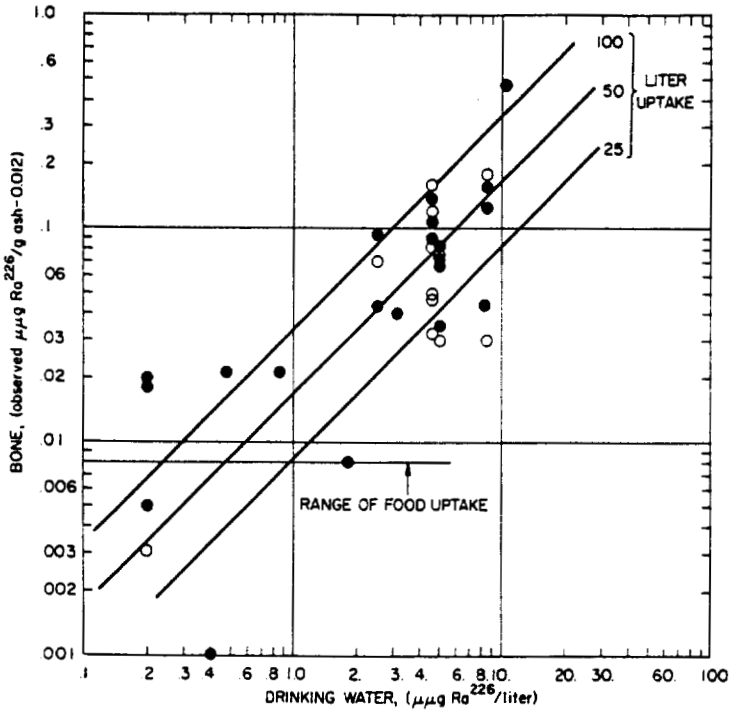

Figure 17

Correlation of Ra^{226} in bone and drinking water. Solid circles represent individuals who have lived all their lives in a single city, with constant use of well water with high radium content. Open circles represent older life-long residents of a single city with exposure to high radium content drinking water for more than the last 30 years but not for their entire lives.

To evaluate the effect of age on whole-body content, the stability of the radium content of the water supply must be evaluated over the lifetime of the subjects. Since teeth have a very much smaller rate of turnover than bone, teeth from individuals of varying ages would give a good indication of the level of radium in the drinking water in past times. The radium content of teeth from life-long residents of the city (No. 9, Table 15) with 4.6 $\mu\mu g$/liter in its drinking water are summarized in Table 16. The average of 0.10 $\mu\mu g/g$ ash is almost identical to the 0.11 $\mu\mu g/g$ ash found for bone from residents from this same city. This may be fortuitous since, as shown in Figure 18, the radium content of both bone and teeth decrease with age.

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Table 16

Ra^{226} in human teeth from city with 4.6 $\mu\mu g/l$. Ra^{226} in drinking water

Sample number	Age	g ash	Ra^{226}, $\mu\mu g/g$ ash ± E*
236	47	2.05	0.046 ± 0.010
68	57	1.35	0.060 ± 0.006
140	54	0.69	0.060 ± 0.012
137	29	0.93	0.069 ± 0.013
139	33	1.64	0.083 ± 0.007
136	35	1.53	0.085 ± 0.010
134	37	1.30	0.112 ± 0.011
70	26	1.52	0.147 ± 0.020
138	30	1.13	0.264 ± 0.015
Average			0.103

*0.9 statistical counting error.

This apparent decrease of radium content with age correlates with known changes in the water source for this city. Prior to 1896, all water came from springs or shallow artesian wells, which are presumed from current measurements of similar sources to be of low radium content, and it was not until 1922 that all of the water came from deep-well sources. Because of the small number of samples and the rather recent changes in the water source, no conclusion can be made about the effect of age on whole-body content. It would, however, appear that teeth are a sensitive indicator of drinking water radium content in the past and can be used to estimate the radium content of populations on which other methods are not feasible.

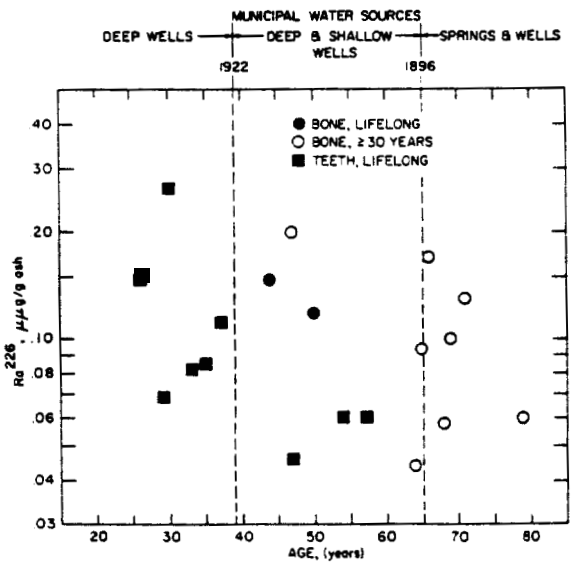

Figure 18

Variation in Ra^{226} content of bone and teeth with age and drinking water source.

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IV. Summary

Measurement of the skeletal and soft tissue contents have been made on normal humans whose primary source of radium was their food. An average whole-body content of 0.36×10^{-10} g is inferred from these measurements. This corresponds to a whole-body uptake equal to 22 times the daily intake. In agreement with other investigators on non-occupationally exposed humans, no correlation with age was noted.

For lifetime uptake, accretion of radium by bone was found to correlate well with that in the drinking water, and the whole-body content for a standard man was found to correspond to that contained in about 50 liters of water. An attempt was made to evaluate the effect of age on whole-body content but because of the small number of samples and recent changes in water source, no conclusion can be made at this time. Teeth appear to be a useful indicator of the radium content of populations on which other methods are not feasible.

I should like to express my appreciation to Asher J. Finkel, M. D., Director of the Health Division, for contacting many of the individuals who have contributed time and samples to this study. I am extremely grateful to all of the dentists, surgeons and pathologists too numerous to mention here for the many teeth and bone samples. Thanks are due Robert W. Anderson for obtaining and verifying residence and water source histories. Acknowledgement is made to Frank H. Ilcewicz, Ronald B. Burke and Janet A. Gondry for the many analyses which were required. Thanks are also due to Dr. Andrew F. Stehney for encouragement and many helpful discussions.

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