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

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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 x 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

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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.

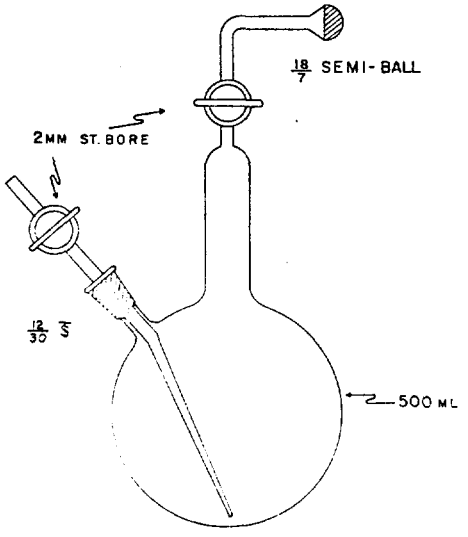

Figure 15

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.

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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.

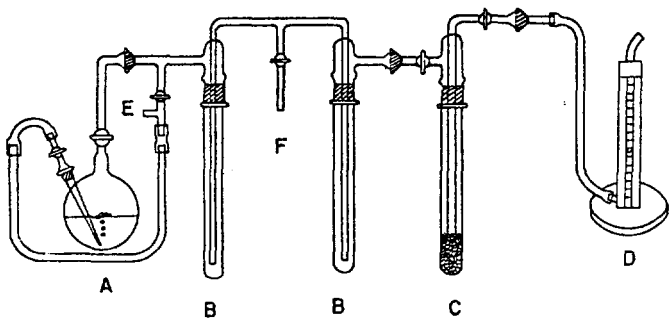

Figure 16

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

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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	-	"	229	vertebra	0.19	0.031 ± 0.037	0.024
	"	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	231	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.

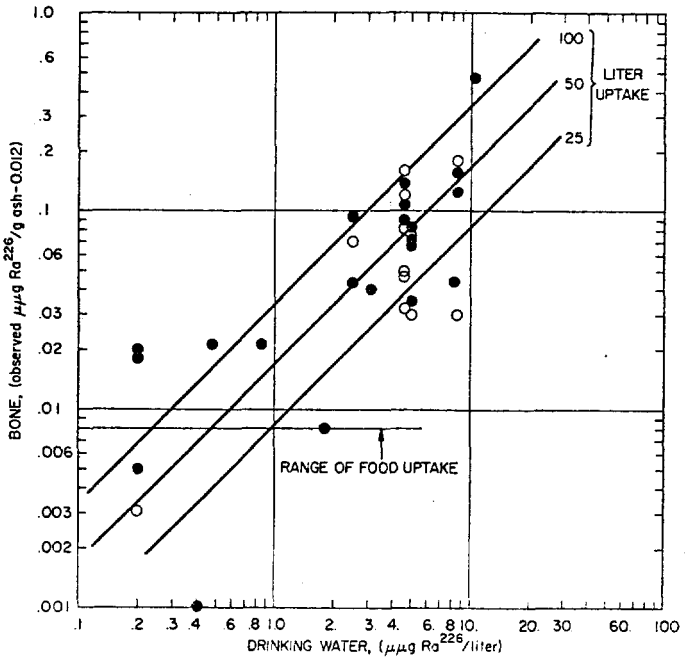

Figure 17

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.

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.

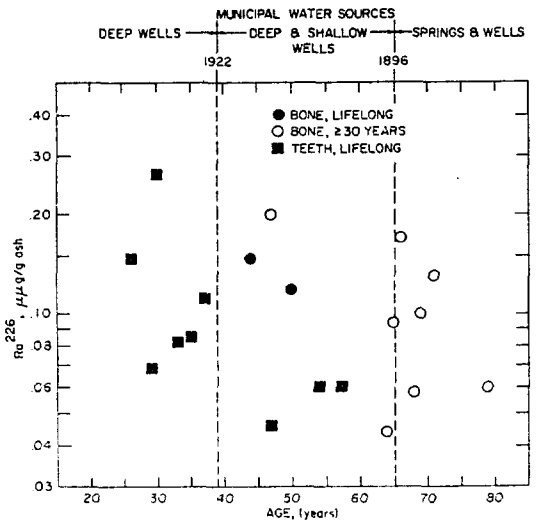

Figure 18

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.

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