THE METABOLIC AND DOSIMETRIC SIGNIFICANCE OF ^{210}Pb TO ^{226}Ra RATIOS IN THE BONE OF RADIUM DIAL PAINTERS*†

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Abstract—The concentration of ^{210}Pb and ^{226}Ra was measured at 89 different sites in the skeletons of two deceased radium dial painters. Using an exponential model for both ^{210}Pb and ^{226}Ra clearance from the skeleton, the data indicate:

1. ^{210}Pb and ^{226}Ra have equal biological half-lives.
2. Variation in the ratios of ^{210}Pb to ^{226}Ra in individual long bones follows a distinct and reproducible pattern. The value of the ratio is lowest in the end pieces of long bones, and highest in the shaft. The ratio is high in rib and low in vertebra. These variations have been interpreted as arising from differences in the ^{222}Rn retention factors at the sites analyzed. These retention factors have been calculated, and vary from 0.20 to 0.57 compared to the skeletal average of 0.33. It is felt that the variations in the ^{222}Rn retention factors reflect fundamental differences in bone structure, namely crystal size, mineral density, and amorphous mineral content.
3. For a ^{226}Ra burden carried 50 years, it is estimated that 5–6% of the average alpha dose delivered to the skeleton results from ^{210}Pb decay.
4. The vertebra would appear to be the bone of choice to be used in estimating total body burden of ^{226}Ra.
5. In a given dial painter, ^{226}Ra concentrations with respect to ash content, varied by a factor of 15 between the highest and lowest values.

THE longest lived daughter of ^{226}Ra is 21.4 year ^{210}Pb. An important intermediate daughter product is the noble gas, 3.83 day ^{222}Rn. In an individual, such as a radium dial painter, who has carried an elevated skeletal burden of ^{226}Ra for many years, the average skeletal ratio of ^{210}Pb to ^{226}Ra is a function of several variables. These include the ^{222}Rn retention factor, the biological half-lives of ^{210}Pb and ^{226}Ra, the physical half-life of ^{210}Pb, and the time elapsed since exposure. These factors which control the average skeletal ratio of ^{210}Pb to ^{226}Ra, likewise control the ratio at specific skeletal sites. However, it does not necessarily follow that specific sites in the skeleton will have the same values for biological parameters that are found for the total skeleton.

In the evaluation of the data to be presented, long time clearance of radium from the skeleton is assumed to be exponential. The bases for this assumption are recent studies which indicate that long term clearance of radium from the skeleton can be described by a single exponential function, and that the average biological half-life of radium is 15–20 years.[1, 2] Schroeder and Balassa have made a compilation of existing data concerning the distribution of stable lead in human tissue, including bone.[3] Analysis of the data for bone, wherein the concentration of lead was measured as a function of age for 79 different individuals, indicates that

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the concentration of lead in bone increases exponentially with age, the half period being approximately 20 years. Therefore, it is also assumed that skeletal elimination of lead can be approximated by an exponential function.

As regards the retention of ^{222}Rn by the skeleton, it has been shown by many workers that the average skeletal retention factor for ^{222}Rn is 0.33,[4] the remainder being expired via the lungs. The rate at which specific skeletal sites lose radon is not known, however, and since the structure of bone is not uniform, it is possible that the ^{222}Rn retention factor is not constant and equal to 0.33 at all skeletal sites, but is dependent upon the locus of ^{226}Ra deposition. If the retention of radon is dependent upon the locus of formation, there are two important consequences. First, the dose delivered by a given amount of deposited ^{226}Ra is a function of the fraction of ^{222}Rn retained. Secondly, it has been postulated by Rowland *et al.*[5] and Mays *et al.*[6] that the fraction of ^{222}Rn retained by bone is controlled by the size of the bone crystals and the mineral density in the region of the ^{226}Ra deposit. Hence, variability of the ^{222}Rn retention factor from site to site within the skeleton, if such is the case, would reflect variability in the fundamental structure of bone.

It was, then, the purpose of the study to be described to measure the ratio of ^{210}Pb to ^{226}Ra at as many skeletal sites as possible in individuals having old, elevated burdens of ^{226}Ra with the expectation that the data would yield information concerning variability within the skeleton of ^{222}Rn retention factors and the relative biological half-lives of ^{210}Pb and ^{226}Ra. This information, in turn, would permit more refined dose calculations to be made, and hopefully lead to qualitative estimates of the variations in the fundamental structure of bone.

BRIEF CASE HISTORIES OF THE INDIVIDUALS STUDIED

The ratio of ^{210}Pb to ^{226}Ra has been measured at 89 different sites in the skeleton of two deceased female radium dial painters. The first case, New Jersey Department of Health No. 5281, was born in 1898. She was continuously exposed as a dial painter from 1914 to 1917.[7] In 1956, her right leg was amputated as a result of an osteogenic sarcoma of the femur. Her body burden of ^{226}Ra, as measured by whole body counting in 1962, was estimated to be 0.54 μCi.[8] She died in 1964 of an acute coronary occlusion. At autopsy, bone specimens were obtained, and thirty-two bone sites taken from eleven different bones, representing about 11% of the total skeleton, were analyzed for ^{210}Pb and ^{226}Ra.

The second case, New Jersey Department of Health No. 5278, was born in 1893. She was continuously exposed as a dial painter in 1917.[7] Her burden of ^{226}Ra was estimated in 1961, by whole body counting, to be 0.046 μCi.[8] She died in 1965 of diffuse metastatic carcinoma. A total of fifty-seven different bone specimens, taken at autopsy from fifteen different bones, were analyzed for ^{210}Pb and ^{226}Ra. Approximately 20% of the skeleton was analyzed.

ANALYTICAL METHODS

All bone samples analyzed were kept frozen and sealed in polyethylene bags prior to analysis.

Each bone section was weighed, ashed for 16 hr at 600°C, and the resulting ash weighed. From these data, the percent ash of each sample was computed. Temperatures in excess of 600°C were avoided less lead be lost by volatilization.

The weighed ash was dissolved in hydrobromic acid, lead carrier added, and ^{210}Pb determined according to the procedure of Petrow and Cover.[9] After the separation of lead, the ash solution was treated with nitric acid to remove bromide ion, the bulk of the free nitric acid removed by evaporation, and radium coprecipitated with lead sulfate after the addition of lead and sulfuric acid. The ^{226}Ra content of the sample was then determined according to the procedure of Petrow *et al.*[10, 11]

ASSUMPTIONS TO BE CONSIDERED

In the introduction, it was pointed out that the ^{222}Rn retention factor and the biological half-lives of ^{210}Pb and ^{226}Ra can affect the value of the ^{210}Pb and ^{226}Ra ratio at any given skeletal site, and in the total skeleton. There are also other factors, including time of bone storage after death and the ^{222}Rn loss rate

during storage, and the ^{210}Pb content of ingested dial paints.

For the first case, 5281, the samples were stored, frozen and sealed in polyethylene for one year between autopsy and analysis. The loss of ^{222}Rn during this year of storage must be considered. Compared to the approximately 67% average ^{222}Rn loss assumed during life, there can be two extremes for the storage period, 100% loss, or zero loss. It has been assumed, for purposes of this study, that ^{222}Rn loss during storage was zero, or very nearly so. This is not a purely arbitrary choice, since the samples were stored, frozen and sealed, and Mays has shown that ^{222}Rn loss from frozen bones is very low.[12] Since the samples were stored for only one year, any error that would arise in the calculations due to a 10% to 20% ^{222}Rn loss during storage would be small. Therefore, for Case 5281, all calculations are based upon a radium burden carried for 49 years to death, during which an average ^{222}Rn loss rate of 67% is assumed. Finally, a zero ^{222}Rn loss rate during the year of storage is assumed.

For Case 5278, the storage period was only 3 months, and hence, any storage error is even less than for Case 5281. The exposure is assumed to have occurred in mid-1917, so 48 years elapsed between exposure and death. All calculations are based on a burden carried for 48 years during which the average ^{222}Rn loss rate was 67%, and 3 months of storage at zero ^{222}Rn loss.

A second factor to be considered is the ^{210}Pb content of the ingested dial paints, for if there were substantial amounts of ^{210}Pb in the paints, then there would be a source of skeletal ^{210}Pb other than ingrowth from ^{226}Ra deposited in the skeleton. The fraction of ingested ^{210}Pb that would be deposited in the skeleton is not known. As will be shown, the data for both cases analyzed indicate that ^{226}Ra and ^{210}Pb formed from ^{226}Ra decay are cleared from the skeleton at equal rates, and this is confirmed by excreta analysis of ^{210}Pb and ^{226}Ra for Case 5281.[13] These data will also be considered later. Therefore, the assumption will be made, in the absence of actual data on the fate of ingested ^{210}Pb, that equal fractions of ingested ^{210}Pb and ^{226}Ra are fixed in the skeleton. The question, then, is what is the likely ^{210}Pb content of a ^{226}Ra preparation used in the period 1914–17, and what effect would this amount of ^{210}Pb have on the results.

The source of ^{210}Pb in the earth is from the decay of ^{238}U. The half-life of ^{238}U is 4.5×10^9 years. The age of the earth is estimated to be about 3.5×10^9 years. That is, about 40% of the uranium present at creation has decayed to ^{206}Pb. Were all the lead formed still present in the mineral, from an original one gram of uranium, there would be remaining 0.6 g of uranium and 0.35 g of lead. Associated with this 0.6 g of uranium would be 0.18 μCi of ^{226}Ra. Hence, in the mineral, one would expect a lead-to-radium mass ratio of 2×10^6. In the analysis of many uranium minerals for lead and uranium, the author has found that the stable lead-to-uranium ratio can vary from about 0.15 to as high as 10. This is the result of many factors, including leaching, recrystallization, and the incorporation of stable lead in the mineral of origin other than uranium decay. However, it is safe to say that the ratio of stable lead to radium is always greater than 10^5. Therefore, in order to prepare ^{226}Ra of high specific activity, as is needed for dial paint formulation, well over 99% of any lead present would have to be separated from the radium. Hence, freshly prepared ^{226}Ra would be very nearly free of ^{210}Pb, since chemical separation processes do not make isotopic distinctions.

The second consideration is how long the radium was stored between purification and use. The period of exposure for the two cases analyzed was 1914–17 and 1917. Radium was discovered in 1898. The maximum age, then, was 16–19 years for the radium used in the paints. In fact, however, during World War I, demand for radium was so great that the material was used almost immediately after preparation,[14] and hence, there was very little time for ^{210}Pb ingrowth. Since ^{210}Pb ingrowth in one year is only 3% of the ^{226}Ra content, and probably less due to ^{222}Rn leakage, it is safe to assume that the ^{210}Pb content of the paints was less than 3% of the ^{226}Ra content, which is negligible.

BASIS FOR THE CALCULATIONS

If one accepts the qualifying assumptions made above concerning sample storage and the

^{210}Pb content of ingested radium paints, there remain two possible causes which can significantly affect the value of the ^{210}Pb to ^{226}Ra ratio at any given skeletal site: the biological half-lives of ^{226}Ra and ^{210}Pb, and the ^{222}Rn retention factor.

The effect of the biological half-lives of ^{210}Pb and ^{226}Ra on the value of the ^{210}Pb to ^{226}Ra ratio at any given skeletal site can be massive. If the relative difference between the biological half-lives of the two radionuclides is large, this alone can influence the value of their ratio to an extent that dwarfs all other factors. If the biological half-life of ^{210}Pb is very long relative to that of ^{226}Ra at a given site, the ratio will be large. If, on the other hand, ^{210}Pb is cleared much more rapidly than ^{226}Ra, the ratio will be small. Therefore, the effect of the ^{210}Pb and ^{226}Ra biological half-lives on the value of the ratio should be considered for three different situations: where the ^{210}Pb biological half-life is greater than, less than, or equal to that of ^{226}Ra.

The Bateman expression for the ratio of ^{210}Pb to ^{226}Ra activity in bone at some time t after the deposition of ^{226}Ra can be expressed as follows:

$$\frac{^{210}\text{Pb}}{^{226}\text{Ra}} = \frac{A^0_{\text{Ra}} \cdot R \cdot \lambda^P_{\text{Pb}}}{\lambda^P_{\text{Pb}} + \lambda^B_{\text{Pb}} - \lambda^P_{\text{Ra}} - \lambda^B_{\text{Ra}}} \cdot \frac{\left[\exp\left[-\left(\lambda^P_{\text{Ra}}+\lambda^B_{\text{Ra}}\right)t\right] - \exp\left[-\left(\lambda^P_{\text{Pb}}+\lambda^B_{\text{Pb}}\right)t\right]\right]}{A^0_{\text{Ra}} \exp\left[-\left(\lambda^P_{\text{Ra}} + \lambda^B_{\text{Ra}}\right)\right]}$$

A^0_{Ra} = initial ^{226}Ra concentration,

t = years elapsed since exposure,

R = ^{222}Rn retention factor,

λ^P_{Pb} = physical decay constant for ^{210}Pb, years $^{-1}$,

λ^P_{Ra} = physical decay constant for ^{226}Ra, years $^{-1}$,

λ^B_{Pb} = biological decay constant for ^{210}Pb, years $^{-1}$,

λ^B_{Ra} = biological decay constant for ^{226}Ra, years $^{-1}$.

Since there is no way of knowing the values of λ^B_{Ra} and λ^B_{Pb} at a given skeletal site, the equation, as written cannot be solved. However for the special case where $\lambda^B_{\text{Pb}} = \lambda^B_{\text{Ra}}$, the equation simplifies to

$$\frac{^{210}\text{Pb}}{^{226}\text{Ra}} = \frac{R \cdot \lambda^P_{\text{Pb}}}{\lambda^P_{\text{Pb}} - \lambda^P_{\text{Ra}}}\left[1 - \exp[-(\lambda^P_{\text{Pb}} - \lambda^P_{\text{Ra}})t]\right]$$

Therefore, when ^{210}Pb and ^{226}Ra have equal biological half-lives, the value of the ratio is independent of their magnitude, regardless of their value. Since the value of R for the total skeleton is known, and is equal to 0.33, one can calculate the value of the ratio when t is known, and when $\lambda^B_{\text{Pb}} = \lambda^B_{\text{Ra}}$. Furthermore, if the average skeletal ratio is greater than that calculated for the special case of equality of ^{210}Pb and ^{226}Ra biological half-lives, it can be said that ^{210}Pb has a longer biological half-life than ^{226}Ra, and conversely, if the average skeletal ratio is less than that calculated for the special case of equality, ^{210}Pb has a shorter biological half-life than ^{226}Ra. Despite the handicaps of a surplus of unknowns, the value of the skeletal ratio permits one to say whether ^{210}Pb is eliminated faster, slower, or at the same rate as ^{226}Ra, to the limits of certainty permissible in our knowledge of R and t.

THE SIGNIFICANCE OF THE MEASURED AVERAGE SKELETAL RATIOS

For Case 5281, with an assumed ^{222}Rn retention factor of 0.33, a 49-year post-exposure history, and one year of bone storage at a zero ^{222}Rn loss rate, the ratio of ^{210}Pb to ^{226}Ra can be calculated for the special case of equal lead

and radium biological half-lives, using the following expression:

$$\frac{^{210}Pb}{^{226}Ra} = \frac{0.33 \times 0.0324}{0.0324 - 0.0004}\left[1 - e^{-0.0320 \times 48}\right] e^{-0.0324 \times 1} + 1 - e^{-0.0324 \times 1} = 0.286$$

which is simply the Bateman expression for the ratio already presented corrected for ^{210}Pb ingrowth and decay during one year of sample storage.

From this it follows that for an average ^{222}Rn retention factor of 0.33, a ratio greater than 0.286 is the result of lead being cleared more slowly than radium, and a ratio less than 0.286 is the result of lead being cleared more rapidly than radium.

In Table 1 are presented the data, for the analysis of 32 bone sites taken at autopsy, from 11 different bones, for Case 5281. The total weight of ash analyzed was 259 g. If we consider her post-amputation ash content to have been 2300 g, it can be seen that 11% of the total skeleton was analyzed. Considering the quantity of ash analyzed and the number of bones and bone sites involved, the total sample is very likely representative, and the average ^{210}Pb and ^{226}Ra concentrations determined are equal, or very nearly equal, to her skeletal concentration of these two radionuclides. This is confirmed by the average ^{226}Ra concentration in the 259 g analyzed, 259 pCi/g ash. Her post-amputation body burden was 0.54 μCi. For an estimated skeletal ash weight of 2300 g remaining after amputation, an average ^{226}Ra concentration of 235 pCi/g ash would be expected. Allowing for the uncertainty in the estimate of her skeletal ash content, the agreement is good and the sample, therefore, is very likely representative.

The average skeletal ^{210}Pb to ^{226}Ra ratio, as determined radiochemically, is 0.266. This is very nearly equal to the value of 0.286 to be expected when the ^{222}Rn retention factor is 0.33 and ^{210}Pb and ^{226}Ra have equal biological half-lives. The data suggest that on the average, lead and radium are cleared from the skeleton at equal or very nearly equal rates. The fact that ^{210}Pb and ^{226}Ra are cleared at equal rates is confirmed by excreta analyses performed at the Argonne National Laboratory on a one-week feces and urine collection obtained from Case 5281 before her death.[13] The ratio of ^{210}Pb to ^{226}Ra found in the excreta was 0.255. The ratio in the skeleton found in this study, corrected for the ratio change during one year storage, is 0.243, nearly perfect agreement with the Argonne excreta data.

The data for the second case, 5278, are given in Table 2. Based upon a single whole body count, this individual had a ^{226}Ra burden of 0.046 μCi. A total of 57 different bone sites, taken at autopsy from 15 different bones, were analyzed. The total ash weight analyzed was 607 g, or 22% of an assumed total skeletal ash content of 2800 g. For a body burden of 0.046 μCi, an average ^{226}Ra concentration of 16.4 pCi/g ash would be expected. The average ^{226}Ra concentration found was 15.4 pCi/g ash, indicating that the sample was representative.

For a ^{222}Rn retention factor of 0.33, the ^{210}Pb to ^{226}Ra biological half-lives can be calculated, as before,

$$\frac{^{210}Pb}{^{226}Ra} = \frac{0.33 \times 0.0324}{0.0324 - 0.0004}\left[1 - e^{-0.0320 \times 48} \times e^{-0.0324 \times 0.25}\right] + 1 - e^{-0.0324 \times 0.25} = 0.266$$

The average skeletal ratio, as determined from the analysis of bone, is 0.284, once again very close to that predicted for equal ^{210}Pb and ^{226}Ra biological half-lives.

It would appear, then, that the average skeletal ratio of ^{210}Pb to ^{226}Ra, as determined radiochemically, is consistent with equal biological half-lives for ^{210}Pb and ^{226}Ra, provided one assumes that the clearance of both nuclides is exponential, and the ^{222}Rn retention factor is 0.33. Equal clearance rates of ^{210}Pb and ^{226}Ra from the skeleton for Case 5281 are confirmed by the very close agreement between the ^{210}Pb to ^{226}Ra ratios in the skeleton and in the excreta. Furthermore, Holtzman[15] has calculated a biological half-life of stable lead in the skeleton of 17.4 years, and the data in ref. 3 indicate a half-life of 20 years, both in close agreement with the average biological half-life of radium determined in refs. 1, 2, about 15-20 years.

Table 1. *Analysis of Case* 5281 *Bone for* ^{210}Pb *and* ^{226}Ra

Sample	Bone	g ash	% ash	^{210}Pb pCi/g ash	^{226}Ra pCi/g ash	$^{210}Pb/^{226}Ra$
2786	L. Tibia	12.15	30.0	29.4 ± 1.47	118 ± 5.90	0.249 ± 0.017
2787	,,	6.61	38.5	23.3 ± 1.16	88.0 ± 4.40	0.266 ± 0.019
2794	,,	10.20	24.2	148 ± 7.40	691 ± 34.6	0.214 ± 0.015
2795	,,	4.27	14.6	93.4 ± 4.67	415 ± 20.8	0.225 ± 0.016
* Weighted Average		33.23		72.6 ± 2.42	326 ± 10.9	0.222 ± 0.010
2847	L. Ribs, 6 & 7	2.61	14.8	92.1 ± 4.60	279 ± 14.0	0.330 ± 0.023
2849	,,	3.20	16.5	82.4 ± 4.12	236 ± 11.8	0.349 ± 0.024
2850	,,	2.60	18.9	68.4 ± 3.42	196 ± 9.8	0.349 ± 0.024
2851	,,	2.48	13.6	116 ± 5.80	331 ± 16.6	0.349 ± 0.024
2852	,,	2.48	10.8	102 ± 5.10	309 ± 15.5	0.330 ± 0.023
Weighted Average		15.37		92.9 ± 1.82	268 ± 5.25	0.346 ± 0.010
2716	L. Humerus	3.96	25.9	58.8 ± 2.94	210 ± 10.5	0.280 ± 0.020
2720	,,	3.95	26.3	126 ± 6.30	424 ± 21.2	0.295 ± 0.021
2725	,,	12.50	33.9	26.1 ± 1.30	86.7 ± 4.34	0.301 ± 0.021
2730	,,	8.14	42.4	21.0 ± 1.05	69.9 ± 3.49	0.301 ± 0.021
Weighted Average		28.55		43.4 ± 1.16	146 ± 3.90	0.297 ± 0.011
2697	L. Radius	3.21	33.8	26.1 ± 1.30	84.0 ± 4.20	0.310 ± 0.022
2700	,,	1.20	15.4	148 ± 7.40	628 ± 31.4	0.235 ± 0.016
2704	,,	4.20	21.0	233 ± 11.7	1030 ± 51.5	0.227 ± 0.016
Weighted Average		8.61		144 ± 4.66	621 ± 20.1	0.232 ± 0.011
2811	Sternum	8.87	8.70	70.7 ± 3.53	186 ± 9.40	0.380 ± 0.027
2813	,,	3.88	7.09	69.6 ± 3.48	220 ± 11.0	0.316 ± 0.022
Weighted Average		12.75		70.4 ± 2.68	196 ± 7.47	0.359 ± 0.019
2752	Mandible	3.77	13.6	139 ± 6.95	467 ± 23.4	0.298 ± 0.021
2764	,,	2.11	13.5	151 ± 7.55	521 ± 26.1	0.291 ± 0.020
Weighted Average		5.88		143 ± 5.20	486 ± 18.1	0.294 ± 0.015
2766	Vertebra, Thor.	14.9	14.4	64.5 ± 3.22	291 ± 14.6	0.222 ± 0.016
2863	Vertebra, Cerv.	25.0	15.4	74.4 ± 3.72	296 ± 14.8	0.252 ± 0.018
Weighted Average		39.9		71.0 ± 2.62	295 ± 10.9	0.240 ± 0.012
2842	L. Fibula	3.99	17.1	110 ± 5.50	476 ± 23.8	0.231 ± 0.016
2844	,,	3.51	16.1	19.8 ± 0.99	67.0 ± 3.35	0.294 ± 0.021
Weighted Average		7.50		68.0 ± 2.96	284 ± 12.4	0.240 ± 0.015
2816	L. Ulna	8.23	28.6	92.1 ± 4.60	358 ± 18.0	0.256 ± 0.018
2822	,,	1.96	14.4	88.8 ± 4.44	411 ± 20.6	0.215 ± 0.015
Weighted Average		10.19		91.0 ± 3.81	368 ± 15.4	0.247 ± 0.015

Table 1. *Analysis of Case* 5281 *Bone for* ^{210}Pb *and* ^{226}Ra*—Cont.*

Sample	Bone	g ash	% ash	^{210}Pb pCi/g ash	^{226}Ra pCi/g ash	$^{210}Pb/^{226}Ra$
3088	Calvaria	34.85	50.0	26.1 ± 1.30	108 ± 5.40	0.242 ± 0.017
2871	L. Femur	20.96	31.8	23.5 ± 1.18	66.6 ± 3.33	0.354 ± 0.025
2877	,,	11.35	44.2	55.5 ± 2.78	197 ± 9.85	0.283 ± 0.020
2884	,,	9.16	28.6	77.6 ± 3.88	264 ± 13.2	0.294 ± 0.021
2891	,,	8.21	30.9	152 ± 7.60	542 ± 27.1	0.280 ± 0.020
2896	,,	12.52	16.7	82.6 ± 4.13	322 ± 16.1	0.256 ± 0.018
Weighted Average		62.20		66.1 ± 1.56	234 ± 5.52	0.282 ± 0.009
Total Skeletal Average		259		68.6 ± 0.75	259 ± 2.82	0.266 ± 0.004

* Weighted average $= \dfrac{\Sigma \text{sample weight} \times \text{nuclide concentration}}{\Sigma \text{sample weight}}$.

The assumption concerning the ^{222}Rn retention factor as being 0.33 can be questioned, in that various laboratories use values ranging from 0.3 to 0.4.[16] However, Vennart *et al.*,[4] in a survey of the published work of many laboratories making *in-vivo* ^{226}Ra measurements, concludes that the ^{222}Rn retention factor is equal to 0.33. At any rate, were the retention factor either 0.3 or 0.4, the conclusion concerning the equal biological half-lives of ^{226}Ra and ^{210}Pb would not be greatly changed. Considering the data for Case 5281, if the ^{222}Rn retention factor was taken as 0.4 and the biological half-life of ^{226}Ra as 16 years, the calculated biological half-life for ^{210}Pb is 13.3 years. On the other hand, were the retention factor 0.3, the biological half-life of ^{210}Pb calculates to be 17.7 years. The range of the biological half-life of ^{210}Pb is 13.3 to 17.7 years relative to ^{226}Ra taken as 16 years. This is not an enormous range. However, there are other factors, both chemical and physical, that suggest that equality of biological half-lives for ^{226}Ra and ^{210}Pb formed from ^{226}Ra decay is not only reasonable, but likely. When ^{210}Pb is formed, it is formed very close to the parent ^{226}Ra deposit, probably in the same crystal or an adjacent crystal. Both ions are bivalent. Both ions have similar ionic radii. The chemistry of lead and radium is similar in a large number of reactions. Therefore, while the biological half-life of ^{210}Pb could be somewhat smaller or larger than that of ^{226}Ra, it is the author's belief that they are equal based upon the arguments presented.

It should be pointed out that the proposed equality is only for ^{210}Pb formed from ^{226}Ra decay since in this case a constraint is imposed upon the system in that the birth place of ^{210}Pb is controlled by the locus of radium deposition. It by no means proves that if lead and radium were deposited independently in the skeleton, the equality would hold. However, data already cited concerning the biological half-life of stable lead[3,15] indicate that independently deposited lead has a half-life similar to that of ^{210}Pb formed from ^{226}Ra decay.

The conclusion that ^{210}Pb and ^{226}Ra have equal biological half-lives is in disagreement with that of Holtzman.[15] Analyzing about 10 pieces of bone taken from five different individuals, he concluded that ^{210}Pb is cleared more slowly than ^{226}Ra. However, this work suffers from several weaknesses. First, the samples were stored for as much as 10 years before being analyzed, thereby magnifying possible errors arising from the unknown ^{222}Rn retention factor during storage. Only a few pieces of bone were analyzed for each case. Finally, it was assumed that the ^{222}Rn retention factor is invariant at every site in the skeleton, an assumption which,

Table 2. Analysis of Case 5278 Bone for ^{210}Pb and ^{226}Ra

Sample	Bone	g ash	% ash	^{210}Pb pCi/g ash	^{226}Ra pCi/g ash	$^{210}Pb/^{226}Ra$
3719	L. Tibia	19.74	17.9	12.6 ± 0.63	51.4 ± 2.57	0.246 ± 0.017
3721	,,	9.68	23.2	6.55 ± 0.33	23.0 ± 1.15	0.284 ± 0.020
3723	,,	9.65	32.6	4.24 ± 0.21	14.0 ± 0.70	0.302 ± 0.021
3737	,,	18.79	48.1	1.81 ± 0.09	5.40 ± 0.27	0.335 ± 0.023
3731	,,	25.03	29.6	6.31 ± 0.32	21.7 ± 1.08	0.294 ± 0.020
Weighted Average		82.89		6.60 ± 0.18	24.3 ± 0.68	0.272 ± 0.011
3760	L. Rib, 7	3.36	13.9	3.60 ± 0.18	9.91 ± 0.50	0.362 ± 0.025
3762	,,	2.16	11.7	3.87 ± 0.19	9.70 ± 0.48	0.400 ± 0.028
Weighted Average		5.52		3.71 ± 0.13	9.84 ± 0.35	0.378 ± 0.019
3588	L. Femur	14.54	43.5	2.93 ± 0.15	8.40 ± 0.42	0.350 ± 0.025
3592	,,	16.34	47.2	2.46 ± 0.12	6.45 ± 0.32	0.380 ± 0.027
3598	,,	11.22	43.0	2.80 ± 0.14	8.56 ± 0.43	0.327 ± 0.023
3604	,,	6.83	33.4	5.50 ± 0.28	21.2 ± 1.06	0.260 ± 0.018
3605	,,	19.77	22.1	9.42 ± 0.47	37.1 ± 1.85	0.255 ± 0.018
3611	,,	13.73	34.7	2.11 ± 0.10	5.42 ± 0.27	0.389 ± 0.027
3612	,,	13.43	26.4	4.73 ± 0.24	17.6 ± 0.88	0.267 ± 0.019
3614	,,	19.95	21.0	4.64 ± 0.23	17.3 ± 0.86	0.267 ± 0.019
Weighted Average		115.81		4.52 ± 0.10	16.1 ± 0.36	0.282 ± 0.009
3750	L. Metatarsal	2.48	19.6	4.90 ± 0.24	16.3 ± 0.81	0.301 ± 0.021
3752	,,	3.21	24.8	10.3 ± 0.51	37.1 ± 1.85	0.278 ± 0.020
Weighted Average		5.69		7.95 ± 0.31	28.1 ± 1.09	0.284 ± 0.016
3636	L. Clavicle	4.02	19.1	3.30 ± 0.16	9.34 ± 0.46	0.352 ± 0.025
3641	,,	3.75	20.8	2.37 ± 0.11	6.56 ± 0.32	0.362 ± 0.025
Weighted Average		7.77		2.84 ± 0.10	8.00 ± 0.29	0.356 ± 0.018
3772	L. Rib	1.31	11.2	3.76 ± 0.18	7.70 ± 0.38	0.480 ± 0.034
3774	,,	4.63	14.7	3.49 ± 0.17	9.00 ± 0.45	0.386 ± 0.027
Weighted Average		5.94		3.54 ± 0.14	8.72 ± 0.35	0.406 ± 0.023
3732	L. Fibula	4.93	22.8	8.71 ± 0.43	46.7 ± 2.33	0.186 ± 0.013
3736	,,	7.23	37.4	2.72 ± 0.13	8.88 ± 0.44	0.306 ± 0.021
3738	,,	6.80	19.8	7.52 ± 0.35	43.2 ± 2.16	0.174 ± 0.012
Weighted Average		18.96		5.98 ± 0.29	31.1 ± 1.52	0.192 ± 0.013
3713	Vertebra	23.06	11.9	2.99 ± 0.15	14.7 ± 0.74	0.206 ± 0.014
3708	Calvaria	53.79	53.2	2.75 ± 0.13	11.7 ± 0.58	0.235 ± 0.016
3712	,,	64.59	52.6	2.02 ± 0.10	6.57 ± 0.33	0.308 ± 0.022
Weighted Average		118.38		2.36 ± 0.08	9.00 ± 0.32	0.262 ± 0.013

Table 2. Analysis of Case 5278 Bone for ^{210}Pb and ^{226}Ra—Cont.

Sample	Bone	g ash	% ash	^{210}Pb pCi/g ash	^{226}Ra pCi/g ash	$^{210}Pb/^{226}Ra$
3753	R. Rib, 6	1.00	5.8	4.66 ± 0.23	12.7 ± 0.64	0.367 ± 0.026
3757	,,	1.69	14.8	4.60 ± 0.23	12.1 ± 0.60	0.381 ± 0.027
3759	,,	2.07	22.0	2.34 ± 0.11	5.41 ± 0.27	0.430 ± 0.030
Weighted Average		4.76		3.63 ± 0.11	9.32 ± 0.28	0.389 ± 0.016
3627	L. Ulna	10.80	25.5	4.86 ± 0.24	13.9 ± 0.70	0.350 ± 0.025
3633	,,	4.60	44.7	3.69 ± 0.18	11.5 ± 0.58	0.321 ± 0.023
3635	,,	4.35	25.6	4.92 ± 0.24	14.1 ± 0.70	0.350 ± 0.025
Weighted Average		19.75		4.60 ± 0.15	13.4 ± 0.44	0.344 ± 0.016
3671	L. Humerus	3.47	32.9	2.41 ± 0.12	6.04 ± 0.30	0.399 ± 0.028
3673	,,	4.33	36.6	2.94 ± 0.14	8.00 ± 0.40	0.368 ± 0.026
3675	,,	5.18	46.3	2.74 ± 0.13	7.70 ± 0.38	0.355 ± 0.025
3677	,,	6.70	44.4	2.56 ± 0.12	7.16 ± 0.36	0.359 ± 0.025
3681	,,	2.77	37.9	2.74 ± 0.13	7.70 ± 0.38	0.355 ± 0.025
3684	,,	3.16	25.7	2.78 ± 0.13	7.80 ± 0.39	0.356 ± 0.025
3687	,,	4.52	22.4	9.05 ± 0.45	31.2 ± 1.56	0.290 ± 0.020
3690	,,	3.68	32.2	4.58 ± 0.27	18.0 ± 0.90	0.310 ± 0.022
3694	,,	4.90	12.0	4.91 ± 0.24	14.7 ± 0.74	0.336 ± 0.024
Weighted Average		44.79		3.74 ± 0.07	11.2 ± 0.20	0.333 ± 0.008
3765	R. Rib, 7	1.71	7.6	3.58 ± 0.17	10.9 ± 0.54	0.327 ± 0.023
3769	,,	2.83	20.2	2.37 ± 0.11	7.65 ± 0.38	0.310 ± 0.022
Weighted Average		4.54		2.84 ± 0.10	8.86 ± 0.31	0.320 ± 0.016
3615	L. Radius	2.83	24.0	6.89 ± 0.34	29.4 ± 1.47	0.234 ± 0.016
3618	,,	6.52	43.3	2.24 ± 0.11	6.90 ± 0.34	0.325 ± 0.023
3621	,,	4.29	47.5	2.50 ± 0.12	7.02 ± 0.35	0.357 ± 0.025
3626	,,	3.53	28.9	6.41 ± 0.32	23.6 ± 1.18	0.271 ± 0.019
Weighted Average		17.17		3.93 ± 0.10	14.1 ± 0.36	0.278 ± 0.010
3643	R. Femur	15.87	45.2	3.04 ± 0.15	8.31 ± 0.42	0.366 ± 0.026
3645	,,	10.94	47.7	2.75 ± 0.14	7.56 ± 0.38	0.364 ± 0.026
3649	,,	12.62	48.5	2.96 ± 0.15	7.44 ± 0.37	0.399 ± 0.028
3654	,,	10.55	43.0	3.90 ± 0.20	12.0 ± 0.60	0.323 ± 0.023
3658	,,	7.75	36.5	4.92 ± 0.25	14.7 ± 0.74	0.335 ± 0.023
3659	,,	18.19	24.6	9.40 ± 0.47	35.9 ± 1.80	0.262 ± 0.018
3661	,,	28.76	23.9	7.95 ± 0.40	26.8 ± 1.34	0.296 ± 0.021
3665	,,	24.24	26.5	3.00 ± 0.15	7.74 ± 0.39	0.387 ± 0.027
3669	,,	8.00	23.8	7.37 ± 0.37	24.2 ± 1.21	0.304 ± 0.021
Weighted Average		140.92		5.16 ± 0.11	16.6 ± 0.36	0.311 ± 0.010
Total Skeletal Average		606.95		4.38 ± 0.46	15.4 ± 0.16	0.284 ± 0.004

as will be shown later, is highly questionable.

There is, therefore, very little reason to doubt that ^{210}Pb and ^{226}Ra are eliminated from the skeleton at very nearly equal rates, and that the two rates, if not exactly equal, are sufficiently similar to justify the use of the simplified Bateman expression that results when the two rates are equal. This has several important consequences. First, since long-term measurements of ^{226}Ra elimination rates can be made by whole body counting, the ^{226}Ra data can be used to estimate stable lead elimination rates as well, provided radiogenic and indigenous lead are eliminated similarly. Secondly, for individuals whose radium exposure occurred at some unknown time, excreta analyses for ^{210}Pb and ^{226}Ra can be used to establish the ratio of ^{210}Pb to ^{226}Ra in the skeleton, and hence, the date of exposure can be calculated, assuming a ^{222}Rn retention factor of 0.33. Third, the whole body skeletal dose due to ^{210}Pb, ^{210}Bi, and ^{210}Po can be more accurately calculated.

CONSIDERATION OF THE VARIABILITY IN INDIVIDUAL RATIOS

Far more difficult to interpret than the value of the average skeletal ratio of ^{210}Pb to ^{226}Ra are the variations in the ratio values found for the different skeletal sites. For Case 5281, the ratios varied from 0.214 to 0.380, and for Case 5278, from 0.174 to 0.480. The variability in the ratios is not random in nature. As will be noted from the data in Tables 1 and 2, there is a distinct pattern in long bone, with regard to ratio and radium concentration. In general, for a given long bone, the greater the ^{226}Ra concentration, the lower the ratio. The striking correlation between the ^{226}Ra concentration in a bone and the ratio value can be seen in Table 3. Equally striking is the fact that each bone is an independent entity with respect to this correlation. For neither case does the correlation exist for all the long bones taken as a single group. However, as can be seen from Case 5278, where both femurs were analyzed, they are very similar. In general, however, ratio values for tibia, fibula and radius tend to be considerably lower than for humerus and femur. There is also a striking correlation between the ratio value and the locus of radium deposition. For a given long bone, high radium concentration and low ratios are found in the end pieces of the bone, with the shaft having high ratios and low radium concentrations. This is particularly true, for both cases, in the fibula, tibia, and radius, where the end pieces have ratios lower than the skeletal average, while the shaft values are equal to or higher than the skeletal average. Humerus and femur, on the other hand, while showing the same pattern as regards ratio, radium concentration, and locus of deposition, have, for both cases, nearly every ratio higher than the average skeletal ratio.

All ratios found in rib, again for both cases, are higher than the average skeletal ratio, whereas vertebra have ratios much lower than the average. Therefore, bone characteristics are, without question, exerting an influence on the value of the ratio.

There are two mechanisms which could result in the observed ratio variations, each quite different:

1. While the average skeletal elimination of ^{210}Pb and ^{226}Ra occurs with equal biological half-lives, non-equality of these rates at discrete skeletal sites might account for the observed differences in ratio values.
2. Variation in the ^{222}Rn retention factors from site to site in the skeleton is a possible cause.

It is difficult to believe that differences in the clearance rates of lead and radium at discrete skeletal sites is the decisive factor controlling ratio variation. If one assumes the ^{226}Ra biological half-life to be constant throughout the skeleton and equal to 16 years, and that the ^{222}Rn retention factor is constant and equal to 0.33, then to account for the ratio variations noted, the biological half-life of ^{210}Pb would have to vary from 5 years to 50 years, depending upon the site analyzed. This wide range of ^{210}Pb biological half-lives, from one-third to three times that of ^{226}Ra, is difficult to accept, especially when one considers the added constraint that despite the wide range of possible differences, the average biological half-lives of both radionuclides are equal. For this reason, differences in metabolic clearance rates are not considered to be a likely cause of the observed differences in ratio.

Table 3. A Comparison of ^{210}Pb to ^{226}Ra Ratios and ^{226}Ra Concentrations in Long Bone

Bone and Case	^{226}Ra pCi/g ash	^{210}Pb/^{226}Ra
L. Tibia, 5278	51.4	0.246
,,	23.0	0.284
,,	21.7	0.294
,,	14.0	0.302
,,	5.40	0.335
L. Tibia, 5281	691	0.214
,,	415	0.225
,,	118	0.249
,,	88.0	0.266
L. Radius, 5281	1030	0.227
,,	628	0.235
,,	84	0.310
L. Radius, 5278	29.4	0.234
,,	23.6	0.271
,,	7.02	0.357
,,	6.90	0.325
L. Fibula, 5281	476	0.231
,,	67.0	0.294
L. Fibula, 5278	46.7	0.186
,,	43.7	0.174
,,	8.88	0.306
L. Humerus, 5281	424	0.295
,,	210	0.280
,,	86.7	0.301
,,	69.9	0.301
L. Humerus, 5278	31.2	0.290
,,	18.0	0.310
,,	14.7	0.336
,,	8.00	0.368
,,	7.80	0.356
,,	7.70	0.355
,,	7.70	0.355
,,	7.16	0.359
,,	6.04	0.399
L. Ulna, 5281	411	0.215
,,	358	0.256
L. Ulna, 5278	14.1	0.350
,,	13.9	0.350
,,	11.5	0.321
L. Femur, 5281	542	0.280
,,	322	0.256
,,	264	0.294
,,	197	0.283
,,	66.6	0.354
L. Femur, 5278	37.1	0.255
,,	21.2	0.260
,,	17.6	0.267
L. Femur, 5278—*cont.*	17.3	0.267
,,	8.56	0.327
,,	8.40	0.350
,,	6.45	0.380
,,	5.42	0.389
R. Femur, 5278	35.9	0.262
,,	26.8	0.296
,,	24.2	0.304
,,	14.7	0.335
,,	12.0	0.323
,,	8.31	0.366
,,	7.74	0.387
,,	7.56	0.364
,,	7.44	0.399

Next, we come to the question of whether variation in ^{222}Rn retention factors can be regarded as the cause of ratio variations. Of all the possible causes of ratio variation, differences in ^{222}Rn retention factors have, by far, the greatest dosimetric significance, and it is important to establish whether ^{222}Rn retention is variable. First, however, it would be helpful to consider the presently accepted theory of ^{222}Rn loss from the skeleton.

Rowland *et al.*[5] have postulated a ^{222}Rn recoil hypothesis to explain the loss of this radionuclide from the skeleton. Mays *et al.*[6] have proposed a similar mechanism. The essence of their postulate is that if a recoiling ^{222}Rn atom, which at birth has 86 keV of kinetic energy, and a mean range in bone of about 200–300 Å,[5] comes to rest within a bone crystal, it will very likely decay within that crystal, since diffusion rates of gases in a crystal are slow, relative to the half-life of ^{222}Rn, 3.83 days. If the recoiling atom comes to rest within the organic bone matrix, where there is a constant flow of fluids, then ^{222}Rn will escape due to the ease of gaseous diffusion in liquids as compared to crystalline solids. The escape path is diffusion into the circulation, followed by exhalation.

Whether the recoiling ^{222}Rn atom comes to rest within a crystal or the organic matrix is controlled by several variables. The size of the crystal is, of course, very important. The larger the crystal, the higher the probability that the ^{222}Rn atom will lose its kinetic energy before escaping from the crystal. It is currently believed that bone crystals are tablet shaped, being 200–300 Å long and 50 Å thick.[17] However, it is believed that in long bones, crystals are larger in the shaft than in the ends.[17]

Mineral density in the region of a recoiling ^{222}Rn atom can affect the ^{222}Rn retention factor. Mineral density can be defined as the fraction of the total density in the recoil volume that is mineral. The greater the mineral density, the greater should be the ^{222}Rn retention factor, since mineral density simply reflects the probability of a recoiling ^{222}Rn atom coming to rest in the crystalline mineral portion of bone. Ash content of a bone is an index, although not an exact measure of mineral density. The greater the ash content, the greater the mineral density. In long bones, the shafts have a considerably higher ash content than the end pieces, and hence, one would expect a higher mineral density in the shafts and a greater retention factor.

A third factor, and one very difficult to evaluate, is the recent discovery[18] that a portion of the mineral content of bone is amorphous. Gaseous diffusion rates in an amorphous solid are much greater than in a crystal. Many early workers noted that if radium is co-precipitated with barium sulfate, ^{222}Rn retention is nearly quantitative, while, on the other hand, amorphous solids such as ferric hydroxide or silica gel lose radon almost quantitatively. Commercial sources of ^{220}Rn (thoron), which consist of ^{228}Th co-precipitated with ferric hydroxide, are available. These release a large portion of ^{220}Rn, despite the fact that the half-life of ^{220}Rn is only 55 sec. Therefore, the greater the fraction of mineral bone that is amorphous, the greater should be the ^{222}Rn loss rate. The work cited[18] indicates that for long bone, the amorphous mineral fraction is lower in the shaft than in the ends.

In long bones, crystal size is greatest in the shaft, the ash content, and hence, the mineral density is highest in the shaft, and the shaft contains relatively less amorphous mineral than the end pieces. All three factors would favor higher ^{222}Rn retention in the shaft relative to the bone ends, and, therefore, higher ^{210}Pb to ^{226}Ra ratios in the shafts. This is precisely the effect noted for long bones, higher ratios in the shaft relative to the ends. Therefore, what we know about crystal size, mineral density, and amorphous mineral content of long bones supports the hypothesis that variable ^{222}Rn retention factors are the cause of the observed differences in the ratio of ^{210}Pb to ^{226}Ra.

As was mentioned earlier, ratios in rib are high, and ratios in vertebra are low. Both vertebra and rib have a low ash content. Therefore, if ^{222}Rn retention controls ratio values, ash content is not the dominant factor. Crystal size and/or the amorphous mineral content must override mineral density. It is for such reasons that it is so important to establish the true cause of ratio variations. If ratio variations are due to differences in ^{222}Rn retention factors, then we have a potential tool for studying bone structure.

As a result of the fact that ^{210}Pb and ^{226}Ra have equal biological half-lives, it is a simple matter to calculate what the ^{222}Rn retention factor was at a site where the ^{210}Pb to ^{226}Ra ratio is known. For both Case 5281 and 5278, were there no ^{222}Rn loss, the ^{210}Pb to ^{226}Ra ratio would be 0.850, as calculated from the Bateman expression already presented. Therefore, the retention factor at any given site is the quotient of the determined ratio and 0.850. For the range of ratios found for the two cases, 0.170 to 0.480, the retention factors must vary from 20% to 57%. The dosimetric significance of this will be considered later.

It can be argued that translocation of ^{210}Pb and ^{226}Ra could produce the variations in the ratio observed. As lead and radium are cleared from bone and enter the blood stream, a portion of both radionuclides is resorbed by the skeleton. This process of elimination followed by resorption can, of course, alter the value of the ratio at any given skeletal site. However, in order for this mechanism of translocation to account for the observed ratio variation, the process would have to occur with the same pattern as has been observed for the ratios themselves. That is, ^{210}Pb would have to be preferentially

resorbed at certain skeletal sites, namely the shafts of long bones and in rib, at the expense of resorption in vertebra and the ends of long bones, where low ratios have been observed. In order to determine whether such a preferential deposition of lead occurs within the skeleton, the distribution of stable lead in the skeleton has been studied. The rationale behind this approach is that resorbed ^{210}Pb should deposit in the skeleton in an identical manner to stable lead entering the blood stream via the gut or the lungs.

Bone samples from an individual who died at the age of 62 were analyzed for stable lead. The data are presented in Table 4.

The pattern of stable lead deposition is not consistent with an hypothesis of preferential deposition of ^{210}Pb via resorption. In fact, if anything, the opposite pattern is indicated.

Table 4. Analysis of Human Bone for Stable Lead

Sample	μg/g ash
Radius, Shaft	87
Tibia, Shaft Piece 1	97
Tibia, Shaft Piece 2	92
Femur, Shaft	77
Femur, End Piece 1	106
Femur, End Piece 2	131
Femur, End Piece 3	154
Vertebra	132
Rib	120
Calvarium	88

For the femur, the lead concentration is lowest in the shaft, and highest in the end pieces, precisely opposite to the pattern noted for the ratios in the femur of the two dial painters. The vertebra has a high lead concentration, whereas the ratio in vertebra was found to be low for both dial painters. For the dial painters, the highest ratios were found in rib. This is not the case as regards the concentration of stable lead. From these data, it is inferred that translocation is not the cause of the observed variations in ratio. In the writer's opinion, differences in ^{222}Rn retention factors are the dominant cause of the variations in ratio.

ESTIMATION OF BODY BURDEN FROM THE ANALYSIS OF A SINGLE BONE

As the data in Tables 1 and 2 indicate, there is a wide range of ^{226}Ra concentrations in the skeleton of an exposed individual. For Case 5281, the range of ^{226}Ra concentrations found was 67–1030 pCi/g of ash, with the average being 259 pCi/g of ash. If one attempted to measure the body burden of an individual by analysis of a single piece of bone, it would be difficult to do so with any degree of accuracy whatsoever, but for one exception, the vertebra. For Case 5281, the two pieces of vertebra analyzed had ^{226}Ra concentrations of 291 and 296 pCi/g of ash, as compared to the average value of 259 pCi/g of ash. For Case 5278, the single piece of vertebra analyzed gave a result of 14.7 pCi/g of ash as compared to the average of 15.4 pCi/g of ash found as the average. Based upon a sampling far too small to be conclusive, it would appear that vertebra would be the most reliable index of body burden.

DOSIMETRIC CONSIDERATIONS

Using an exponential model for ^{210}Pb and ^{226}Ra clearance from the skeleton, with both radionuclides having equal bilogical half-lives and an average skeletal ^{222}Rn retention factor of 0.33, it has been possible to predict the actual average ^{210}Pb to ^{226}Ra ratio in the skeleton of two radium dial painters. This suggests that the model is valid and, therefore, applicable to the calculation of the average skeletal alpha dose. Only the alpha dose will be considered since there is little reason to doubt that bone is the critical organ, and the bulk of the dose to bone is from alpha radiation.

For Case 5281, the terminal burden of ^{226}Ra is known, 0.54 μCi. Were it not for the amputation of her right leg, her terminal burden of ^{226}Ra would have been approximately 0.66 μCi. In calculating dose, it is this value that will be used, since she carried her leg most of her life. Four whole body measurements, taken over a period of 3 years, indicate that her radium biological half-life was 20 years.[1] The author, however, through measurement of ^{228}Th to ^{228}Ra ratios in her skeleton, calculated her radium biological half-life to be 10.4 years.[19] Both values will be used in calculating the average skeletal alpha dose. It is assumed that the

skeletal mass was 10,000 grams, and the average ^{222}Rn retention factor equal to 0.33. It is also assumed that all ^{210}Po formed *in vivo* decayed *in vivo*. The time elapsed since exposure was 49 years.

Taking 10.4 years as the biological half-life of ^{226}Ra (and of ^{210}Po), her initial burden of ^{226}Ra was 17.1 μCi. Up to the time of death, approximately 16.4 μCi of ^{226}Ra were removed from the skeleton. Of this total, 0.105 μCi of ^{226}Ra actually decayed in her skeleton. The dose delivered by the decay of this much ^{226}Ra and one-third that amount of ^{222}Rn, ^{218}Po, and ^{214}Po is 5160 rad. The dose due to ^{210}Po was calculated as being derived from the total number of ^{210}Pb atoms that decayed *in vivo*. This calculated to be 2.93×10^{13} atoms, and the associated dose, 258 rad. Hence, the total dose was 5418 rad, of which 4.8% results from ^{210}Po.

Assuming a biological half-life of 20 years for radium, the initial burden of ^{226}Ra was 3.97 μCi. A total of 3.31 μCi was removed from the skeleton, of which 0.039 μCi ^{226}Ra disappeared via decay. The resulting dose from ^{226}Ra, ^{222}Rn, ^{218}Po, and ^{214}Po was 1920 rad. The ^{210}Po dose was 120 rad, and 5.9% of the total dose resulted from ^{210}Po decay.

It can be seen that the fraction of the total dose derived from ^{210}Po decay is rather insensitive with regard to the magnitude of the biological half-lives of ^{226}Ra and ^{210}Pb, and it is safe to say that for long periods of exposure, the ^{210}Po dose is approximately 5–6% of the total alpha dose.

Calculations based upon data presented elsewhere,[19] indicate that the total dose arising from the ^{228}Ra decay chain was approximately 5200 rad, assuming a 10.4 year biological half-life for radium. Therefore, it would appear that her total radium dose was 10,000 rad, or less, depending upon the biological half-life of radium used in the calculation. Since it is unlikely that the radium biological half-life is less than 10.4 years, the figure of 10,000 rad is the upper dose limit. It might seem inconsistent, at first, that smaller doses are associated with longer biological half-lives of radium. This arises from the fact that the initial radium burden is calculated from a known terminal burden, and becomes smaller as the radium biological half-life increases.

Considering the individual sections of the skeleton, the average ^{222}Rn factors calculated for these sections, the ^{226}Ra concentration in each section, and a biological half-life of radium of 10.4 years, the highest dose calculated due to the ^{226}Ra decay chain is 13,000 rad, and the lowest dose, 840 rad, as compared to the skeletal average of 5418 rad.

For Case 5278, whole body counting indicated her biological half-life for radium to be about 10 years. Using this figure, her average skeletal dose from ^{226}Ra was calculated to be 390 rad, with a range of 110 rad to 840 rad for the individual bone sections.

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