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RADIUM TOXICITY STUDIES

R. E. Rowland, J. Jowsey and J. H. Marshall

I. Quantitative Observations on Ra^{226} in Bone

Autoradiographic studies have shown that the pattern of deposition of radium in bone is similar to that of calcium. Studies of both human and animal bone show radium to be taken up in areas of appositional growth in the process of mineralization of the newly formed osteoid. We define these regions of high concentration of radium as "hot spots", while the more uniform deposition which appears throughout the bone tissues is called the diffuse reaction. These terms, hot spot and diffuse reaction, as we use them, are descriptive terms referring to the appearance of an autoradiograph and are not associated with any one mechanism.

Autoradiographs of Ra^{226}-burdened bone are easily evaluated quantitatively. For this we employ an Eastman Kodak NTA plate with a 25μ thick emulsion. The number of alpha tracks observed in this emulsion after a suitable exposure to a bone section depends upon the following:

1. The quantity of Ra^{226} per unit volume of bone.
2. The fraction of the radon produced which escapes from the bone.
3. The range of the alpha particles in bone. We have demonstrated that approximately 70% of the radon produced in a bone section embedded in methyl methacrylate escapes, the same fraction that escapes from the bone *in vivo*.[1]

Thus, for every radium atom which emits an alpha particle, there will be four alphas emitted by its daughters, of which only 30% will be emitted in bone. The average range of these alpha particles in bone (ρ = 2 g/cm^3) is 30μ. Hence, we write the number of radium disintegrations per unit time per unit volume as

$$\frac{4 \times \text{No. of tracks}}{2.2\ \alpha\text{'s per Ra disintegration}} \times \frac{1}{\text{Time}} \times \frac{1}{\text{Area} \times 30\mu.}$$

From a volume of bone 30μ thick one quarter of the alphas produced will reach the surface and produce a track within the emulsion. Hence above we multiply the number of tracks by four. Since there are 2.2 alpha particles emitted in bone per radium disintegration (1 + 0.3 x 4), we divide the number of alphas produced by 2.2 and obtain the number of radium disintegrations per unit time per unit volume.

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To obtain this activity in terms of microcuries, we simply divide by 3.7×10^{10} disintegrations/sec:

$$\mu c \text{ Ra}^{226}/\text{mm}^3 = 1.9 \times 10^{-8} \frac{T}{A \times t}$$

where T = the observed number of tracks in area A.
A = the area in mm^2
t = the time of the exposure in days

The quantitative study of Ra^{226} depositions in bone yields information on the following:

1. The dose in microscopic regions of bone.
2. The relationship between the diffuse and the hot spot activities in bone. Dose distributions and the correlation with damage to bone structure will be reported at a later date.

After a single injection of radium, the specific activity of the blood drops rapidly, and the mineral formed during this period will have a constantly changing specific activity. A narrow band of bone a few microns wide will be labeled with the isotope, but this band is too narrow to allow evaluation of an average specific activity of the mineral. However, when the specific activity of the blood is elevated for a considerable period of time as in the Elgin State Hospital patients,[2,3] or as may be assumed to have occurred in the dial painters, then the activity in regions of appositional growth covers a cross-sectional area of dimensions sufficiently large to allow the evaluation of the specific activity by means of an autoradiographic measurement of activity and a microradiographic measurement of density. In these cases we can define a hotspot to diffuse ratio with some precision. The ratio must be clearly understood to refer to the ratio of the specific activities, but not to the rates of uptake of the radioactive label in the two regions.

The diffuse uptake of radioactivity we picture as taking place as long as the specific activity of the blood is elevated. The appositional growth process, instead, labels a closing osteon with the specific activity of the blood, and thus, to a first approximation, the activity found is independent of the length of time the blood level was elevated. Of course, a region labeled as a result of appositional growth can later be diffusely labeled, but this is a second order effect.

In Table 25 are shown the quantitative values of hot spot and diffuse activities obtained to date for a number of cases. Here we have used the concept of "uniform label," which we define as the body burden divided by the bone volume. The bone volume is calculated assuming a wet density of 2 g/cm^3, and assuming the bone weight is 10% of the body weight. This

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"uniform label" would be expected to describe the distribution occurring in an animal which had always a constant level of a radioactive tracer in the blood. Under hot spots we have listed the value of the highest specific activity found for a large haversian system. Under diffuse we have placed a range of values which were obtained from counting tracks over areas ten times as large as a typical hot spot. Finally, we have calculated the ratios hotspot to diffuse activity, hot spot to "uniform label," and diffuse to "uniform label,"

Table 25

Summary of the autoradiographic observations

Case	Radium acquired	Body burden at death, μc	Body weight, kg	Uniform label, $\mu c/mm^3$	Measured activity		Calculated ratios		
					Hot spot, $\mu c/mm^3$	Diffuse, $\mu c/mm^3$	Hot spot / Diffuse	Hot spot / Uniform label	Diffuse / Uniform label
R	Elgin patient 41 weekly inj. at age 32 yrs. Died 56 yrs.	3.6	55	1.3×10^{-6}	2.9×10^{-5}	$.67-.98 \times 10^{-6}$	30-43	22	0.5-0.8
I	Elgin patient 18 weekly inj. at age 27 yrs. Died at 53 yrs.	0.7	55 assumed	0.25×10^{-6}	1.3×10^{-5}	0.2×10^{-6}	65	52	0.8
103	Medically, I-V inj. at age ~35 yrs. Died ~64 yrs.	6.8	57.5	2.4×10^{-6}	2.6×10^{-5}	0.7×10^{-6}	37	11	0.3
118	Drank Ra water at age 48 yrs. Died at 84 yrs.	10.5	52.	4.0×10^{-6}	6.7×10^{-5}	$2.2-3.4 \times 10^{-6}$	20-30	17	0.55-0.85
302	Dial Painter Painted at age 15 for 2 mo. Died at 47 yrs.	3.8	68	1.1×10^{-6}	2.5×10^{-6}	$1.0-2.0 \times 10^{-6}$	12.5-25	23	0.9-1.8
	Dial Painter, Painted at age 21 yrs for 1 yr. Died at age 54 yrs.	2.7	55 assumed	0.98×10^{-6}	3.4×10^{-5}	$0.14-1.6 \times 10^{-6}$	21-240	35	0.14-1.6
Dog R	Age ~1-2 yrs. Single I.V. Inj. Sacrificed at 1 mo.	16	9.1	35×10^{-6}		$22-32 \times 10^{-6}$			0.6-0.9

These values were all obtained from cortical bone sections, usually from the femur, but in some instances from the tibia. Note that in these long bones the activity associated with the diffuse label is of the order of magnitude of one-half the "uniform label" defined above as shown by the last column in this table.

In Table 26 we list the activities found in various bones of Elgin patient I. In this patient the radium burden had been carried for 26 years. While no great importance can be placed on samples ranging from 100 to 300 g, it is still apparent that most of the activity remaining is in the long bones. These gross samples were evaluated by gamma-ray spectroscopy by Dr. C. E. Miller.

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Table 26

Bone radium content, Elgin patient I

Bone	Sample, g	Average Activity $\mu c/g$	Activity Uniform label
Femur	285	3.5×10^{-4}	2.4
Vertebrae and rib	74	0.91×10^{-4}	0.62

II. The Loss of Ra^{226} from Human Bone

The Elgin State Hospital patients form an extremely important and valuable group for study, for they were given known quantities of Ra^{226} at weekly intervals for varying lengths of time. Thus we know the total input and can assume, without great error, the specific activity of the blood during the period over which the I.V. injections were given.

In Fig. 32 is shown a blood specific activity curve for a human given weekly injections of $10\mu c$ of Ra^{226}. This was drawn by transforming the blood curve for Sr^{85} in humans(4) into a curve for Ra^{226}, the transformation used being that required to transform an observed Sr^{90} curve for beagles(5) into an observed Ra^{226} curve for beagles.(6) Admittedly, this is not the most satisfactory manner of obtaining a blood curve, but we feel that at least it gives the proper order of magnitude to be expected.

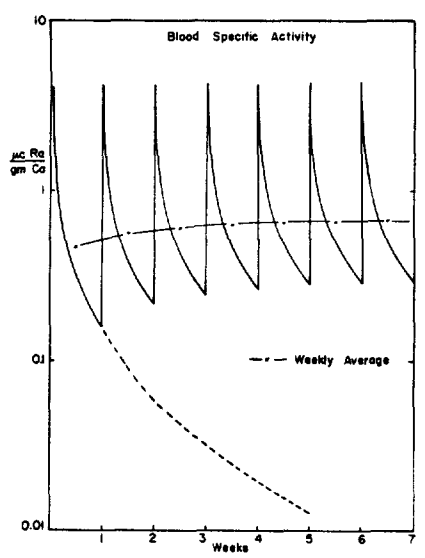

Fig. 32 Postulated blood specific activity in a human receiving weekly injections of 10 c Ra^{226}. The highest value plotted for each week is the value occurring at one hour.

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Also indicated in Fig. 32 is the average value of the blood specific activity over each weekly interval. Note that this curve seems to approach a constant value after several weeks. This average value is of considerable importance, for if there is no discrimination against the radium from blood to bone, then regions of appositional growth should be built at this specific activity during the period over which the injections were given.

In Table 25, given in part I of this report, are tabulated bone specific activities for Elgin cases R and I. The values for R are quite good, having been obtained from studies of many different bone sections, while those from I must be considered tentative at present as they have been made from only a very few observations. From Fig. 32 we would expect hot spots to show a specific activity of the order of 0.7 μc Ra^{226}/g Ca.

Transforming this value from units of μc Ra^{226}/g Ca to units of μc Ra^{226}/mm^3 of bone, using a microradiographically determined value of 1.3 g of hydroxyapatite per cm^3 of bone (for human bone)(7) where hydroxyapatite is 40% calcium by weight, we obtain 36×10^{-5} $\mu c/mm^3$. This is the specific activity we would expect a forming osteon to incorporate if it were built during the 41 or 18 week interval in patients R or I. However, from Table 25, we find that this value is 12 and 27 times greater than the highest activities found in these two cases. If these systems are thought to have been formed after the injection period, then examination of the blood activity curve indicates they were formed within a few weeks after the injections stopped, and one must postulate that all original systems formed during the period of the injections have been resorbed. This consideration convinces us that these are osteons formed during the injection period, not afterward.

Thus it is apparent that either radium is not built into forming osteons at the specific activity of the blood, or that a loss of activity from these regions has occurred without resorption taking place. To describe this removal of a label without removal of the total calcium within a system we use the term diminution. We are tacitly assuming that the results we obtain have not been greatly modified by radiation damage.

The magnitude of the diffuse label in these two Elgin cases is quite informative. While the period over which the specific activity of the blood was elevated differs by a factor of two, the actual retained radium levels after 26 years differ by a factor of five, and the observed diffuse levels also differ by a factor of five. The dependence of the magnitude of the diffuse label upon the body burden is quite evident in the last column of Table 25, where the ratio of the diffuse level to the uniform label is tabulated.

Since roughly the same diffuse to uniform label relationship exists one month after injection in the dog (see Table 25) and 26 or more years later in the human, it is tempting and not unreasonable to assume that the

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magnitude of the diffuse radium label in the bone has decreased with time, apparently at the same rate as the activity in the regions of appositional growth.

While we must emphasize that these results are tentative, it is apparent that they indicate resorption per se may play a relatively minor role in the long-term removal of radium from cortical bone. It is obvious that the mechanisms herein discussed are of decisive importance in the description of radiation dosimetry in bone following intake of bone-seeking radioelements. Before entering into this aspect of our investigation, however, we feel that it is necessary first to extend our observations to suitable human material in order to assess the extent to which our preliminary impressions apply.

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Th^{228}, Ra^{228}, AND Ra^{226} IN NORMAL BONE AND IN BONE SPECIMENS FROM RADIUM PATIENTS

D. E. Wallace and A. F. Stehney

I. Normal Bone

Table 27 summarizes data obtained in the course of testing the dibutyl phosphate (DBP) method for measurement of Th^{228} in bone.[1] Groups of bone samples from the first cattle were analyzed at intervals of about 6 months. The Th^{228} content increased with time, indicating that appreciable amounts of Ra^{228} were present and that Th^{228} was growing into the sample according to the genetic relationship known to exist for these nuclides:

$$Ra^{228} \xrightarrow[T\ 1/2=6.7\ yr]{\beta^-} Ac^{228} \xrightarrow[T\ 1/2=6.13\ hr]{\beta^-} Th^{228} \xrightarrow[T\ 1/2=1.90\ yr]{\alpha} Ra^{224} \xrightarrow[T\ 1/2=3.64\ d]{\alpha} \text{others}$$

Since the history of intake of Ra^{228} and Th^{228} was not known for these samples, it was not possible to calculate the Ra^{228} content by application of equations of radioactive growth. However, extraction of Th^{228} by DBP leaves Ra^{228} in the aqueous solution, and a subsequent extraction of Th^{228} which has been allowed to grow in can now be used to calculate the amount of Ra^{228} present. This was done for the group of samples from which Th^{228} was extracted in July and August, 1956; a second extraction in June, 1957 provided a value of 0.57 $\mu\mu c$ Ra^{228}/g of ashed bone calculated to Aug. 1, 1956. Comparison of this value with the series of Th^{228} values indicates that most of the Th^{228} was present because of growth from Ra^{228} instead of independent deposition in the bone.

Table 27

Th^{228}, Ra^{228}, and Ra^{226} in normal bone

Sample	Bone specimen	Date of analysis	No. of samples	$\mu\mu c$ Th^{228}/g[a]	$\mu\mu c$ Ra^{228}/g[a]	$\mu\mu g$ Ra^{226}/g[a][b]
Cattle - 1	Tibia	July-Aug., 1956	4	0.25±0.01[c]	0.57±0.02	
		Nov.-Dec., 1956	3	0.30±0.01[c]		
		Apr.-May, 1957	5	0.36±0.01[c]		0.66±0.03
Cattle - 2	Tibia	Oct., 1957	4	0.11±0.01[c]		0.22±0.02
Normal Human - 1	Cortical Tibia	Dec. 4, 1956	1	0.016±0.004		$0.07^{+0.01}_{-0.06}$
Normal Human - 2	Cortical Tibia or Femur	Apr. 3, 1957	1	0.007±0.004		0.016±0.006

[a] Radioactivity per g of bone ashed at 620°C.

[b] Analyses for Ra^{226} were performed by radon emanation techniques (Reference 9). The large uncertainty indicated for low values of Ra^{226} was caused by an uncertainty of of 0.02 $\mu\mu g$ in the blank correction.

[c] The mean deviation from the mean value for replicate samples was well within the estimated error of 5 to 10% for a single measurement.

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The concentration of Ra^{226} in these samples was only slightly greater than that of Ra^{228}. It is likely that only 3 or 4 years elapsed from birth of the cattle to time of analysis, so decay of 6.7-yr Ra^{228} should not greatly distort the implied intake of approximately equal amounts of these two isotopes by the animal from its environment. The uniformity of values for different samples implies that intake of radium was continuous and, therefore, from natural sources, but the concentrations of radioactivity are considerably higher than those found for normal human bone (see below). An average value of 0.03 $\mu\mu g$ Ra^{226}/g ashed bone would be expected from the natural body burden of about 10^{-10} g Ra^{226} for humans.

Another sample of bone was obtained from Wisconsin cattle whose history was known. Ra^{228} values have not yet been obtained, but comparison of the first measurements of Th^{228} with Ra^{226} values indicates again that approximately equal amounts of the isotopes of radium are present (Table 27). On the other hand, the concentrations of radioactivity are only one-third of those found for the first cattle although much higher than those for humans. It appears that a ruminant diet may be considerably higher in radium than that of humans. The first of the cattle may have come from a region having an unusually high Ra to Ca ratio in the environment; the sample was purchased locally but the origin of the animal is unknown.

The bone samples from the two normal humans listed in Table 27 were analyzed primarily to provide an estimate of Th^{228} normally present in human bone for comparison with figures from radium patients. The results given are tentative because of the low concentrations and the small amount of sample available. However, no detectable radioactivity (<0.005 $\mu\mu c$ Th^{228}) was found in reagent blanks carried through the analysis. Because the samples represent accumulation of radioactivity over several decades, one may assume that the Ra^{228} is in equilibrium with Th^{228}. Thus it appears from these analyses that Ra^{228} may be present in amounts equal to or greater than Ra^{226} if one includes corrections for decay of Ra^{228}.

One conclusion which may be drawn from the data of Table 27 is that estimates of the natural radioactivity of humans must include Ra^{228} which has been largely neglected in the past. One reason for this neglect is that very little is known about the rate of intake of Ra^{228}. However, Ra^{228} in amounts comparable to or greater than those of Ra^{226} has been reported for numerous radioactive hot spring waters.(2) In addition, preliminary work on the water supplies of certain communities near Chicago has shown that Ra^{228} is present in concentrations approximately equal to those of Ra^{226} (3-8 $\mu\mu g$ Ra^{226}/l). Previously, water from these sources was found to contain Ra^{224} and Ra^{226} in equal amounts, but no Th^{228} was found.(3)

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II. Bone Specimens from Radium Patients

It is now widely recognized that, in addition to Ra^{226}, sufficient Ra^{228} may have been present in the radium preparations taken by some patients to contribute appreciably to the radiation dosage to the bone.(4) Thus, measurement of the Ra^{228} content of specimens of bone from radium patients is of importance in determining whether correlation of bone damage with the Ra^{226} burden alone is valid. Since no means of directly measuring Ra^{228} in bone with sufficient sensitivity is available as yet, the Th^{228} content is being used as a measure of Ra^{228} in the present work. Assuming radioactive equilibrium to exist, the amount of Ra^{228} at the time of radium intake can then be calculated and compared to the Ra^{226} content of the same sample of bone.

Table 28 summarizes data on bone specimens from six radium patients. The code numbers in Column 1 are those used in the report by Looney, et al.,(5) which describes the case histories of these radium patients. Under the heading of Th^{228}, values are given for the amount of Th^{228} per g of bone ashed at 620° C as of the date given. In the last column, the ratio of Ra^{228} to Ra^{226} at the time of intake is expressed in per cent values for each bone specimen. Ra^{228} values were obtained by extrapolating to the approximate time of dosage, using calculations of radioactive growth of Th^{228} and the assumption that direct deposition of Th^{228} need not be considered. These figures were not corrected for natural amounts of Ra^{228}, deposited during the entire life of the subject, so the extrapolation may provide values that are much too high for those cases in which the concentration of Th^{228} was very small. It should be noted that considerable variation in the concentration of Ra^{226} was found for bone specimens from each patient (as previously reported by Norris and co-workers(6) for Case No. 112) and that the concentration of Ra^{228} varied similarly so as to provide ratios for each patient which were constant within reasonable limits of error.

The amount of Th^{228} present (Col. 8, under Th^{228}) was sufficiently high in only three cases to be considered positive identification of intake of Ra^{228} with the Ra^{226} dose. The highest ratio of Ra^{228} to Ra^{226} was 0.02 at the time of intake for a patient (designated by * in Table 28 because no literature reference for this patient is available as yet) who painted radium dials in the 1920's. Patient 112, who had the next highest ratio, took radium orally for therapeutic purposes in 1928. A bone specimen from this patient was reported by R. D. Evans to contain a Th^{228} to Ra^{226} ratio of 0.0024 ± 0.0005 several years ago;(7) allowing for radioactive decay, this value is in reasonable agreement with our Th^{228} to Ra^{226} ratio of 0.001 in 1957. The calculated ratio of Ra^{228} to Ra^{226} at the time of ingestion is 0.01. The third patient, number 313, was a dial painter who seems to have ingested very little Ra^{228}.

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Table 28

Th^{228}, Ra^{228}, and Ra^{226} in specimens of bone from radium patients

Patient	Body burden, $\mu g\ Ra^{226}$	Intake route[a]	Approx. yr. of intake, t_1	Bone specimen	Weight of ashed bone, g	Th^{228}		$\mu\mu g\ Ra^{226}/g$[b]	% Ra^{228} at t_1 relative to Ra^{226}
						Date	$\mu\mu c/g$		
112	1.3	Oral-Med.	1928	Humerus	2.99	7-12-56	0.16 ± 0.01	180	1.2 ±0.1
				Humerus	1.88	8-15-56	0.12 ± 0.01	160	1.0 ±0.1
				Humerus	3.13	7-10-57	0.53 ± 0.03	550	1.4 ±0.1
313	1.3	Oral-Dial	1924	Femur	1.36	4-26-56	0.02 ± 0.01	520	0.08±0.04
				Femur	1.36	5- 4-56	0.06 ± 0.01	730	0.15±0.04
				Femur	1.44	10-26-56	0.02 ± 0.01	280	0.14±0.04
				Vertebra	1.22	7- 9-57	0.07 ± 0.01	1420	0.11±0.02
*	1.3(?)	Oral-Dial	1924	Femur	2.42	7- 9-57	0.54 ± 0.03	560	2.1 ±0.1
R	3.1	Intravenous	1931	Tibia	1.67	5- 7-56	< 0.006	290	< 0.02
				Tibia	2.1	5- 7-56	< 0.005	190	< 0.03
				Tibia	4.70	10-12-56	0.005±0.002	430	(0.01)
				Femur	5.95	7-10-57	0.013±0.002	1220	(0.01)
				Femur	4.94	7-10-57	0.004±0.002	330	(0.01)
103	5.3	Intravenous	1925	Femur	4.68	10-12-56	0.021±0.004	1240	0.03±0.01
				Femur	5.93	7-10-57	0.020±0.005	1090	0.04±0.01
118	2 to 10	Oral-Med.	1918	Vertebra	1.04	4-26-56	<0.01	3220	<0.013
				Vertebra	1.36	5- 4-56	0.04 ± 0.01	5430	0.03±0.01

[a] "Oral-Med." denotes radium water taken as a medicine; "Oral-Dial" refers to radium dial painter.

[b] Errors in measurement of Ra^{226} are estimated to be less than 5 per cent.

* No literature reference available for this patient.

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Patient R is one of the series of patients at Elgin State Hospital who received intravenous injections of radium. Solutions of radium believed to be of the same composition as given to those patients have been reported to contain no more than 0.05 per cent Ra^{228} relative to Ra^{226}, corresponding to a maximum of 0.66 per cent at time of injection.(8) As seen in Table 28, our values for Th^{228} in bone specimen from Patient R do not exceed the values found in "normal" bone, and an extrapolation to time of injection yields a Ra^{228} to Ra^{226} ratio of only 10^{-4}, even without attempting to correct for the natural Th^{228} content of human bone.

Patients 103 and 118 received radium as a therapeutic measure, Number 103 by intravenous injection and the other orally. In both cases, the Ra^{228} content seems to have been negligible.

It is evident that the total radiation dosage from Ra^{228} was minor compared to that of Ra^{226} for each case listed in Table 28. However, the presence of Ra^{228} may be relatively more hazardous if its 1.9-yr Th^{228} descendant migrates to a different portion of the bone--such as the organic matrix.

Specimens from patients 115, 313, R and 118 were extracted with hot ethylenediamine (EDA) as a means of separating the organic constituents. No detectable Th^{228} was found in the organic portion. However, these results are inconclusive in that the absolute amount of Th^{228} in the whole bone was small and several years had elapsed since the bone material had died.

On the other hand, positive results might be expected for samples from the first cattle listed in Table 27 from which a large supply of fresh bone was available. The comparison of inorganic and organic portions given in Table 29 shows that less than 5 per cent of the Th^{228} in each sample was recovered in the organic fraction. Probably an even smaller percentage was actually extracted by the EDA, because contamination with inorganic matter is indicated by the large weight of residue in the organic fraction for the three samples in which Th^{228} was found. Such contamination was also demonstrated by the presence of Ra^{226} in the organic residues.

The results in Table 29 do not fully resolve the question as to migration of Th^{228} in bone because there was no assurance that all the organic matter was extracted by EDA. In particular, the tissue closest to the bone crystals may have been inaccessible to the EDA. That the amount of organic material left behind may be small is shown by the fact that bone transplants are made successfully after removal of proteins with EDA.

We wish to thank Dr. Robert J. Hasterlik of the Argonne Cancer Research Hospital and Drs. Asher Finkel and William P. Norris for bone specimens from radium patients. We are grateful to Commander G. W. Hyatt of the National Naval Medical Center, Bethesda, Maryland, for providing a sample of normal bone from the Tissue Bank Department, and to Captain F. Losee, also of Bethesda, for valuable advice on treatment of bone with ethylenediamine.

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Table 29

Comparison of Th^{228} in inorganic and organic portions of bone from Cattle-1

Sample	Fraction[a]	Weight of ashed residue,[b] g	Th^{228}	
			Date	Total Amount, $\mu\mu c$
1	Inorganic	4.13	8-15-56	1.02 ± 0.06
1	Organic	0.055	8-17-56	< 0.01
2	Inorganic	4.73	11-23-56	1.54 ± 0.09
2	Organic	0.38	11-27-56	0.03 ± 0.02
3	Inorganic	4.46	11-23-56	1.33 ± 0.08
3	Organic	0.56	11-27-56	0.03 ± 0.01
4	Inorganic	5.26	5-23-57	1.92 ± 0.12
4	Organic	0.31	5-23-57	0.10 ± 0.02

[a]Analyses were also made on the filter paper thimble which contained the bone sample during extraction with EDA; no Th^{228} was found.

[b]Weights of residues from the organic fraction include material leached from the glassware as well as portions of the inorganic fraction of bone carried over mechanically.

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THE HUMAN SPECTROMETER

C. E. Miller

I. Introduction

In the period since the last report[1] a second iron-shielded facility has been constructed. The reduction in the gamma-ray background of an 8 by 4 inch NaI(Tl) crystal as the detector was surrounded with various thicknesses of Fe is reported. Extensive data is given for 0, 3/4, 1½, 3, 5½ and 8 inches of Fe. After the background with the steel present was obtained, the detector was surrounded with 1/8 inch of lead to determine the effects of lining the room with sheet lead. Summation of the data clearly indicates that little is gained by building Fe shields thicker than 5 inches until radioactively cleaner photomultipliers become available.

The radioactive body content of numerous subjects has been measured during the past 6 months. A 2-year follow-up study was made of an Am^{241} subject. Four subjects who had inhaled radioactive material were brought to Argonne National Laboratory from other AEC installations. The isotopes were identified, the amount determined and their location ascertained.

A search for former dial painters in the vicinity of Chicago has yielded the identity and location of many subjects containing various amounts of Ra^{226}, with prospects of eventually locating several hundred. Thus far 42 individuals have been measured prior to clinical studies now under way. Because of the low quantity in some of them, it has been necessary to evaluate the uncertainty in a radium measurement due to fluctuations in the subject's Cs^{137} and I^{131} arising from fallout and in K^{40} due to the subject's musculature. A lung-equivalent phantom which may be used to calibrate a detector for inhalation accidents is discussed.

II. Gamma Ray Background of Iron Room II

The complete success of this Division's human spectrometer has led to emulation of the facilities by scientific groups all over the world. Judging from requests for engineering drawings and visits, at least 25 projects, modeled directly on the Argonne type, are contemplated, and other groups are modifying the design for special purposes. A few groups are investigating other shielding materials with the hope of economizing on initial construction costs. Although the effect of steel shielding on the counting rate of NaI(Tl) crystals has been reported several times by our group,[2] it seems advisable to continue to report pertinent information as it accumulates.

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The construction of this Division's second Iron Room provided the opportunity to re-examine the attenuation of the background scintillation spectrum with a large 8 by 4 inch NaI(Tl) crystal as the walls of the room were made progressively thicker. The data tabulated in this report should prove valuable in the construction and design of other low-level facilities since background attenuation of bulky shieldings can be checked during the course of their construction and the data compared to the curves published in this report. Excessive deviation will warrant investigation and may lead to the recognition of contamination problems as they occur. The data in this report will also allow the calculation of the sensitivity and of the accuracy possible with a low-level room of given wall thickness. Others interested in particular types of measurement can determine beforehand the feasibility of an Iron Room for their particular purpose.

Comparison with Other Types of Shielding

All data were taken with the same NaI(Tl) crystal (8 inches in diameter, 4 inches thick) used in Iron Room I. The walls, floor and ceiling of the second room were built up uniformly to each scheduled interval so that all surfaces were of equal thickness while the background attenuation was being determined. Moreover, assessment was made of the shielding effect of 1/8-inch sheet lead placed immediately around the crystal at various wall thicknesses.

The scintillation spectrum of this large crystal in Iron Room II is plotted in Fig. 38 and recorded in Table 39 by thirty-one 50-kev wide bands. A wider segment of the spectrum was also obtained using bands 100 kev wide covering the energy spectrum from 50 kev to 3.150 Mev; during this series of tests the sheet lead around the crystal was not used since 1/8 inch of lead would not substantially affect it at higher energies. These data are given in Table 40.

After the walls had been built up to their full 8-inch thickness, the inside surfaces of the room were covered with 1/8-inch thick sheet lead. The low-energy background pulses were reduced but not to the degree expected from the results obtained with Iron Room I (see below). The background with the crystal surrounded with sheet lead was higher than without the additional lead on the walls, thus demonstrating that this particular lead was contaminated.

To demonstrate the pattern of attenuation, the total background c/m in the neighborhood of the K^{40} peak (1.325 - 1.575 Mev band) were determined for each thickness and plotted as the solid line in Fig. 39. This curve is apparently composed of two components, one of which may be considered as due to natural activity originating entirely outside of the walls, and this fraction should decrease as the thickness of the wall is increased. The second component is due to the activity in the detector,

Table 39

Gamma-ray background reduction

Energy band, kev	0" Fe	3/4" Fe	$1\frac{1}{2}$" Fe	$1\frac{1}{2}$" Fe +Pb	3" Fe	3" Fe +Pb	$5\frac{1}{2}$" Fe	$5\frac{1}{2}$" Fe +Pb	8" Fe	8" Fe +Pb
	Counts per minute									
25-75	3707	507	256	152	142	85.6	88	61.7	76.1	56.5
75-125	5693	1231	566	197	261	99.2	142	60.9	120	54.6
125-175	4768	1622	816	216	315	106	168	62.0	141	56.0
175-225	3665	1445	768	226	280	107	151	63.7	125	56.5
225-275	2674	1141	624	226	224	105	119	61.4	96.2	53.6
275-325	2018	905	498	227	185	102	96.9	56.5	80.3	49.6
325-375	1543	728	407	216	154	97.1	81.8	54.2	66.2	46.9
375-425	1168	589	345	203	138	92.9	69.1	51.7	60.1	45.9
425-475	1033	526	313	205	136	94.7	70.6	53.2	59.9	47.2
475-525	936	476	290	192	125	88.5	69.1	52.5	59.7	43.9
525-575	894	413	243	167	94.4	68.1	49.9	39.9	42.0	36.7
575-625	814	362	210	151	83.3	65.5	43.7	36.2	36.5	32.3
625-675	602	293	179	131	72.2	58.5	37.2	32.9	31.0	28.6
675-725	526	263	157	121	64.6	53.6	34.5	30.0	27.1	26.3
725-775	485	243	145	112	60.6	49.7	31.5	29.0	26.4	24.0
	(30,527)	(10,745)	(5,816)	(2,743)	(2,334)	(1,273)	(1,251)	(746)	(1,047)	(659)
775-825	467	217	132	105	55.6	45.3	27.5	25.8	24.7	23.4
825-875	436	205	122	95.6	49.8	41.7	25.7	23.2	20.3	19.3
875-925	416	194	112	86.8	44.5	37.9	21.9	20.6	19.0	18.3
925-975	387	177	103	80.0	40.7	34.0	20.4	18.1	16.3	14.7
975-1025	326	157	94.3	72.6	36.4	30.9	17.6	16.0	14.7	13.3
1025-1075	313	146	84.5	68.0	32.9	27.9	15.8	14.5	13.3	11.8
1075-1125	306	136	80.5	65.0	30.2	25.3	13.0	11.6	11.0	10.2
1125-1175	287	130	74.2	58.6	26.9	22.4	11.8	9.6	8.8	8.5
1175-1225	262	118	66.0	54.1	24.4	20.5	10.4	8.9	7.3	6.5
1225-1275	253	108	62.7	52.0	22.8	19.0	9.4	8.5	7.4	6.7
1275-1325	268	112	60.7	48.6	21.5	18.7	9.3	8.3	6.6	7.3
1325-1375	303	122	64.5	51.5	22.4	19.4	9.5	8.6	7.5	6.7
1375-1425	405	146	75.8	57.4	24.5	21.1	9.7	8.7	8.8	7.3
1425-1475	426	159	79.5	60.9	25.3	20.4	9.4	8.8	7.5	7.4
1475-1525	331	122	58.0	46.7	18.9	15.0	8.4	7.5	6.1	6.3
1525-1575	180	77.2	38.9	32.6	13.7	11.0	6.4	5.4	4.9	5.2
	(5,363)	(2,326)	(1,310)	(1,035)	(491)	(411)	(226)	(204)	(184)	(173)

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Table 40

Gamma-ray background reduction

Energy band, kev	0" Fe	3/4" Fe	1½" Fe	3" Fe	5½" Fe	8" Fe
	Counts per minute					
50-150	10,192	2,874	1,437	582	315	263
150-250	6,174	2,575	1,384	500	272	224
250-350	3,504	1,622	891	330	178	152
350-450	2,236	1,124	662	269	147	122
450-550	1,895	887	521	213	120	103
550-650	1,346	660	381	148	82.6	70.1
650-750	1,040	510	300	122	64.9	56.3
750-850	906	425	248	98.4	52.5	44.6
850-950	792	368	211	81.7	41.1	33.9
950-1050	658	296	172	66.8	30.6	25.5
1050-1150	585	266	147	54.3	24.1	19.6
1150-1250	528	224	131	46.7	18.8	15.4
1250-1350	688	254	137	47.2	18.9	15.1
1350-1450	818	307	154	48.1	19	16.4
1450-1550	322	148	81	27.2	12.2	9.8
	(31,683)	(12,540)	(6,855)	(2,633)	(1,397)	(1,170)
1550-1650	185	82	49.2	17.9	9	7.2
1650-1750	160	72.7	41.8	16.3	7.6	5.6
1750-1850	101	48.3	30	12.0	6.1	4.5
1850-1950	68.6	35.9	22.2	10.9	5.5	4.4
1950-2050	73.3	35.8	22.1	9.6	4.8	3.7
2050-2150	78.1	39.6	22.4	9.8	4.8	4.0
2150-2250	64.7	31.2	20.0	9.4	4.3	3.6
2250-2350	55.9	27	17.1	7.5	3.8	3.5
2350-2450	60.9	29.4	17.2	7.3	3.7	3.3
2450-2550	80.4	35.6	18.9	8.4	3.8	3.1
2550-2650	60.7	29.4	18.2	6.7	3.0	2.9
2650-2750	22.9	13.4	9.2	4.4	2.8	2.6
2750-2850	10.8	6.4	5.2	3.6	2.5	2.5
2850-2950	7.2	5.5	4.1	3.3	2.7	2.2
2950-3050	4.6	4.5	3.6	2.6	2.7	2.5
3050-3150	4.9	4.7	3.7	3.0	2.7	2.4
	(1,040)	(501)	(305)	(133)	(70)	(58)

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detector mount, inner plates of the room and cosmic rays. This value should not change appreciably as steel is added to the outside of the room. Subtracting 34 c/m from the background at each wall thickness yields the straight dashed line (Fig. 39), a result consistent with the hypothesis that within this band, 34 c/m originate from cosmic rays and internal activity within the confines of the room and less than 1 c/m originates outside the room.

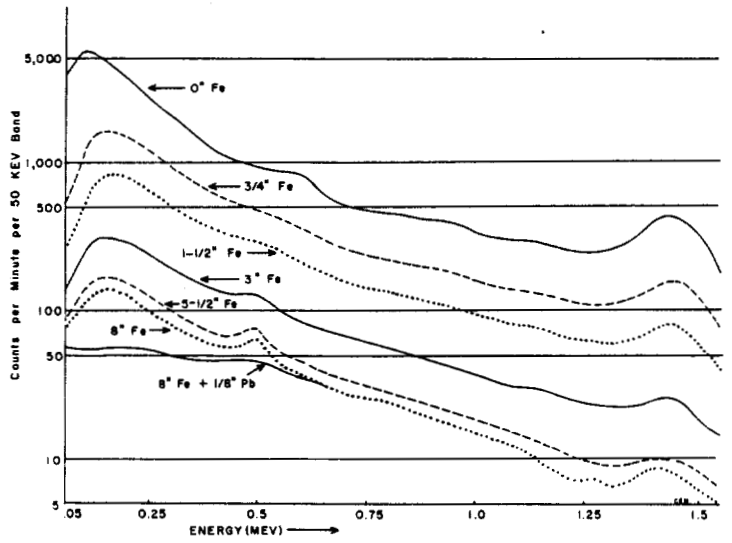

Fig. 38 Scintillation spectra of gamma-ray background of Steel Room II as a function of wall thickness.

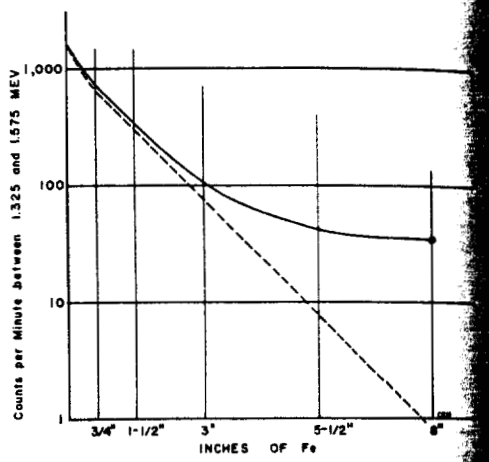

Fig. 39. Gamma-ray background of Steel Room II between 1.325 and 1.575 Mev as a function of wall thickness.

A similar analysis performed across the spectrum indicates that with 8-inch Fe walls about 1 c/m per 50-kev band at the low energies and 0.2 c/m per 50-kev band at the higher energies were originating outside the room. The empirically determined straight lines determined from the calculations at all these other energy spans had practically identical slopes. The mass absorption coefficient calculated from these slopes is 0.045; namely, the mass absorption coefficient of iron from gamma rays in the 1.5 - 2 Mev region. The reason for the fact that the same mass absorption coefficient is found for low energies of the spectrum is not immediately apparent, but this finding could conceivably be due to the fact that most of the low energy pulses observed are due to the degraded high-energy rays from the Ra and Th present in the surrounding concrete.

From the above studies, it seems evident that 95% of the background inside the 8-inch Iron Room is indigenous to the room and cosmic rays; therefore, any practical amount of additional shielding will not significantly reduce the background.

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The data of Table 39 demonstrate also that the low-energy background can be reduced by adding 1/8 inch lead on the interior walls. These low-energy rays which are absorbed by the lead apparently originate within the steel. It has not been demonstrated whether these counts are due to the interaction of cosmic rays or due to contamination of the steel.

III. Iron Room I

The reduction in background realized by covering the floor and one wall of Iron Room I with 1/8 inch sheet lead was reported in the last semiannual report.(1) The remaining interior surfaces have since been covered with lead and the reduction in background noted. An additional shielding of 1/8-inch sheet lead was positioned immediately around the crystal and a further reduction in background noted. This further reduction cannot be easily interpreted, but it may in part be due to the result of shielding the detector from natural airborne activity such as radon and other gases within the enclosure, like A^{41} emitted by the research reactor and N^{13} from the cyclotron. Additional studies will be made and reported at a later date.

The data obtained with Iron Room I and the comparable data from Iron Room II are assembled in Table 41. A comparison of the data of the two rooms seems to indicate that some material used in Room II is contaminated.

The radium and K^{40} present in the photomultiplier tube have been found to be responsible for 275 c/m of the background of the 8 by 4-inch crystal between 25 kev and 1.575 Mev. It has been calculated that the K^{40} content of the crystal would account for an additional 170 counts. These two spectra were subtracted from the actual background. The residual spectrum, suffering from considerable statistical uncertainties, suggests the presence of gamma rays at 510 kev, 860 kev and 1.3 Mev, and the presence of a small amount of Ra^{226}.

It should be realized that although it is desirable to reduce the background as much as possible when counting low activity samples, the background itself, in the region below 800 kev, is not of primary importance when measuring the activity of the human body. As will be shown in a later section, the variations of I^{131} and Cs^{137} in the body seem to be the factors limiting the ultimate sensitivity of the present experimental setup.

IV. A Follow-up Study of an Americium Accident

A chemist who had received a puncture type wound of the hand while working with Am^{241} was studied on September 8, 1955 and the results reported.(3) This individual was brought back on July 25, 1957 and the

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Table 41

Comparison of backgrounds in rooms I and II

Energy band, kev	8" Fe walls		8" Fe + 1/8" Pb on walls		8" Fe + 1/8" Pb on walls + 1/8" Pb around crystal	
	Room I	Room II	Room I	Room II	Room I	Room II
	Counts per minute					
25-75	79.4	76.1	76.2	118	60.9	63.1
75-125	110.5	119.8	66.7	96.4	50.4	59.3
125-175	113	140.8	71.4	98.5	50.5	59.4
175-225	98.4	124.5	69	97.9	52.5	60.2
225-275	80.1	96.2	63.7	85.4	48.7	54.1
275-325	64.8	80.3	58.1	74.2	45.1	50
325-375	55.4	66.2	51.9	64	41.4	49.4
375-425	50.8	60.1	46.1	57.1	41.4	46.1
425-475	50	59.9	48	53.8	40.6	44.1
475-525	44	59.7	46.1	52.7	40.1	43.3
525-575	36.8	42	38.3	41	34.6	36.4
575-625	33.6	36.5	34.4	35.6	30	33.5
625-675	28.5	31	30.5	30.4	24.7	28.6
675-725	26.6	27.1	26.7	28.4	24.7	27.4
725-775	26.5	26.4	24.7	25	25.2	25.7
	(898)	(1047)	(752)	(958)	(611)	(680)
775-825	24.2	24.7	23.4	23.8	22.7	23.6
825-875	21.4	20.3	20	20.9	20.4	20.0
875-925	20.5	19.0	19.9	18.9	17.6	18.7
925-975	17.1	16.3	17.5	15.9	16.7	17.1
975-1025	14.7	14.7	15	14.3	13.3	14
1025-1075	12.5	13.3	12.6	12.9	12.3	12.8
1075-1125	10.9	11	11	11.5	10.5	11.1
1125-1175	9.2	8.8	10.2	9.9	9.4	9.2
1175-1225	8.1	7.3	8.2	7.9	8.5	8.7
1225-1275	7.3	7.4	7.8	7.4	7.2	7.7
1275-1325	7.6	6.6	8.2	7.3	7.7	7.9
1325-1375	8.2	7.5	8	7.3	8.7	7.8
1375-1425	8.4	8.8	9.4	7.5	8.7	8.2
1425-1475	8.8	7.5	9.1	7.6	8.3	7.4
1475-1525	6.8	6.1	6.9	6.4	6.7	6.5
1525-1575	5.4	4.9	5.7	4.9	4.8	4.6
	(191)	(184)	(193)	(184)	(184)	(186)

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study of his gamma activity was repeated with the same collimated crystal. The body burden and body distribution was identical with that found almost 2 years ago, indicating no appreciable migration within the interval.

The hand was scanned using a 1/4 by 1/4-inch NaI(Tl) medical scintillation probe, previously described.[4] The bulk (ca. 90%) of the remaining material was found to be concentrated in one area, less than 1/4 inch in diameter, just below the surface about 1/8 inch to the side of the old incision line. Figure 40 shows a 1/2-inch diameter circle drawn around the outside of the crystal holder. The activity was located in the darker erythematous area. Utilization of this type of scintillation probe in the operating room should permit ready excision of the contaminated tissue.

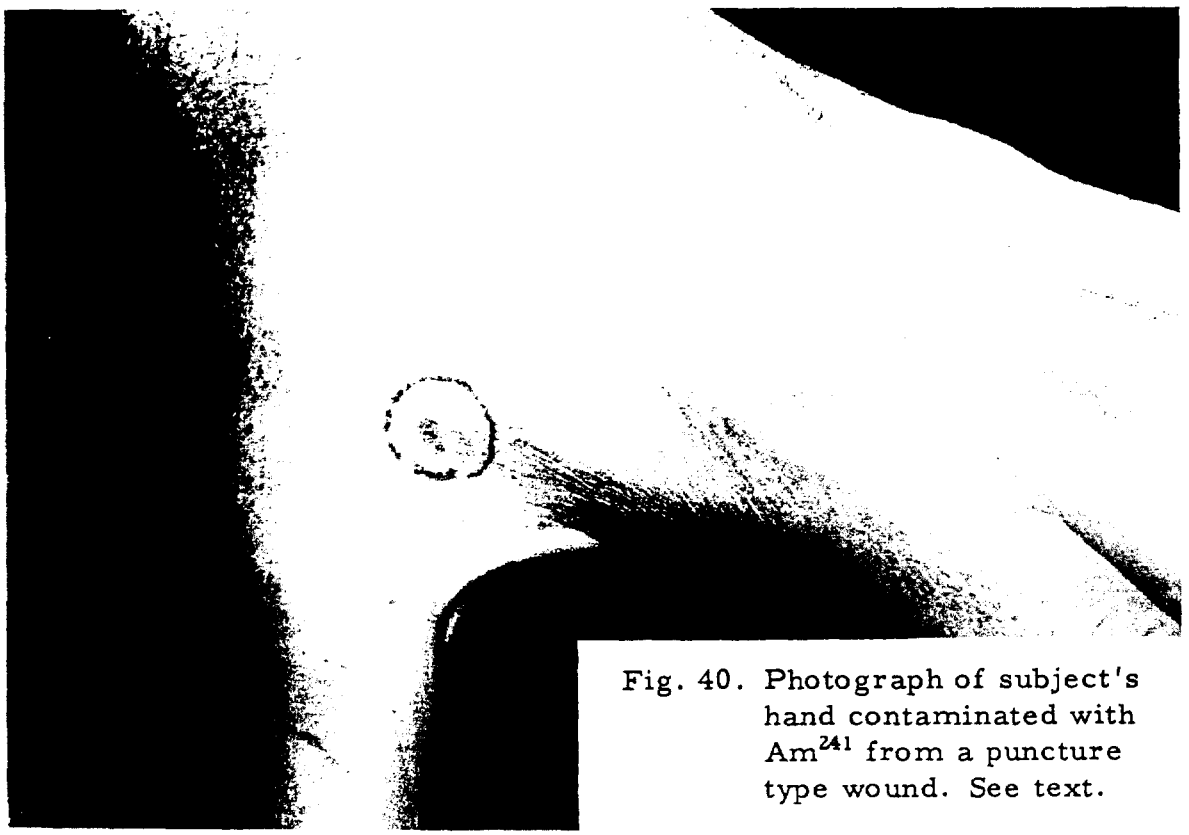

Fig. 40. Photograph of subject's hand contaminated with Am^{241} from a puncture type wound. See text.

V. The Presence of I^{131} Fallout in Humans

The net *in vivo* spectrum of CEM was obtained 8-1-57 in the course of the long-range evaluation of Cs^{137} in humans. The low-energy end of the spectrum was observed to be higher than was to be expected from previous

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observations (Fig. 41). The run was repeated the following day with identical results. In order to pursue the matter further, the subject's net in vivo spectrum of 3-22-57 was subtracted from the anomalous spectrum, and the spectrum of Fig. 42 resulted. The main radioactive isotope responsible for the result was found to be I^{131} with some Ru^{106}-Rd^{106} and Zn^{95}-Nb^{95} present. The Cs^{137} content was also slightly higher than as of 3-22-57. A collimated crystal was placed over the subject's thyroid and a distinct I^{131} spectrum was evident.

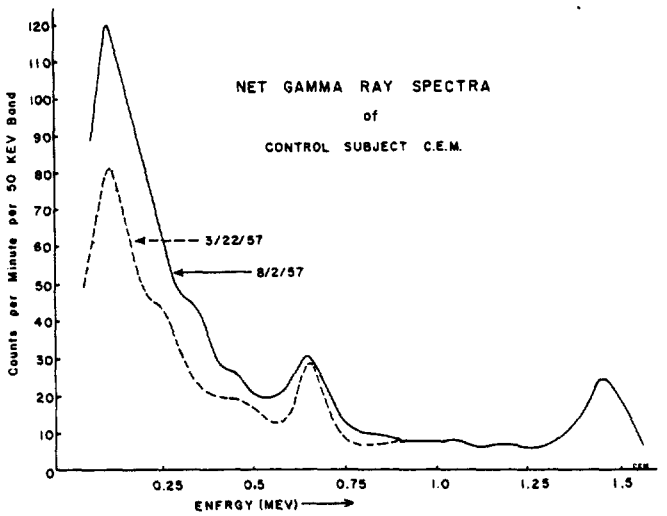

Fig. 41. Net gamma-ray scintillation spectra of control subject CEM.

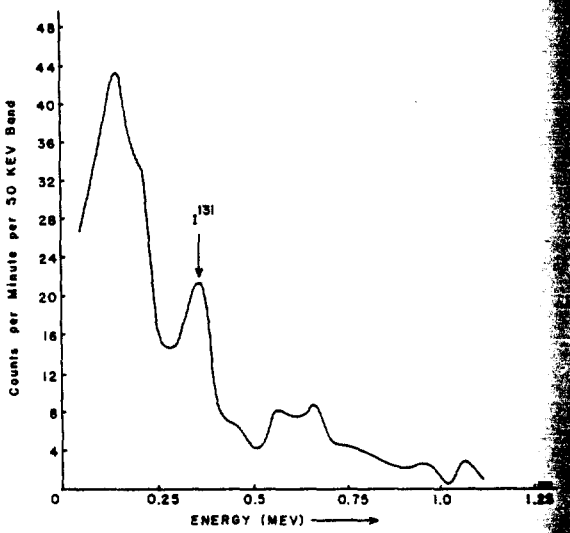

Fig. 42. Increase in net spectrum of control subject CEM from 3-22 to 8-2-57.

All the other control subjects were then checked near that date, and amounts varying from 10^{-10} to 10^{-9} c of I^{131} were found.

Additional subjects from other parts of the U.S. were also checked, and the I^{131} photopeaks observed were of the same order as those found in local residents.

VI. Ra^{226} and Th^{232} Activity in Humans

Interest in the long-term effects of radioactivity in humans has focused our attention on individuals who, by one means or another, have accumulated radium or thorium in their bodies for long periods of time. Contemporary literature indicates that thousands of people in all parts of the world have accumulated varying amounts of radioactive materials

from industrial employment or medical treatment. However, the number of individuals adequately studied,[5,6] is not sufficient to establish, within satisfactorily narrow limits, the incidence of pathological manifestations as a function of radioactive burden. Moreover, this limitation in number is aggravated by the fact that many of the subjects were selected for study by the appearance of clinical symptoms. It is obvious that an extension of this type of study to unselected large groups is most desirable. In this laboratory, in addition to the inquiry on the relationship between incidence of bone sarcoma with radium content of potable water of Illinois by A. F. Stehney, H. F. Lucas, and H. Auerbach, effort has not been spared to locate groups of individuals suited to the purpose of this task.

Radium Subjects

In the last few months, many individuals formerly connected with the radium dial paint industry have been traced by virtue of persistent efforts requiring a great deal of time mostly devoted to personal contacts with the subjects involved and to word-of-mouth education as to the strictly scientific intent of our labors. Response has ranged from the most enthusiastic cooperation to downright suspicious hostility, but the over-all effect has been gratifying. By means of group pictures at the time of employment, autograph and address books, Christmas card lists, etc., a nonindifferent group of about 300 people has been identified. Of these 42 have already been measured and found to have from 10^{-9} to 3×10^{-6} c of Ra^{226}. It is hoped to continue the study in the immediate future and to establish, if possible, the entire numerical potential in the vicinity.

Thorium^{232} in Humans

Two groups of thorium-burdened people seem suitable for study: industrial workers of thorium refineries and patients injected with Thorotrast.

Thorium refineries have been handling thorium compounds for many years, and undoubtedly many workers in these plants must carry detectable amounts. Two hundred people are located in the vicinity of the Laboratory and hence potentially available for study. Since the main route of contamination is the lung, the physical studies in prospect are not expected to be simple. One subject - connected with the firm for several decades - was measured as a test some time ago with the $4 \times 1\frac{1}{2}$ inch NaI crystal and showed Ra^{228} and Ra^{224} photopeaks corresponding to about 50 mg of Th^{232} in equilibrium with its daughter products.

Thorotrast (ThO_2) was injected into humans during the early thirties for various diagnostic purposes. The potential usefulness of this group, however, remains to be elucidated because of the unsettled question of life

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expectancy. It is to be noted, however, that the injected group is undoubtedly large (a single incomplete survey[7] reports nearly 5,000 cases in the United States alone), and that since the work was performed mostly by radiologists in hospitals, and records are available, there exists a good opportunity of follow-up. Moreover, since the volumes injected do not vary greatly and - judging from work reported to date - the injection of ThO_2 is followed by a quantitatively fairly well defined pattern of deposition and excretion - it might be possible to evaluate the series by relatively modest effort without a great number of physical measurements beyond those required to establish without doubt the dosimetric features of the element. To date one Thorotrast patient has been measured and it is reported below:

Study of Thorotrast in Humans

The in vivo gamma ray spectrum of a patient who received 10 cc of Thorotrast intravenously in 1939 was obtained utilizing the "one-meter-arc" and the "standard-chair" techniques. In addition, a rough estimate of the body distribution was determined by positioning the crystal 1 cm above the umbilicus, abdomen and head. Both the absolute quantity and the ratio of the photopeaks to backscatter troughs were much higher at the head indicating that a considerable amount of the material was still present at the locus of the injection in the neck.

The 1-meter-arc spectrum and the standard-chair spectrum are identical if the value of the ordinates of the latter are reduced by a factor of 5.5. This ratio is higher than the 4.1 ratio found in the past for subjects containing Ra^{226}, Cs^{137} and K^{40}. This may be due to the unusual anatomical features of this subject, only 5' 2" tall but weighing 218 pounds; it may also be due to the fact that the material was not uniformly distributed throughout the body volume.

The spectra were analyzed in the following manner: The gamma-ray spectrum of a normal unexposed subject of the same height and weight was subtracted from the spectrum of this Thorotrast patient, yielding a net thorium spectrum as shown in Figs. 43 and 44. The spectra obtained from both a RdTh source and an old Thorotrast source (1937) buried in a 28-cm phantom were normalized so that the area under the 2.62 Mev ThC' photopeak matched the area under the patient's ThC" 2.62 Mev photopeak. The normalized RdTh spectrum was then subtracted from both the patient's net spectrum and the normalized Thorotrast spectrum. The difference spectra, thus obtained, which are due to MsThII, should have been identical if the MsThII in the patient had been in equilibrium with its daughters and the scattering from the phantom was representative of the patient as a whole. From the portion of the patient's and phantom's spectra above 1 Mev, there is no reason to question the latter assumption.

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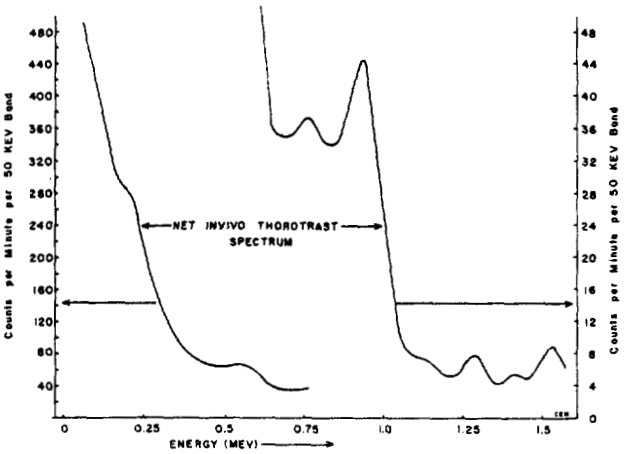

Fig. 43. Net in vivo scintillation gamma-ray spectrum from 25 kev to 1.6 Mev obtained from a Thorotrast patient.

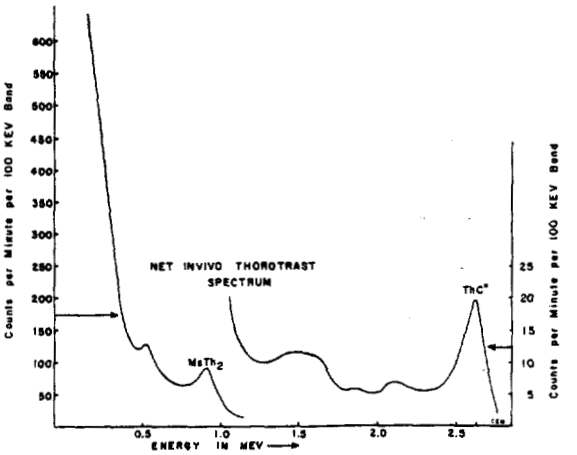

Fig. 44. Net in vivo gamma-ray scintillation spectrum from 50 kev to 2.8 Mev obtained from a Thorotrast patient.

Since the subject was found to contain 1.22×10^{-7} c of MsThII and 1.1×10^{-7} c of ThC" or 12% more MsThII than the source, it seems likely that the patient was eliminating about 12% of the daughter products between MsThII and ThC". The value compares very well with the value found by Hurst et al. for the elimination of Ra^{224} from Thorotrast patients.(8)

VII. Observations and Calculations Concerning the Error of Ra^{226} Measurements *in vivo*

The 1-meter-arc technique as described in another report(9) is the absolute method for determining the amount of radioactivity in a human body. In practice, measurements are made with the subject in the standard chair.(10) The reasons for using the chair are that the subject is more confortable, the subject's body can be more reliably positioned, and the crystal can be moved closer to the subject's body with a resultant higher counting rate and greater sensitivity.

As discussed in an earlier report(11) the chair position cannot be calibrated using a phantom since all parts of the body are not equally distant from the crystal. For certain purposes, such as measurement of K^{40} content, the chair can be "calibrated" directly by the administration of tracer doses of K^{42} to a standard subject. However, for obvious reasons, this technique is not applicable in the study of long-lived bone-seekers.

A radium and thorium calibration factor for the chair has been determined indirectly. A number of subjects with relatively large burdens of these elements have been counted both in the chair and on the arc. The

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absolute amount in microcuries has been determined from the arc data. The net in vivo radium or thorium spectrum in c/m per 50-kev energy band obtained with the chair has been divided by the amount in microcuries found by the arc technique. The resultant parameter (counts per minute per μc per band, in the chair) can be used to calculate other subjects' body content directly from the chair measurements.

The gamma-ray spectrum of a radium patient is analyzed as follows: The net gamma-ray spectrum of a normal unexposed control subject of the same height, weight, and general build is subtracted from the net gamma-ray spectrum of the radium subject yielding the in vivo radium spectrum. The total counts per minute in each of three energy bands (75 kev to 375 kev, 525 kev to 675 kev, and 775 kev to 1.175 Mev) are divided by the factor evolved above, giving three independent evaluations for the subject's body content. The three values should agree if the net spectrum is due entirely to radium. The values will not agree if the patient's Cs^{137} or K^{40} level differed from that of the control's, or if there are any additional isotopes present. This may be the case when the Ra^{226} burden is low.

The theoretical uncertainty in a radium measurement is determined by the statistical variations in background in the spectrum of a normal unexposed control subject and in the in vivo radium spectrum. The actual measured values for background, CEM's natural and the calibration factors for radium are given in Table 42. The statistical variations to be expected for a 50-minute counting period have been computed for background and subject CEM.

Table 42

Uncertainty in in vivo radium measurements

Standard chair technique, counting time of 50 minutes

Energy band, kev	Background, c/m	CEM		In vivo c/m per μc of Ra	Theoretical uncertainty, μc of Ra	Realistic uncertainty, μc of Ra
		Gross, c/m	Net c/m			
75-375	408.5 ± 2.9	714.6 ± 3.8	306.1 ± 4.7	11,000	6.1×15^{-4}	3.10^{-3}
525-675	103.7 ± 1.4	168.2 ± 1.8	64.5 ± 2.3	1,650	2.1×10^{-3}	1.2×10^{-2}
775-1,175	105.5 ± 1.75	215.3 ± 2.1	64.8 ± 2.8	1,500	2.8×10^{-3}	2×10^{-3}

The statistical variation in the patient's net data due to his natural K^{40} and Cs^{137} and due to the room background will be the same as that given for the "Net CEM" column. Subtracting the net CEM control data

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from the patient's net data yields a net difference with a statistical uncertainty of $\sqrt{2}$ times the net CEM uncertainty. The minimum theoretical uncertainty in a calculated body content (expressed in μc of radium) is found by the following expression:

$$\text{Minimum uncertainty} = \frac{\text{variation of net CEM} \times \sqrt{2}}{\text{c/m per c of radium } \underline{\text{in}}\ \underline{\text{vivo}}} .$$

This value could be realized if the control subject were measured, given a tracer and remeasured within a few days.

In actual practice, the uncertainty is much greater since the amount of Cs^{137} depends upon an individual's diet. Control subject CEM's net counting rate was observed to increase by 164 c/m in the low energy band, 23.5 c/m in the middle band and 13.5 c/m in the high energy band between 3-22-57 and 8-2-57 because of I^{131} fallout intake. The errors resulting from using this individual's March spectrum as a control spectrum in August would result in errors of 1.5, 1.4 and 1×10^{-8} c, respectively.

The variation resulting by the substitution, for purposes of control, of people of the same height and weight at the same location on the same date would not be quite as extreme. Data from the control subjects suggest that a variation of 30 c/m in the lower band, 20 c/m in the middle band and 3 c/m in the upper band are representative of actual fluctuations leading to realistic uncertainties of 3×10^{-9}, 1.2×10^{-8}, and 2×10^{-9} c. See Table 42. The measured values for the radium dial painters support these as realistic uncertainty values.

VIII. Standard Deviations in the Measurement of Potassium in Humans

The uncertainty in measurement of K^{40} in a human being depends upon several factors such as the person's build, the relative position of the crystal with respect to the body, length of counting time, and energy band. The first two factors were examined and the results reported in a previous report from this Laboratory.(11) The remaining two factors have been examined and shown in Fig. 45 where a typical scintillation spectrum from an "uncontaminated" individual is shown.

Three energy bands were chosen as follows: Band I: 0.775 Mev to 1.275 Mev, Band II: 1.325 Mev to 1.575 Mev, and Band III: 0.775 Mev to 1.575 Mev, which includes Bands I and II. A counting time of 50 minutes was used for the background run and 10 minutes for the subject's run. The counting rates of background, subject J. P. (gross and net counts) and the attendant standard deviations are shown for each energy band.

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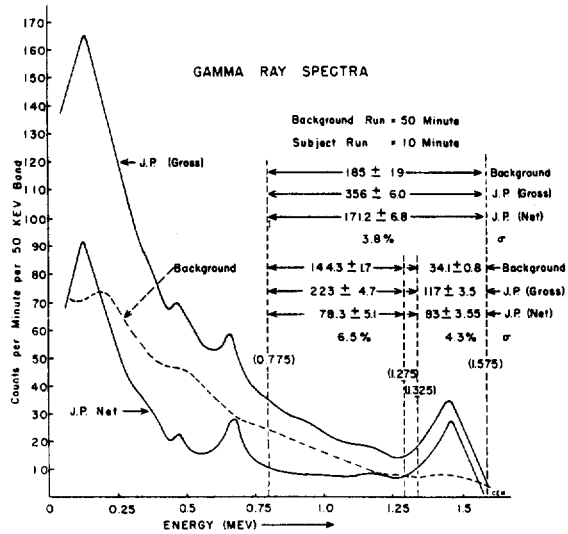

Fig. 45. Gamma-ray scintillation spectrum of background, gross and net, of typical uncontaminated subject. The standard deviations of the net spectrum, assuming a 10-minute run, have been calculated for 3 bands.

It seems apparent that it is possible to determine a subject's K^{40} level with a standard error of 4%, using a 10 minute run.

In the past we have calculated a human's K level by using Bands I and II independently. Agreement between the two resulting values (by calibration with K^{42}) is a proof that the K^{40} value is correct and that the subject did not contain any appreciable amount of any other element emitting gamma rays in these bands.

IX. The Measurement of Natural Gamma Activity in Humans above 1.5 Mev

It has been observed for several years that with an 8-inch by 4-inch crystal a net human in vivo gamma-ray spectrum yielded about 6 counts per minute between 1.7 and 2.8 Mev over a background of 50 counts per minute. This section of the energy spectrum was examined in detail (1) to explain the source of these counts, and (2) to determine the possibility of measuring the natural radium and thorium content of humans by detecting the high energy gamma rays from these isotopes. In theory, greater accuracy might be achieved using this energy band since no other isotope occurring in humans emits gamma rays above the 1.46 Mev of K^{40}.

On the basis of data obtained from subjects with relatively large Ra^{226} burdens, the 6 c/m would suggest a body content of about 10 times the natural value, since it would require a burden of 6×10^{-9} c of radium or 1.2×10^{-9} c of thorium to yield 6 c/m. A. F. Stehney finds a maximum

of 4×10^{-10} c of radium[12] and predicts about 10^{-10} c of thorium[13] in individuals from the Joliet area. These amounts would account for a total of approximately 1 c/m only.

It has been suspected that the body must be scattering secondary rays from cosmic rays into the detector. The background was obtained with the chair alone, with cans filled with distilled H_2O in place of the humans and with empty cans. The background with the distilled water present was about 5 c/m higher than the other two, demonstrating that the body would increase the apparent background between 1.7 and 2.8 Mev. The increase in background at low energies, while detectable, is not significant when compared to the actual counting rates.

These additional counts are probably the result of cosmic rays interacting in the human or the H_2O. Comparison of the background from 2.85 to 3.25 Mev, which is above the most energetic naturally occurring gamma ray, with the background from 2 to 2.85 Mev, leads to the conclusion that most of the background counts in the latter energy band are due to cosmic rays. The Ra and Th present in unexposed humans might be measured utilizing these energy bands if the cosmic rays' photons produced could be eliminated by anticoincidence counting. The counting rates will be rather low, about 1 c/m, and long counting periods will be required. A 16-hour count, the time usually required for an accurate radon breath test on a normal subject,[12] would give 960 counts due to the same source. This could be measured with about 10% uncertainty of the background. Some advantages might accrue: the subjects may be more confortable during these long counting periods than when being measured on the radon breath apparatus. They would not have to wear a mask, with the attendant discomforts, but would recline in the contour chair. The run could be interrupted and the subjects allowed to walk around for periods of time with no adverse effects. This probability will be kept under consideration.

X. Inhalation Accidents

In the period since the last report, three people who inhaled radioactive dust have been brought to our laboratory from out of town for an estimate of content and biological half-life. One individual, a machinist who inhaled a uranium compound containing U^{235} during a grinding operation, has been measured on three occasions. The other two subjects, who inhaled air-borne Pa^{233}, have been measured twice. All will return at future times for further studies.

The total body content, the amount in the lungs, and the quantity in selected volumes of the body have been measured during each visit. The subject's total body content has been measured using the standard-chair and the 1-meter-arc techniques. The total amounts in the lungs, head, shins, and abdomen have been measured by placing the 8-by-4-inch NaI(Tl) crystal about 1 cm over the ventral thoracic, dorsal thoracic, abdominal

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and lumbar regions and the shins and the head. It is hoped that by repeated measurements the lung clearance rates and the fraction deposited in the bones for each of the two isotopes can be determined with some accuracy and compared to data from excretion studies.

The chest area of each subject was scanned with a collimated detector on his first visit, to determine whether the material was concentrated in any particular area or uniformly distributed throughout the lung volume. Nine points on a grid over the chest were measured. The distances from the top of the subject's head to the horizontal grid lines were 34, 42, and 50 cm, respectively, and the vertical grid lines were the subject's mid-line (over the sternum) and lines 8 cm to the right and to the left of the mid-line. The highest readings were found 8 cm to the right and left of the mid-line, 34 cm from the top of the head. The values on either side of the mid-line, 42 cm down were about 20% less. The points 50 cm down and 8 cm to the right and left were about one-fourth of the top values. The values along the mid-line were somewhat lower than the values on either side, demonstrating that the material was distributed throughout the lungs. Essentially the same distribution was found for all three individuals. These positions will be correlated to lung volumes by superimposing the grid on chest X-rays of each subject to define the chest volume which was actually scanned.

The reading obtained with the crystal 1 cm from the back was identical to the reading obtained with the crystal 1 cm from the chest proving that the material was either located on a plane midway from the front to the back of the individual, or that it was distributed throughout the lung volume.

U^{235} Case Data

The total amount of uranium in the thorax was calculated using the techniques described in the following section. The dates of the measurements and the amounts were:

Date	Total amount in chest area, mg	Decrease, mg
11-2-57	2.75	
11-19-57	2.05	0.7
12-10-57	1.47	0.58

These three values were plotted on a semilog graph, fall on essentially a straight line and indicate a biological half-life in the lungs of 41.5 days. This value is considerably shorter than the value of 125 days usually assumed.

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All of the subject's urine has been collected and analyzed by the employer's bio-assay group. They report that the subject eliminated 0.75 mg during the first period and 0.45 mg during the second period. These values are in substantial agreement with the amount leaving the lungs as estimated by the above measurements. These data demonstrate that most, if not all, of the material is being eliminated from the body after it leaves the lungs and that apparently it is not being deposited in other organs in comparable amounts.

To date, no detectable amount has been found in any organs other than the lungs. The measurement made over the kidney region is a few per cent of the lung value. Measurements made with appropriate phantoms in place of the human demonstrate that the gamma rays detected over the kidney are probably scattered rays originating in the chest volume. More exact measurements of the more sluggish U deposits will be possible after the relatively short-lived lung content has been eliminated.

The half-time of Pa^{233} in the lungs cannot be stated with certainty on the basis of only two measurements. A biological effective half-time, i.e., [T 1/2 (eff) = T 1/2 (phys) · T 1/2 (biol)/T 1/2 (phys) + T 1/2 (biol)] of 22.6 days was found for both subjects. Pa^{233} thus has a biological half-time in the lungs of 129 days which is in agreement with the 112-day half-life found for $RaSO_4$ by Marinelli, et al.(16) but at variance with the half-life in the lung of 66 days for T^{129} and 41.5 days for U^{235} as observed by Miller.(14)

The lung volumes were scanned on the second visit and about the same distribution noted as on the first visit. It was felt that the original distribution might reflect different particle sizes and that the lung clearance rates might also reflect these effects. However, the scans were not exact enough for this detailed study.

XI. Phantom Calibrations for Inhalation Cases

The net counts per minute as obtained from measurements on humans must be converted into an absolute quantity of radioactivity if the measurement (and the facility) is to serve any useful purpose. A constant, such as the net *in vivo* counts per minute per microcurie, must be determined for the particular isotope involved, the particular geometry used and the specific location of the isotope in the body.

The inhalation case is the most difficult one to calibrate since the material is distributed throughout a low-density volume which is surrounded by a denser layer of unknown thickness. The U^{235} and Pa^{233} inhalation cases have enabled us to reappraise and extend some previous observations. The data obtained with U^{235} will be given in detail.

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The expression, counts per minute per microcurie, must be obtained from an appropriate phantom since it is impractical to use tracers in humans for inhalation studies in these cases. The ratio of the area under the backscatter peak to the area under the photopeak may be used to test the "equivalence" of the phantom to the human. In the case of U^{235}, the ratio was established as the area under the curve from 71 kev to 126 kev (the uranium X-ray plus backscatter from the 188-kev gamma ray) divided by the area under the curve from 154 kev to 209 kev (the 188-kev photopeak). The in vivo ratio was found to be 1.14. The measurement and ratio from the back of the subject was identical to the reading taken from the front.

The following tests were made to arrive at an equivalent phantom. A 100-mg uranium source was placed at various depths in a 20-cm thick phantom built up of 1/4-inch sheets of Presdwood. The crystal was positioned 1 cm above the top plate of the stack as it was for the measurement in vivo over the thorax. The ratio was found to vary from 0.8 when the source was placed on top of the phantom to 1.8 when the source was buried the full 20 cm. The phantom ratio as measured at the top surface matched the in vivo ratio when the source was buried under the 1.4 cm of Presdwood. This, of course, cannot be the location of the U^{235} source in vivo. Moreover, it was not possible to distribute sources through a solid phantom which would yield this ratio for both front and back measurements. The contribution from the source buried at the far side increased the ratio considerably above the desired value. This indicated the total mass through the phantom was too great.

A phantom was constructed as shown by Fig. 46 to approximate the mass distribution of the human. The mass at positions X and Y correspond to the denser masses of the chest walls and the spaced area approximates the spongy tissue of the lung cavity. The over-all thickness of the phantom was made equal to the subject's thickness to provide the same geometry.

The source was located at the center of the phantom and the thicknesses used at X and Y varied. Varying the back thickness (Y) varies the area under the backscatter peak only. Increasing the front thickness (X) reduces both the photopeak and the backscatter peak. The phantom uranium spectrum matched the in vivo spectrum as shown by Fig. 47 when 6 cm of Presdwood was used in positions X and Y.

The chest span indicated that the material was distributed through the lung volume rather than concentrated at a point. The calibration of the phantom must therefore be corrected for this effect. The source was placed at the five positions (A,B,C,D,E) shown by Fig. 46 and the spectra obtained. Due to the inverse square effect, the average reading was 7% higher than the reading obtained with the source at the center, point C. The ratio varied from 1.04 when the source was at position A to 1.3 with the source at position E, with an average value of 1.12.

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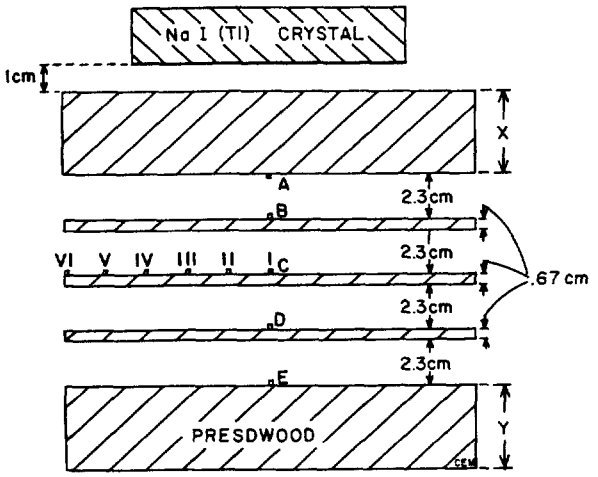

Fig. 46. Cross-sectional view of Presdwood phantom used to reproduce a gamma-ray spectrum obtained from radioactivity in a human's lung.

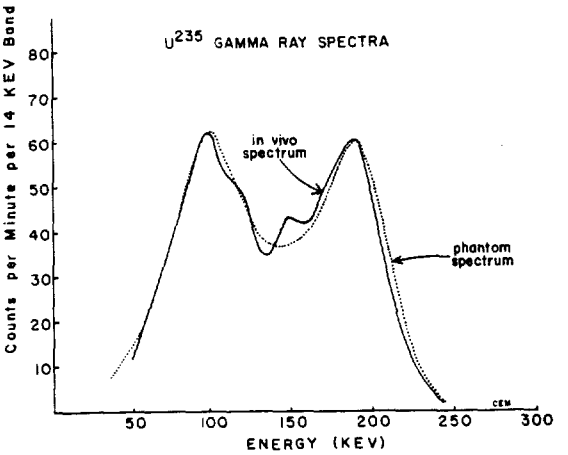

Fig. 47. A comparison of U^{235} gamma ray spectra obtained from the lung-equivalent phantom with an actual lung burden.

The source was then placed at the center of the phantom and at 3, 6, 9, 12, and 15 cm to the left on the mid-plane as shown by positions I through VI. The ratio did not change although the average value for the simulated distributed source was 33% lower than the value at the center.

The counts per minute per milligram of U^{235} as found with the source located at the center was multiplied by 1.07 to account for the inverse square effect and by 0.67 to account for the distribution effect. The net counts per minute as measured from the subject divided by this factor yields the lung content in milligrams.

Essentially the same approach was used during November, 1956 to build up an equivalent lung phantom for two Te^{129} inhalation accident subjects. The same phantom thicknesses and effects were noted at that time.

The spacing between the front thickness (X) and the back thickness (Y) has a critical effect on the ratio. The crystal subtends a particular solid angle to the unscattered gamma rays from the source itself. The solid angle subtended to forward scattered rays in volume X is higher than the solid angle to the direct rays. The effective solid angle for the back scattered rays from volume Y is considerably lower than either of the other two. Bringing the volume Y closer to the source increases the backscatter solid angle and increases the amount of backscatter detected by the crystal. Therefore the spacing between X and Y must be equal to the lung cavity.

The actual spacing used between the masses X and Y is not as critical as when the detector is placed 1 meter from the subject because the solid angles subtended to the front and back surfaces are almost equal. However, the thickness of volume X is still very important since increasing X increases the absorption and thus affects the counting rate per microcurie.

The physical construction of the phantom which is used to calibrate the 1-meter-arc thus depends upon the location of the radioactive element in the body, and this can be important when dealing with relatively soft gamma radiation. The spectrometric trial and error technique(14) will probably result in more accurate values than the empirical approach.(15) This was also suggested by measurements performed. In the case of one radium dial patient, where a phantom 18 cm thick was suggested by empirical methods based on the subject's dimensions it was found that an equivalent phantom 14 cm thick yielded the correct spectral shape.

In the case of a subject who had just drunk a K^{42} tracer, it was necessary to bury a K^{42} source 6 cm deep in a 22-cm thick solid phantom to match the spectral curve obtained from the front and back of the subject.

The author is pleased to acknowledge his indebtedness to Mr. L. D. Marinelli for his valuable collaboration and guidance during the course of this work. Thanks also due to Mr. J. B. Corcoran for his capable technical assistance.

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