ANL-5919

702286

Finkel

Argonne National Laboratory

RADIOLOGICAL PHYSICS DIVISION

SEMIANNUAL REPORT

January through June, 1958

REPOSITORY Argonne Lab

COLLECTION

BOX No.

FOLDER

0016904

PARTIAL DOCUMENT

TABLE OF CONTENTS

	Page
The absorbance change of irradiated organic solvents. Part VI I. B. Berlman	5
Drift velocity of electrons J. C. Bowe	7
Progress report on flux and spectrum measurements of the cosmic ray neutron background R. Grismore, L. D. Marinelli and B. G. Oltman	11
Variable energy neutron source R. Grismore, B. G. Oltman and L. D. Marinelli	14
Radium toxicity studies R. E. Rowland, J. H. Marshall and J. Jowsey	19
The effect of parathyroidectomy on haversian remodeling of bone Jenifer Jowsey, R. E. Rowland, J. H. Marshall and F. C. McLean	27
Calculation of the possible effects of calcium diffusion in bone crystallites J. H. Marshall and R. E. Rowland	35
Positive ion emission H. A. Schultz	47
Determination of Sr^{90} in biological materials D. P. Krause	53
Determination of Ac^{228} D. P. Krause and A. F. Stehney	54
On the stratospheric Sr^{90} fallout P. K. Kuroda	55
Illinois municipal water supplies having unusually high concentration of Ra^{226} H. F. Lucas, Jr.	58
Gamma-ray emitting radioactivity in soil P. F. Gustafson	61
The human spectrometer. Introduction C. E. Miller	64

0016905

TABLE OF CONTENTS

Page

I. The gamma-ray background of Iron Room I (airborne radioactivity)
C. E. Miller and J. B. Corcoran 64

II. Ra^{226} and Th^{232} activity in humans
C. E. Miller 66

III. Techniques which may yield more accurate Ra^{226} measurements *in vivo*
C. E. Miller and O. J. Steingraber. 67

IV. Follow-up of U^{235} inhalation incident
C. E. Miller 71

V. The elimination pattern of Co^{60} acquired by inhalation
C. E. Miller and H. A. May 72

VI. *In vivo* measurement of Cs^{137}
C. E. Miller and L. D. Marinelli 74

Some investigations on the source of background counts in NaI(Tl) crystals
P. F. Gustafson, L. D. Marinelli and S. S. Brar 77

On the nature of the 3.25-Mev peak in large crystals
H. A. May 85

Background of Iron Room II
H. A. May 89

Evaluation of RCA 5-inch tube, Type 7046
H. A. May 91

Optimum performance from large NaI(Tl) crystals
H. A. May 92

The effect of meteorological variables upon the vertical and temporal distributions of atmospheric radon
H. Moses, A. F. Stehney and H. F. Lucas, Jr. 102

Argonne Meteorological Model Towing Tank
P. Frenzen 113

0016906

TABLE OF CONTENTS

Page

Atmospheric diffusion studies
Peter E. Wasko and Harry Moses 125

Staff activities 139

Publications 140

0016907

THE HUMAN SPECTROMETER

Introduction

C. E. Miller

As in previous reports from this Division, under this heading are collected some of the research activities carried out in our low-level facility with scintillation gamma-ray spectrometer. With the availability of three iron-shielded rooms on our site, these activities have expanded to include measurement of radioactivities in soil and some much needed developmental studies concerning stability and magnitude of background pulse height scintillation spectra. To obviate the considerable editorial work necessary to the coordination of all phases of this work, we report under this section only activities that have taken place in the first iron shielded room, and these concern mainly experiments in vivo. The remainder of the work is reported under the authorships of P. F. Gustafson and H. A. May.

I. The Gamma-Ray Background of Iron Room I (Airborne Radioactivity)

C. E. Miller and J. B. Corcoran

The gamma-ray background within a heavily shielded enclosure has been discussed in previous semiannual reports.(1-6) Extensive data are given in the last report(6) regarding the reduction of the gamma-ray background of an 8-inch diameter by 4-inch thick NaI(Tl) crystal as a function of thickness of Fe shielding. The data given herein are, in a sense, an extension of the past reports, for they concern the ultimate sensitivity attainable by our methods. The background of Iron Room I had been observed to fluctuate because of three different airborne radioactivities. At times the fluctuations were traced to the presence of A^{41} from the research reactor or of N^{13} from the cyclotron(4) by the simple inspection of the photopeaks resulting therefrom. The presence of even a very small concentration of either of these products has seriously affected the accuracy of measurements and, at times, has forced a suspension of all runs.*

To overcome this difficulty a storage air system was installed so that air free of radioactivity could be fed to the iron rooms. The storage system consists of two 500-cubic foot tanks which can be filled to 10 atmospheres. Through appropriate valves, air can be pumped into, or used from, either tank independently of the other. Normally, the two tanks would be

*Similar inconveniences were experienced by Dr. P. F. Gustafson in the low-level facility installed in the Biological and Medical Research Division (vide infra).

0016908

filled when the wind is not blowing from the above sources. In the case of a prolonged southerly flow of several days, one tank can be filled and the air aged for 24 hours to allow the 1.83-hr A^{41} (or the shorter lived N^{13}) to decay while air is being used from the other tank. In other instances the difference between routine background spectra has been large enough to yield a true Ra(B+C) spectrum, hence it must be presumed to be due to fluctuations in the amounts of radon daughters present within the room. The possibility that parts of these fluctuations are due to translocations of the active deposit within this room cannot be totally excluded although it seems unlikely because, to our knowledge, there is no substantial electric field present in the room, all electric cables being covered by grounded shields. In order to obtain some idea of the effect produced by the presence of radon in the room, air which was aged for ten days, was used to inflate two plastic weather balloons placed on either side of the crystal which was located in the center of the room. Under these conditions the background decreased from 915 to 860 cpm and climbed to 895 overnight as the balloons deflated partially and basement air diffused into the room. This experiment confirmed our suspicions on the detectable role of Ra(B+C) in the background, but because of the long time taken in the readings, it did not yield any information on either the location of the active deposit or its magnitude as a function of Rn concentration in the room.

To obviate contamination with Rn spilled in room, the latter was sealed on the outside with polyethylene sheeting which was in turn taped to the floor. The room was kept sealed for six days during which the background climbed from 965 to 1212 c/m. The growth of the background, assessed at 0.75, 1.75, 2.75 and 5.75 days after sealing, bore to a typical radon growth curve arbitrary ratios of 4.65, 3.72, 4.45 and 4.26, strongly suggesting that the increase was due to radon growing from a virtually freely emanating Ra source. At the peak the Rn concentration was 3.25 $\mu\mu$c/liter. Upon flushing the now unsealed room with aged tank air (0.002$\mu\mu$c/l.) the concentration fell to 0.15 $\mu\mu$c/l., and the background counting rate dropped to 907 c/m. Left overnight, the Rn concentration climbed to 0.22 $\mu\mu$c/l. and the counting rate to 911 c/m. These data indicate that the background count between 25 kv and 1575 kv increased by approximately 100 c/m per each $\mu\mu$c/l. in the room.

Under the conditions of the experiment either (or both) the walls of the room or the concrete floor may be responsible for the production of radon; from what is known about the radioactivity and porosity of building materials, the floor is by far the most probable. Further study of this problem will lead to improvement in stability of background.

One puzzling feature remains. After sealing the room, if the circulating air blower within the room is kept running, the background does not change appreciably (± 2%), even if the seal is kept as long as five days.

0016909

Apparently circulation of air must contribute to a translocation of Ra (B+C) away from the crystal, for it is difficult to envision this ventilation preventing diffusion of radon from the floor into the room. This phenomenon may be of importance in methods aimed at minimizing and stabilizing that portion of the background count due to decay products of radon.

The authors are pleased to acknowledge their indebtedness to Mr. H. F. Lucas, Jr., for his measurements on the concentration of radon in the air of the steel room.

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

C. E. Miller

As discussed in the previous semiannual report,[6] a relatively large number of individuals who have accumulated significant quantities of either radium or thorium some 30 years ago are the object of a systematic study in this laboratory.

A. Radium Subjects

Eleven women who painted radium watch dials during the 1920's have been brought in for total body gamma-ray measurements. The radium body content of these subjects has ranged from 10^{-9} to 2.5×10^{-6} c of Ra^{226}. Total skeletal x-rays of twenty of the dial painters have been obtained under the direction of A. J. Finkel, M.D., Director of the Health Division, Argonne National Laboratory. To date, the number of people measured is 68, and the number x-rayed is 36.

In addition, the radium body contents of two individuals who received radium therapeutically 20 to 30 years ago have been determined. One individual (H.T.) contained 4μc and the second (C.F.G.) contained 4.2μc. Two individuals, (B.M.) and (W.F.), reported previously from our laboratory, who received the material from the same source had burdens of 1.2μc and 5.2μc, respectively. Since these cases are discovered usually as a result of clinical or roentgenological symptomatology, it should not be surprising that their burdens are, as a whole, well above those found in cases of industrial exposure. What is comforting, but not reassuring, is the fact that the minimum symptomatological burden has not decreased since 1952.

B. Study of Thorotrast in Humans

The evaluation of the long-term effects of both radium and thorium is complicated by the fact that both elements are the parents of long families of radioactive daughters. The problem is particularly difficult in the case

0016910

of thorium since this element decays through a series of 6 others, two of which are long-lived, before it reaches the stable element Pb^{208}. The daughters are metabolized and excreted in different manners. Thus the metabolic pattern of each daughter must be understood in detail before integral dose calculations for particular organs can be made.

Reynolds, et al.[7] calculated the retention and elimination of radium isotopes produced by the decay of thorium parents within the body (calculations and comparison with experimental findings). These results are being verified by clinical studies now in progress following the I.V. injection of Thorotrast to appropriate subjects under medical care. The amount of thorium-232 and of each daughter is determined before the material is administered. The empty vial and hypodermic syringe are measured after the injection to determine the amount of Th^{232} and of each daughter remaining on the walls of the glassware. The quantity of each element administered to the patient is thus known. At a later date, the amount of Ra^{228}, Th^{228}, Ra^{224}, Pb^{212} and Po^{212} present in the liver, kidney and bone is determined.

One case has been studied to date. Measurements have demonstrated that a large fraction ($\sim$75%) of the Ra^{228} daughter remained in the vial. Consequently, it is imperative that the empty vials be measured carefully since a small error in the measured value remaining in the vial results in a large error in the estimate of the quantity administered. This situation does not exist in subjects to whom the material was injected many years ago, for most of the Ra^{228} present is the result of growth from Th^{232} retained in the body.

III. Techniques Which May Yield More Accurate Ra^{226} Measurements *in vivo*

C. E. Miller and O. J. Steingraber

A. Coincidence Techniques

As discussed in the last semiannual report,[6] the practical uncertainty in an *in vivo* radium determination using the tilting chair technique is about 3×10^{-9}c. This large uncertainty is due to the presence in the human body of small quantities of fission products. For example, the relatively abundant 607-kev gamma ray emitted by RaC' cannot be easily distinguished from the 661-kev gamma ray of Cs^{137}. Consequently, the human's normal Cs^{137} entire spectrum cannot be subtracted accurately from the subject's gross spectrum thus preventing an accurate measurement of the RaB gamma rays at 241, 295 and 351 kev.

0016911

Theoretically, the interference of the Cs and K gamma rays may be eliminated by analyzing only those gamma rays which are in coincidence (timewise) with other gamma rays. This is the case of the abundant 607-Mev gamma ray emitted by RaC', which is always emitted in coincidence with one of several higher energy gamma rays. Since Cs and K are both monoenergetic emitters, only chance coincidences and those gamma rays which undergo a Compton scatter in one crystal and then are absorbed in the second crystal would produce a count. The scattered gamma-ray coincidence can be eliminated by placing the crystals on opposite sides of the subject so that a gamma ray would have to scatter through 180° (thereby losing all but 250 kev) to reach the second crystal. The pulses resulting from these back scattered photons can be eliminated by adjusting the pulse height analyzers to reject all pulses of less than 500 kev.

The number of coincidences can be measured in the conventional manner whereby the pulses from one crystal are used to gate the analyzer, and only the pulses from the second crystal are analyzed as to height. Obviously, the number of detected events will be doubled if the pulses from either crystal can gate the analyzer and the pulses from the opposite crystal are sorted. Equally obvious is the fact that the pulse from one crystal should not be able to gate the analyzer and be itself sorted and recorded.

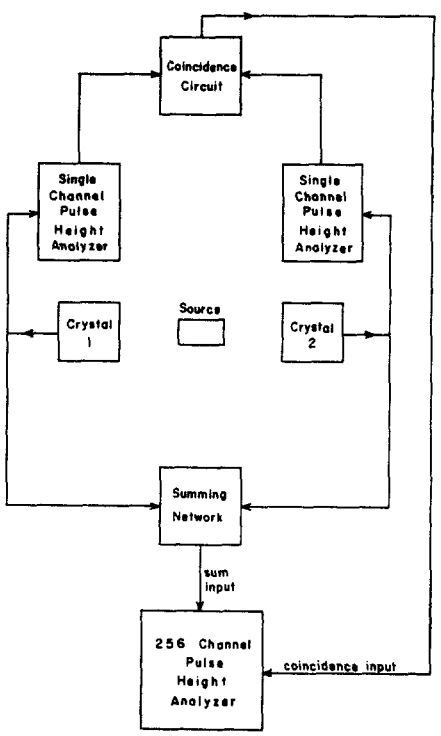

Fig. 26

The electronic equipment which has been used to evaluate the feasibility of this approach is shown in block form by Fig. 26. The pulses from both crystals are fed to a summing network (Σ) and the sum pulse applied to the input of the 256-channel analyzer. In addition, the individual pulses from each crystal are amplified and analyzed by wide-band single channel pulse height analyzers. The output pulses of the single channel analyzers are applied to the coincidence circuit. The coincidence circuit in turn gates on the 256-channel analyzer and permits it to sort the sum pulse whenever pulses are received simultaneously from both single channel analyzers.

Two NaI(Tl) crystals, one $2\frac{1}{2}$ inches in diameter by $2\frac{1}{8}$ inches and one 4 inches in diameter by $1\frac{1}{2}$ inches thick, were used for the preliminary tests. The pulse height discriminators on the single channel analyzers were set to accept any pulse over 500 kev. The upper gate was not used at this time, however, but the possibility of its use should be kept in mind for this would reduce the background since the cosmic ray showers would be removed.

0016912

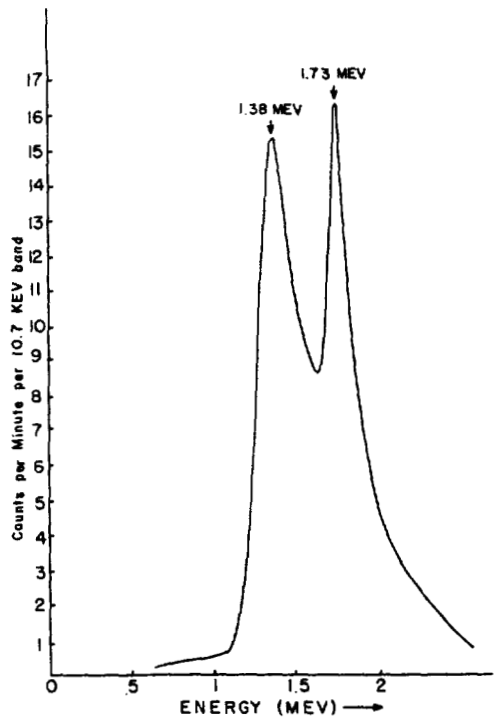

Fig. 27. Sum-Coincidence Spectrum from Ra C'

The spectrum of the sum pulses when 2×10^{-7}c of radium was placed between the two crystals with a 10-cm spacing is shown by Fig. 27. The distinct 1.38-Mev and 1.73-Mev photopeaks are the sums of the 0.607-Mev plus the 0.77-Mev gamma rays and the 0.607-Mev plus the 1.12-Mev gamma rays of RaC'. A less intense coincidence pair which yields a sum peak at 1.54 Mev shifts the high side of the 1.38-Mev peak. Other coincidence pairs at sum energies of 1.85, 2.12 and 2.2 Mev account for the area under the curve above 1.8 Mev.

The gamma-ray background, outside of the iron shield and without the use of an upper limit gate to remove cosmic rays, is low, namely, equal to the counting rate which is obtained from 10^{-9}c of radium. Placing the assembly inside the iron room will reduce background by a factor of 10. An additional reduction will be realized by placing a plastic anticoincidence shield over the assembly to eliminate coincidences due to cosmic rays.

The counting rate from 1300 grams of natural K (ten times the amount present in the total body) placed between the crystals was found to be equal to background. Thus the small amount of K present in a subject's leg will not yield a detectable counting rate.

This system could lead to a more accurate measurement of the RaC' content of the body if sufficiently large crystals are used.

B. Direct Measurement of the Radon Partition Ratio

It is not possible to calculate directly the quantity of Ra^{226} present in the human body using the gamma-ray spectrum of the whole body. The 188-kev gamma ray which is emitted by Ra^{226} in 5.7% of the disintegrations is obscured by the numerous degraded gamma rays from the radium daughters, from the Cs^{137}, and from the K^{40} present in the subject's body. Thus the amount of Ra^{226} present must be determined indirectly by measuring the quantity of the Ra^{226} daughter products.

The body content of RaB and of RaC' can be measured quickly and with high accuracy using gamma-ray techniques. Unfortunately, the body content of Ra^{226} is larger than the body content of RaB or RaC' since a

large fraction of the radon is exhaled. Consequently, an accurate Ra^{226} determination requires, in addition to a measurement of the RaB and RaC', a measurement of either the radon in the breath or a measurement of the Ra^{226}/(RaB RaC') ratio (radon partition ratio).

The addition of a second gamma-ray measurement which would provide the radon partition ratio directly has the obvious advantage that the breath measurements would be unnecessary. The interfering degraded gamma rays from the body which swamp the 188-kev gamma rays from Ra^{226} can be reduced by measuring the spectrum of the kneecap, elbow, shin or ankle with a collimated NaI crystal. These volumes contain a large mass of bone with the least mass of surrounding tissue. The Ra^{226}/(RaB RaC') ratio found for one of these volumes is thus assumed for the total skeleton. The whole body RaB RaC' content is then multiplied by this ratio to obtain the total body Ra^{226} content.

The photopeaks from the 188-kev gamma ray from radium as well as the photopeaks from the 241-, 295-, and 351-kev gamma rays of RaB can be distinguished quite clearly in the spectra obtained from the kneecap and ankle (see Fig. 28). It will be necessary to obtain spectra using appropriate phantoms containing both de-emanated radium and radon in order to peel the spectra from the kneecap and ankle apart to determine the ratio.

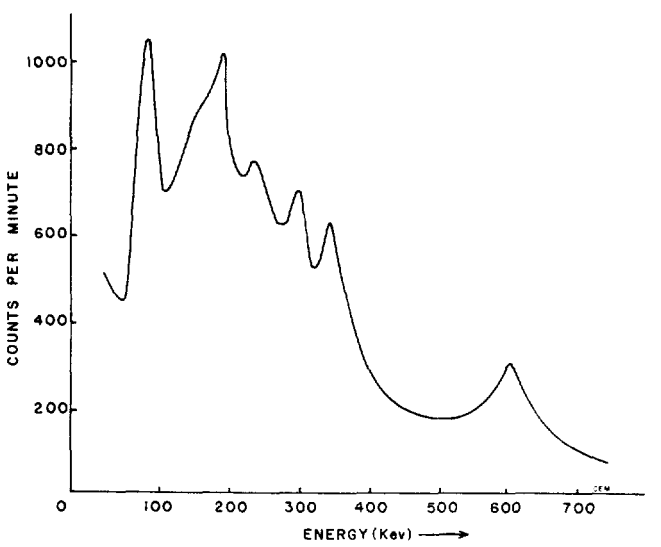

Fig. 28

Gamma ray spectrum of subject EVR's kneecap

A phantom has been constructed using Presdwood to simulate the flesh and using a plaster of Paris bone. Results will be given in a later publication.

IV. Follow-Up of U^{235} Inhalation Incident

C. E. Miller

In a previous report[6] data were given concerning a machinist who had inhaled U^{235} dust during a grinding operation. The quantity present in various volumes of the subject's body had been measured on three occasions during a span of 38 days. The subject was measured a fourth time 98 days after the third measurement. The quantity present in the subject's chest on the four occasions, as well as the quantity eliminated in the excreta between measurements, is given in Table 13. (The excretion measurements were made at the subject's laboratory and are given here with the latter's permission.)

Table 13

Data on U^{235} subject

Date of measurement	Total amount of U^{235} in chest area (mg)	Decrease of U^{235} (mg)	Quantity of U^{235} in excreta during these periods (mg)
November 2, 1957	2.75	0.7	0.74
November 19, 1957	2.05	0.58	0.48
December 10, 1957	1.47	0.77	0.78
March 18, 1958	0.7		
Total		2.05	2.00

The four values do not follow a power function but suggest a short half-time in the lungs of 41.5 days for the first 38 days followed by a longer one. Although the average half-time between the third and fourth measurement is 92 days, this would be a minimal value for the slower phase since the U^{235} may have been eliminated at the higher rate for a period of time after the third measurement. It is now apparent that a measurement 20 days after the third one might have been of considerable value.

The subject's head and feet, which constitute a fair percentage of the subject's skeleton, did not contain a detectable amount of U^{235}. Thus, it is doubtful that the longer half-life is particularly influenced by clearance rates from the bone.

The agreement between the decrease in lung content and the quantity present in the excreta demonstrates that the lung measurements are quantitatively correct if the assumption is made that elimination from other tissues is considerably faster than that taking place in the lungs.

0016915

V. The Elimination Pattern of Co^{60} Acquired by Inhalation

C. E. Miller and H. A. May

Two employees, herein designated A and B, inhaled a small amount of cobalt dust while unpacking a vessel which had become contaminated with Co^{60} dust during a grinding and polishing operation. The vessel had been scrubbed down several times, then wrapped and moved to another area for further cleaning. The employees inhaled the dust which had remained on the vessel and which became airborne as it was unwrapped.

The total body content of subject A was measured on the 2nd, 4th, 7th, 10th, 18th, and 50th day, after the accident. The distribution within subject A's body was measured on the 3rd, 4th, 7th, and 10th day. The total body content of subject B was measured on the 2nd, 4th, and the 24th day after the incident and the distribution within the body was measured only on the 4th day.

Only the data taken on subject A will be discussed since they are, in essence, representative of both cases. Measurements with the standard chair technique and measurements with a collimated crystal were made over various areas of the body, as illustrated in Fig. 29 with the results shown in Table 14.

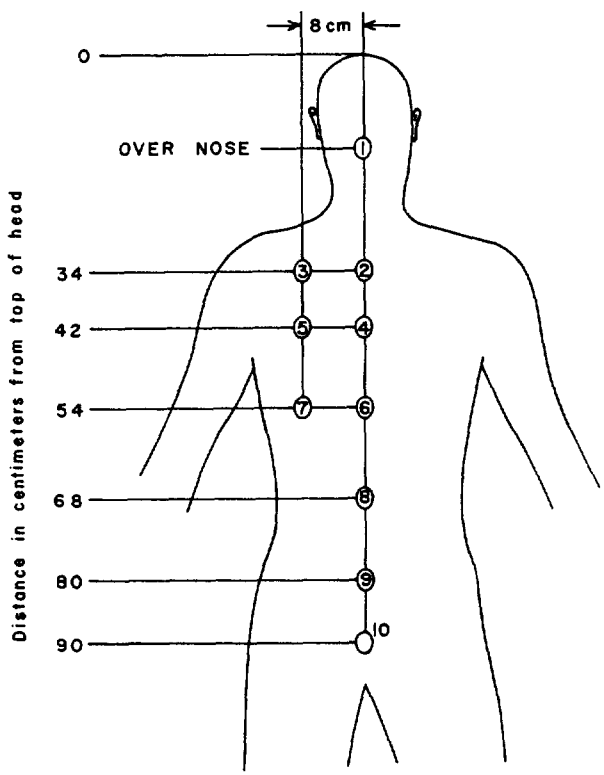

Fig. 29

0016916

Table 14

Co^{60} activities (in arbitrary units) obtained by scanning subject A

Position	1	2	3	4	5	6	7	8	9	10
Date										
6-9-58	-	-	-	Dust inhaled	-	-	-	-	-	-
6-12-58	22	30	23	28	32		42		170	3.5
6-13-58				18		22	26	26	100	
6-16-58	16			4.7		8.4	10	22	6	
6-19-58	10			5.4		7.7	8.7	5		
6-27-58	7			6.5			7.5			

As shown by Fig. 30 the total body content of subject A decreased with a half-time of 1.6 days from the 2nd day through the 7th day and with a half-time of 42 days from the 17th through the 50th day. The content of the chest seems to be decreasing with a half-time of 52 days and the content of the abdomen with a half-time of 62 days.

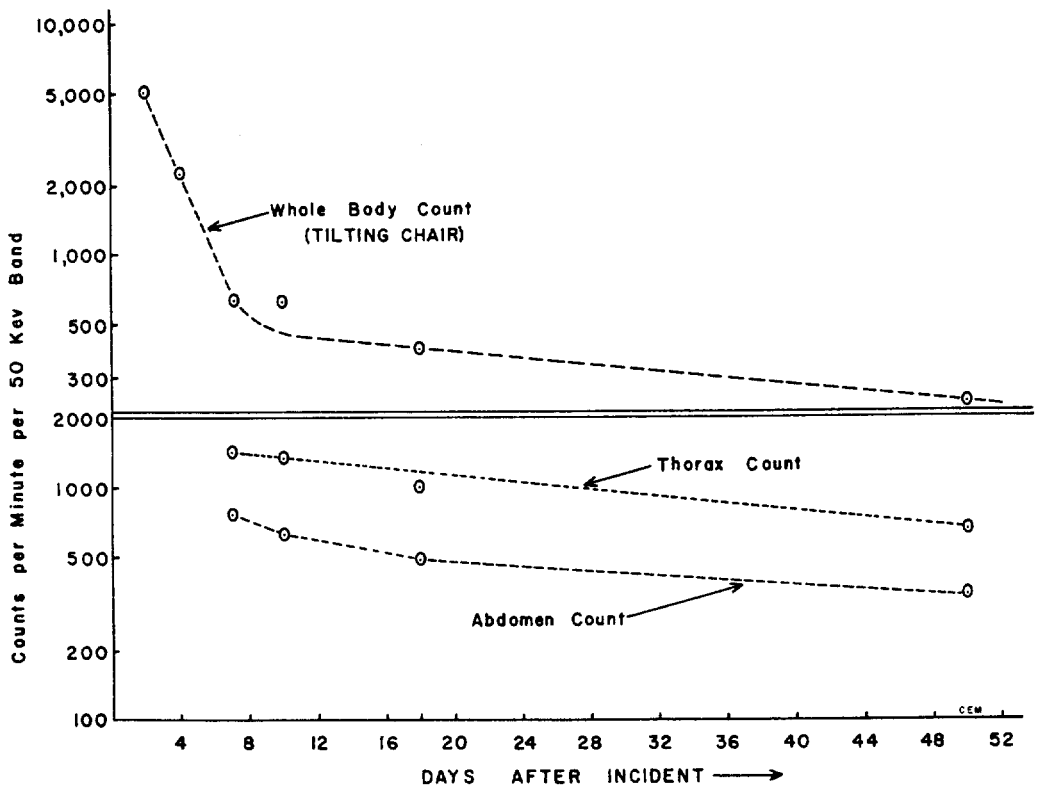

Fig. 30

0016917

The data of Table 14 disclose that the quantity at position No. 4, probably the lymph nodes, decreased until the 7th day and then increased slightly. This phase may account for the longer half-time found over the chest. The 62-day half-time for the abdomen is probably influenced by the presence of the material in the liver.

VI. In Vivo Measurement of Cs^{137}

C. E. Miller and L. D. Marinelli

The radioactive body contents of a group of individuals, living in the vicinity of our Laboratory, have been measured periodically since March 1955, and reported last in ANL-5755. These subjects, members of our Division, do not inhale or ingest any radioactive materials other than that present in ordinary food, water and air. Thus, the radioactivity present in their bodies should be typical of that present in individuals living in the Chicago area.

The Cs^{137} content of 8 subjects, plotted as a function of time since 1955, is shown in Fig. 31. The Cs^{137} content is expressed in units of $\mu\mu c$ Cs^{137} per gram of body K to permit a direct comparison between subjects without regard to body musculature. The Cs^{137} content is seen to have increased from March 1955 until May 1956 at which time it leveled off and remained at essentially a constant value until January 1958. Latest measurements suggest that since then the Cs^{137} content has increased.

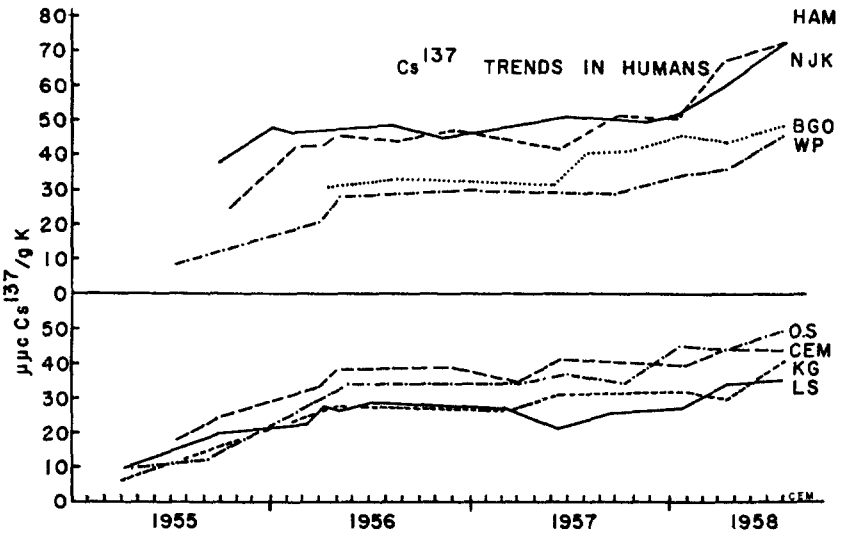

Fig. 31

The gamma-ray spectra of subject HAM obtained on January 16, 1958 and July 28, 1958 (given in Fig. 32) clearly demonstrate the increase in Cs^{137} content in the presence of constant K^{40} content.

0016918

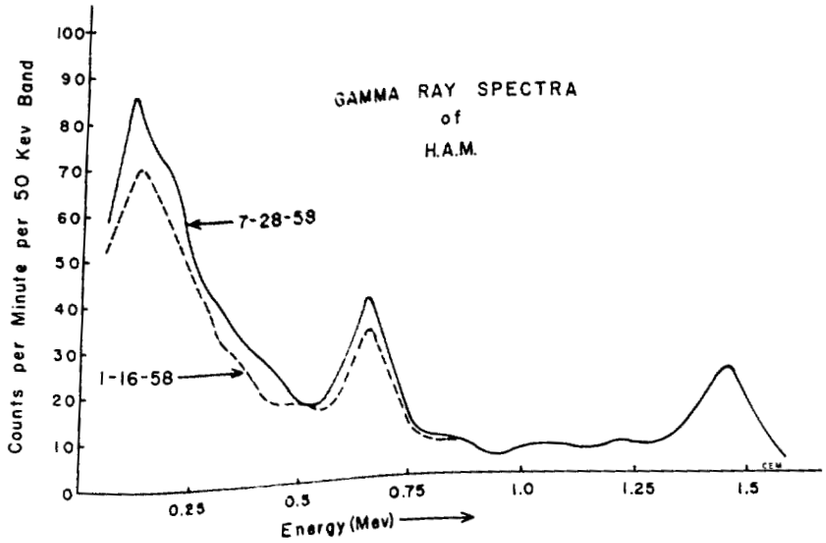

Fig. 32

Details of the Cs^{137} content of 13 subjects on four dates: April 1956, June 1957, April 1958 and July 1958 are given in Table 15. It is obvious that, despite individual fluctuations, the average value has increased from 34$\mu\mu$c Cs^{137}/gK during 1956-1957 to 52$\mu\mu$c Cs^{137}/gK at the present time. Since Cs^{137} has been found to have a biological half-life in man of the order of 150 days, these values should be roughly proportional to the average rate of fallout for the preceding period of 6 months if no abrupt changes have occurred in the dietary habits of the individuals.

Table 15

Cs^{137} in residents of Chicago (42° N)

	Females $\mu\mu$c Cs^{137}/gK					Males $\mu\mu$c Cs^{137}/gK			
	4-56	6-57	4-58	7-58		4-56	6-57	4-58	7-58
K.G.	27	22	33	38	R.R.	21	25	31	39
L.S.	24	31	29	35	W.P.	20	28	35	44
I.S.	24	33	43	52	B.O.	31	31	42	47
C.L.	42	36	51	57	O.S.	30	36	43	48
N.K.	50	50	59	71	H.M.	45	41	65	71
					C.M.	39	41	44	43
					E.M.	-	45	62	64
					J.C.	-	42	55	51
Average	36	34	43	51	Average	33	36	47	52

0016919

References

1. Report on Biological, Medical and Biophysics Programs ANL-5456: 107-123 (1955).

2. Report on Biological, Medical and Biophysics Programs ANL-5518: 45-57 (1956).

3. Radiological Physics Division Semiannual Report ANL-5596: 26-33 (1956).

4. Radiological Physics Division Semiannual Report ANL-5679:22-37 (1957).

5. Radiological Physics Division Semiannual Report ANL-5755:47-61 (1957).

6. Radiological Physics Division Semiannual Report ANL-5829:144-165 (1958).

7. Reynolds, J. C., P. F. Gustafson and L. D. Marinelli. Retention and Elimination of Radium Isotopes Produced by the Decay of Thorium Parents within the Body - Calculations and Comparison with Experimental Findings. ANL-5689 (1957).

0016920