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ARGONNE NATIONAL LABORATORY
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RADIOLOGICAL PHYSICS DIVISION
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THE HUMAN SPECTROMETER

I. Introduction

C. E. Miller

During the past six months, the Human Spectrometer has been devoted largely to the study of certain features of Cs^{137} fallout and its uptake by humans. Thus, the gamma-ray body activities of 11 subjects from Central and South America and of 7 Marshallese have been measured and the trend of cesium burden of our control subjects has been brought up to date. In addition, extensive measurements have been made of the Cs^{137} contents of filter papers collected from an air monitoring program dating back to 1953.

The radium body burdens of two former dial painters have been determined. Studies of the relationship between total and exchangeable K in humans have been started. The gamma-ray spectrum emitted by the liver of a Tridacna clam collected in the Bikini lagoon is given.

The low-energy, gamma-ray background of the Iron Room has been reduced by about 20 per cent with the addition of 1/8 in. of lead to the floor and one wall.

II. In Vivo Measurements of Cs^{137}

C. E. Miller and L. D. Marinelli

Cs^{137} Trends in Humans

The build-up of Cs^{137} as a function of time in four control subjects was reported in graphical form in the preceding Semiannual Report. (1) Because of its obvious importance, all pertinent data from which these curves were plotted have been re-examined in the light of subsequent experience. Figure 18 consists of new curves based on these data incorporating the following changes and additions.

(1) The January, 1956, point on the CEM curve has been dropped. The data had been determined using a 4-in. by 4-in. NaI(Tl) crystal on loan from the Harshaw Chemical Company through the courtesy of E. C. Stewart. This point had been incorrectly plotted since the crystal efficiency factor for the standard 4-in. by 1½ in. crystal was used.

48

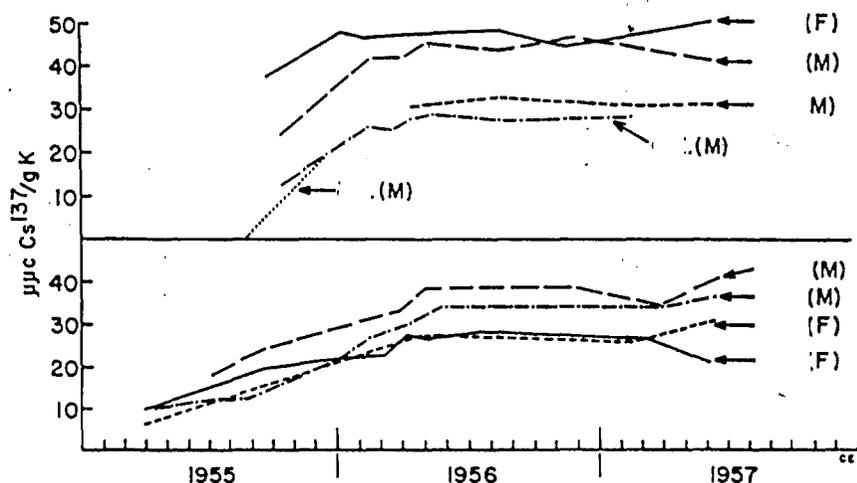


Fig. 18. Cs^{137} trends in humans

(2) The February, 1956, point on the CEM curve has been dropped since an exhaustive examination of the operating log book disclosed that the pulse height analyzer had malfunctioned on other runs during that day.

(3) The June 28, 1955, point on _____ has been corrected for an arithmetical error.

(4) Data taken from April 1, 1955, until July 1, 1955, were not included in the original graph since the subjects had been measured while wearing their own street clothes which may have carried contamination. Since these values are the only existing measurements of Cs^{137} body burdens during this time, the data have been re-examined. In cases where the K body burden has agreed with subsequent values taken under standard conditions, the early data have been assumed to be correct and have been added to the curve.

(5) Curves for five additional subjects have been added. Several runs which gave higher than normal potassium values for female subject _____ were dropped. These high values were discovered to be due to the subject's wearing a foundation garment which had a measurable amount of potassium.

(6) The curves have been extended to June 1957.

49

We conclude that the average Cs¹³⁷ body burdens have remained essentially constant since the spring of 1956. The increase of LS's burden, for example, is offset by a decrease in subject _____ burden. These changes can be ascribed to dietary fluctuations.

The estimate of radioactive contamination of the human body under continual intake of radioactive foods and water would be greatly aided by the establishment of a significant parameter describing the radioactivity of bone. Thus, in the last few years, great efforts have been expended in establishing whether the Sr⁹⁰/Ca ratio in the diet leads to a reliable evaluation of Sr⁹⁰/Ca in the human skeleton. Our first efforts along these lines concerning Cs¹³⁷ led to the speculation as to whether the Cs¹³⁷/K in the diet could be regarded as a reliable parameter for the estimate of Cs¹³⁷ body burden. Contrasted the findings with the Sr⁹⁰/Ca, the Cs¹³⁷/K in the body was found to be 2.2 times the ratio in the urine of a single individual. (ANL-5679, p. 29-30, June-December 1956).

Taking advantage of the constancy of Cs¹³⁷ burden experienced in the last year, the excreta of two of the control subjects (_____ and _____) were collected for several days and analyzed for Cs¹³⁷ and K. The pertinent data obtained are shown in Table 6.

TABLE 6

Subject		
g K/day in urine	2.53	2.0
g K/day in feces	.65	.4
μμc Cs ¹³⁷ /day in urine	33.5	40
μμc Cs ¹³⁷ /g K in urine	13.2	20
ratio $\frac{\text{Cs/K in body}^*}{\text{Cs/K in urine}}$	2.6	1.9

*This ratio has been called assimilation ratio in the body of the paper.

An assimilation ratio of 1.9 was found for subject _____ and 2.6 for subject _____. The average value is 2.2 which is in agreement with the previously reported value for RER. Owing to the low radioactivities due to K⁴⁰ and Cs¹³⁷ in the feces of these subjects and the relatively high content of Ra²²⁶ therein, due to natural intake, it has not been possible to determine the fecal Cs¹³⁷/K by simple gamma ray analysis alone. Efforts to measure this ratio are currently under way. Additional pertinent information is given later in the body of this report, concerning studies on Marshallese subjects of relatively high Cs¹³⁷ content, who, however, could not be considered in equilibrium with their environment at the time of measurement.

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50

World Wide Values

The Cs/K ratios present in the 13 Argonne control subjects (Chicago) are given in Table 7. These values range from 22 to 50 $\mu\mu\text{c Cs}^{137}$ per gram of body potassium with an average value of 36 $\mu\mu\text{c Cs}^{137}/\text{g K}$, irrespective of sex. The spread from the lowest to the highest value yields a ratio of 1 to 2.3.

This ratio was also determined for 11 Central and South American subjects during their visit to our Laboratory. These data are tabulated in Table 8 according to geographical latitude. The values vary from 7 to 21 $\mu\mu\text{c Cs}^{137}/\text{g K}$ with an average value of 14.6 $\mu\mu\text{c Cs}^{137}/\text{g K}$ and yield a spread of 1 to 3.

Since the spread of each group is approximately the same and the dietary influence is pretty well established, the conclusion could be drawn that the subjects with the larger Cs^{137} body burdens were the larger consumers of meat and dairy products as is invariably the case with North American control subjects. The diets of the Argentine subjects, however, include meat and dairy products in quantities wholly comparable to the U.S.A. diets and, moreover, their Cs^{137} body burdens are the lowest of the group. This fact points to the possibility of a real difference in hemispherical fallout.

The values found for people from other areas of the world are given by Table 9. The average for the European subjects is 32 $\mu\mu\text{c Cs}^{137}/\text{g K}$, essentially identical to the Chicago value. The Asian subjects who include little or no milk in their diets are significantly lower.⁽¹⁾

The similarity between the European and Chicago and continental U.S.A. values⁽²⁾ suggests that the fallout in the Northern Hemisphere is approximately uniform.

Although the evidence gathered on a few subjects is certainly not conclusive, it does, nevertheless, focus attention on the various factors affecting the geographical patterns of fallout which have been considered for the case of Sr^{90} .⁽³⁾

The Cs^{137} Atmospheric Content as a Function of Time

In conjunction with our studies of the time trend of Cs^{137} content of humans at Chicago it was thought advisable to inquire into the Cs^{137} content of the atmosphere at the same site. The gross amount of radioactivity present in the air has been measured at Argonne National Laboratory for the past several years. Air pumps located at several different sites at Argonne pull approximately 15 m^3 of air per hour through HV-70 counting filter papers. The filters from some air pumps were changed daily and some were changed weekly. The weekly filter papers were bunched into monthly groups and the gamma-ray spectra of each bundle obtained.

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51

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TABLE 7
CHICAGO SUBJECTS (42°N) DURING JUNE 1957

FEMALES	$\mu\mu\text{c Cs}^{137}/\text{gK}$	MALES	$\mu\mu\text{c Cs}^{137}/\text{gK}$
	22	R.R.	25
	31	W.P.	28
	33	B.O.	31
	36	O.S.	36
	43	H.M.	41
	50	C.M.	41
		E.M.	45
			<u>45</u>
AVERAGE	36	AVERAGE	35

TABLE 8
DATA ON FOREIGN SUBJECTS

COUNTRY	LATITUDE	DATE	$\mu\mu\text{c Cs}^{137}/\text{gK}$
CUBA	22°N	5-23-57	20.0
EL SALVADOR	13°N	"	8.3
COLOMBIA(4,000')	4°N	"	21.0
ECUADOR(10,000')	0°N	"	13.0
BOLIVIA (13,000')	15°S	"	13.0
BRAZIL	20°S	"	16.0
"	30°S	"	21.2
URUGUAY	34°S	"	11.6
ARGENTINA	35°S	"	7.3
"	35°S	"	7.6
CHILE	37°S	"	22.0
		AVERAGE	14.6

TABLE 9

DATA ON FOREIGN SUBJECTS			
COUNTRY	SURJECT	DATE	$\mu\mu\text{c Cs}^{137}/\text{gK}$ $\mu\mu\text{c Zn}^{65}$
EUROPE	ENGLAND	5-16-56	33.5
	"	7-13-56	34.7
	FRANCE	9-21-56	32.7
	BELGIUM	10-30-56	26.5
	SWEDEN	11-29-56	32.2
AUSTRALIA		3-27-57	50.0
ASIA	INDIA	Vb 12-18-56	18.9
	"	Vb "	20.8
	JAPAN	S 7-26-56	24.5
	INDONESIA	S 8-10-56	13.9
OCEANIA	MARSHALL IS.	M 4-5-56	8.5
	"	RC(US)	26.7
	"	TT(US)	31.7
	"	10	65.0
	"	6	69.0
	"	9	73.2
	"	4	79.0
	"	7	95.5
	"	5	1610.0
	"	8	2720.0

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Various amounts of 285-day Ce^{144} - Pr^{144} , 1-year Ru^{106} - Rh^{106} , 27-year Cs^{137} -2.6 m Ba^{137} , and 65-day Zr^{95} - Nb^{95} are present on the filters. The 65-day Zr^{95} - Nb^{95} completely masks the presence of the Cs^{137} on all filters collected since August, 1956. The amounts of Cs present on these latter papers have not been determined to date, although the spectra will be peeled apart when time permits. Unfortunately, all papers collected prior to March, 1953, except for the month of June, 1952, are missing. The amount of Cs^{137} per paper, expressed in arbitrary units, is given by the upper curve of Fig. 19.

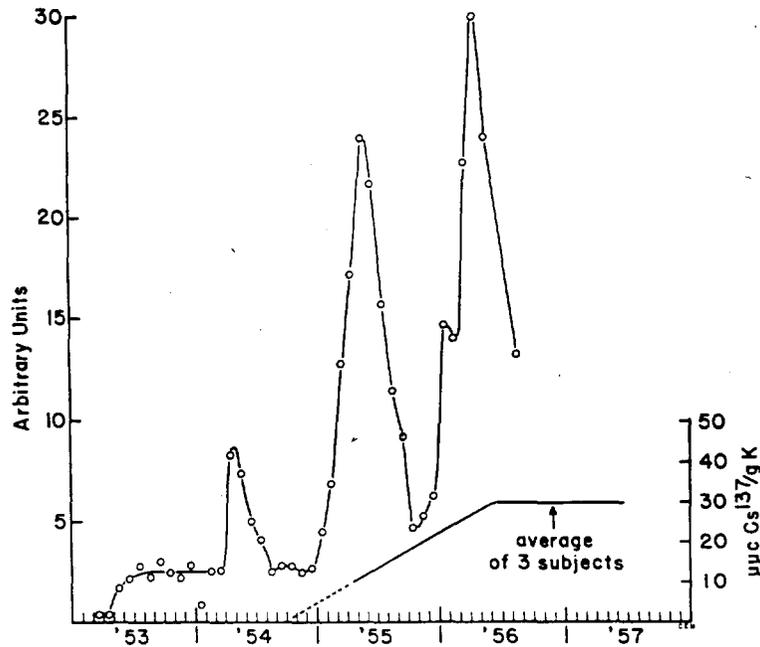


Fig. 19. Cs^{137} activity in filter papers

The Cs^{137} atmospheric contents as shown by Fig. 19, increased by a factor of about 6 in the spring of 1953 and remained at this higher level until January 1955, except for an upward excursion immediately following Operation Castle, March 1954. It is of decided interest that the atmospheric Cs^{137} content, at Chicago at least, did return to its 1953 value in the latter part of 1954. The 1955 excursion is due to the 14 U.S.A. continental tests (Operation Teapot) and the early 1956 excursion is probably due to a combination of the American tests in the Pacific and the Russian tests. The various Russian, English, and American tests have taken place with such regularity since 1955 that the Cs atmospheric content has not returned to its 1953-1954 plateau value. The Cs present on the papers collected since August 1956 is completely masked by Zr^{95} - Nb^{95} from Russian tests during the last half of 1956; efforts will be made to measure it by nondestructive tests.

In view of its possible usefulness, the average shape of 3 cesium trend curves (Subjects and) has been plotted as shown at the lower right hand curve of Fig. 19. It is hoped that upon closer examination of the various factors influencing fallout and its transmittal to man, it will be possible to offer, at some future date, an explanation of the behavior of the lower curve.

III. Measurements on Some Residents of the Marshall Islands

C. E. Miller and O. J. Steingraber

Seven normal, healthy male residents of the Marshall Islands, under the medical supervision of Dr. Robert A. Conard (, Table 9) of the Brookhaven National Laboratory and accompanied by Mr. Jack Tobin (, Table 9) District Anthropologist for the Marshall Islands, were flown to this Laboratory on April 4, 1957 for whole body gamma-ray spectroscopy.

Details on mode and extent of exposure, body content from bioassay and clinical course are described elsewhere.(4,5)

Four of these men (Nos. 4, 7, 6, 9, Table 9), residents of Rongelap, and two others (Nos. 5 and 8), residents of Uterek, were among those evacuated to Kwajelein from the area of heavy fallout some 48 to 78 hours following the thermonuclear detonation of March, 1954. About 3 months later, Nos. 5 and 8 were returned to Uterek and the others moved to Majuro. The seventh man (No. 10) from Majuro, not exposed, served as a control although his diet was not typical since he worked at Trust Headquarters as did Mr. Tobin (No. 11, Fig. 20).

Each subject's gamma-ray spectrum (see Fig. 20) was obtained using the standard chair position. Since the body burdens of the two Utere subjects (Nos. 5 and 8) were relatively high, their gamma-ray spectra could be accurately measured utilizing the one-meter arc technique. The body burden of subjects 5 and 8 was then calculated using the one-meter arc technique described in an earlier report.(1) The one-meter arc net $K^{40} + Cs^{137}$ spectrum of (previously determined) was subtracted from the spectrum of Subject 5, whereupon the curve labeled " K^{40} subtracted" in Fig. 21 was obtained representing the Zn^{65} and "excess" Cs^{137} activity. A Zn^{65} phantom spectrum and a Cs^{137} phantom spectrum were obtained using presdwood phantoms of appropriate thickness. The Zn^{65} phantom spectrum was normalized so that the area under the Zn^{65} photopeak matched the area under the subject's Zn^{65} photopeak and the normalized spectrum subtracted. This yielded the curve labeled " $0.48 \mu c Zn^{65}$ subtracted" in Fig. 21. The Cs^{137} phantom spectrum was then normalized to match the area under the subject's remaining Cs photopeak. This normalized Cs^{137} spectrum was subtracted from the above resultant spectrum and the residual curve label

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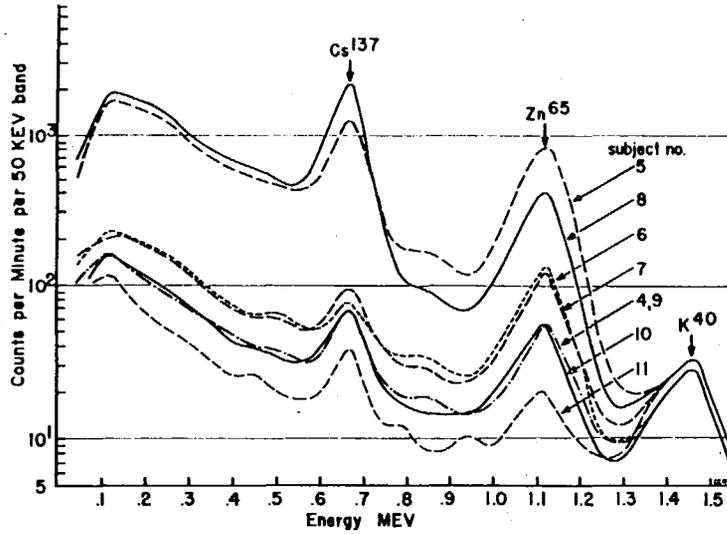


Fig. 20. Net in vivo gamma-ray spectra of Marshallese

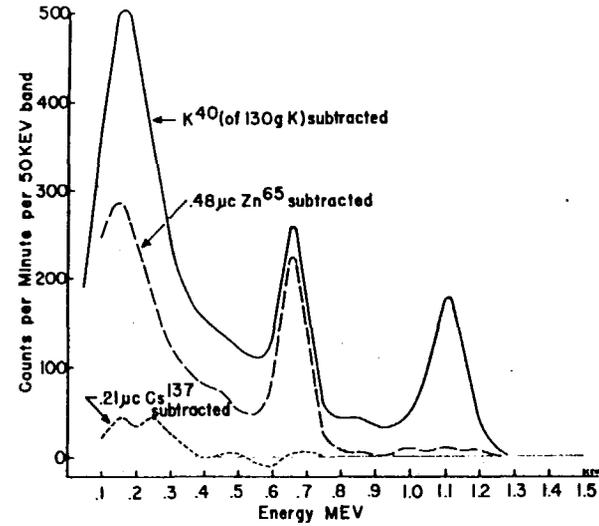


Fig. 21. Analysis of gamma-ray spectrum of Marshallese Subject No. 5

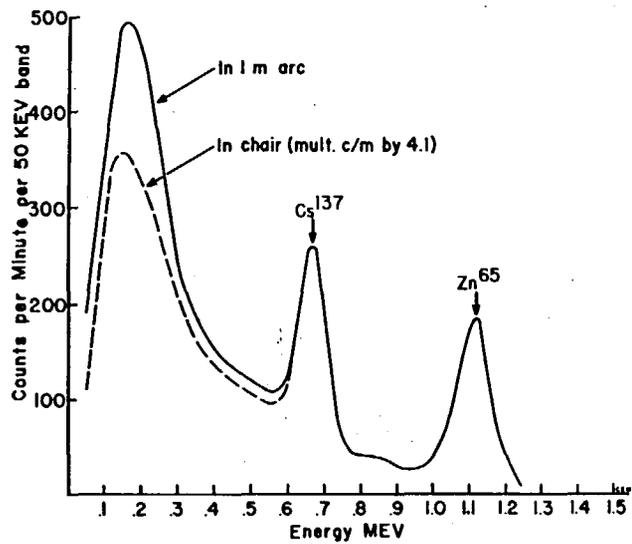


Fig. 22. Net in vivo gamma-ray spectra of Marshallese Subject No. 5

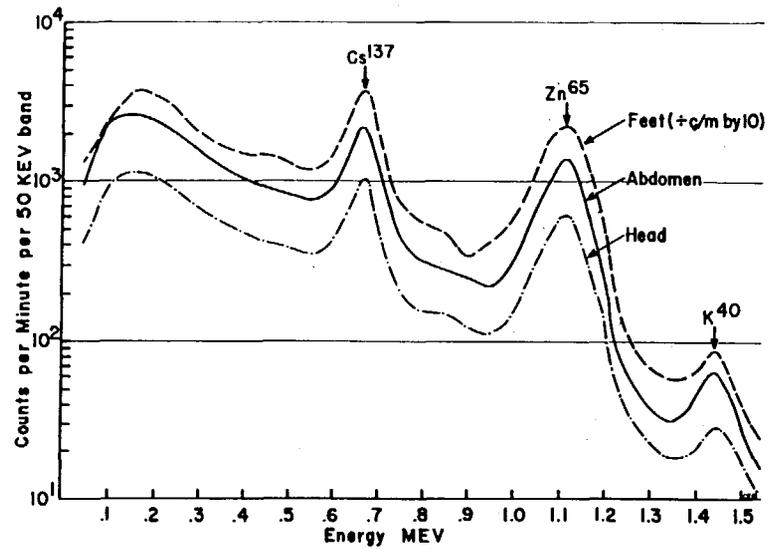


Fig. 23. Net gamma-ray spectra of Marshallese Subject No. 5

55

"0.21 μc Cs^{137} subtracted" was obtained. This residual spectrum below .4 Mev is undoubtedly due partly to the fact that the Cs and Zn phantom spectra are not exactly identical to the in vivo Cs and Zn spectra. It is felt, however, that the burdens of two subjects calculated in this fashion are correct to at least 5 per cent.

Calculating the body burdens of the five subjects who had been measured only in the standard chair presented a problem. The standard chair position cannot be calibrated with a source and a presdwood phantom since all volumes of the body are not at the same distance from the crystal, a condition which is assumed when a person is placed on the one-meter arc

Since administration of the pertinent isotopes to the subject was deemed unadvisable, the standard chair position was cross calibrated to the one-meter arc position using the gamma-ray spectra of Subjects 5 and 8 in the following manner. The appropriate K gamma spectrum of a control subject of the same height and weight was subtracted from the gamma-ray spectrum obtained with Subject 5 in the chair and on the arc. The resulting net in vivo Zn^{65} and Cs^{137} spectrum, from 600 kev to 1.3 me obtained with the chair position was 4.1 times the net in vivo Zn^{65} and Cs^{137} spectrum obtained on the arc. (See Fig. 22) The same ratio of 4.1 was found for the spectra of Subject 8.

The standard chair gamma-ray spectra of the five subjects were analyzed using the identical peel-off procedure discussed above for the one-meter arc technique above and demonstrated with Fig. 21 except that the part of the spectra falling below 600 kev was neglected and the chair to one-meter arc ratio was taken into account.

The values calculated for each subject were given by Table 9 above

To investigate the gross distribution of Zn^{65} in the body, gamma-ray spectra were obtained of Subject 5's feet, abdomen, and head by merely placing the crystal near the sites in question. The results, shown in Fig. demonstrate that $\text{Zn}^{65}/\text{Cs}^{137}/\text{K}^{40}$ ratios do not vary widely.

In order to determine the effective biological half-life of the isotope involved, the Marshallese were supplied with suitable containers and requested to submit their excreta. However, due to sight seeing trips, reporters, newsreel people and other extraneous activities, some samples were lost, and no samples could be regarded as representative of excretion rate over a definite period of time.

In order to gather at least some approximate information the samples were treated as follows: the K, Cs^{137} , and Zn^{65} content of each specimen was measured and the Cs^{137} and Zn^{65} values were expressed in terms of per gram of K. Since the Marshallese were on a standard American diet

about 10 days, they were assumed to be in equilibrium with our diet and to eliminate (as the average of our control group) 2 grams of K per day in the urine and 0.5 gram per day in the feces. Knowing the K contents of the samples collected, this assumption permits a not-too-far-fetched extrapolation to the daily excretion rate.

The Zn^{65} data are given in Table 10. An average of $6.7 \mu\mu c$ of Zn^{65} per $m\mu c$ of Zn^{65} present in the total body is eliminated with each gram of K in the feces. The Zn^{65} amount present in the urine is negligible. On the basis of 0.5 gram of K eliminated per day by the fecal route, $3.4 \mu\mu c$ Zn^{65} per $m\mu c$ Zn^{65} in the body would be eliminated per day. The biological half-time for the element is thus 200 days. An effective biological half-time of 110 days results by adding the effect of the physical half-life of 245 days. This is considerably longer than the published values.

TABLE 10

Subject	Feces	Urine	Body burden	$\mu\mu c$ Zn^{65} per gram K in feces
	$\frac{\mu\mu c Zn^{65}}{g K}$	$\frac{\mu\mu c Zn^{65}}{g K}$	$\frac{m\mu c Zn^{65}}$	$\frac{m\mu c Zn^{65}}{\text{in body}}$
4	no data	-	29.5	-
5	3340	82	482	7
6	536	-	73	7.4
7	236	-	62.1	3.8
8	1340	42	229	5.9
9	183	-	29.5	6.3
10	280	-	29.5	9.7
				average 6.7

The Cs^{137} data on two subjects are given in Table 11; they similarly are based on the American average of 2 grams of K eliminated in the urine per day and 0.5 gram of K in the feces per day. The effective biological half-time was 92.4 days for Subject 5 and 157 days for Subject 8. The average of these values is in essential agreement with values found under controlled conditions.

TABLE 11

Subject	Feces $\frac{\mu\mu\text{c Cs}^{137}}{\text{g K}}$	Urine $\frac{\mu\mu\text{c Cs}^{137}}{\text{g K}}$	Body burden μc	Daily excretion $2 \times (\text{urine}) + 0.5 (\text{feces})$ $\mu\mu\text{c Cs}^{137}$
5	825	575	0.22	412 + 1,150 = 1,562
8	1040	638	0.41	520 + 1,276 = 1,796

IV. Exchangeable Potassium in Humans

C. E. Miller

The measurements of exchangeable potassium in humans are based on the following simplified model. The normal K present in humans is considered to be present in two fractions or pools. The atoms of K in one pool are bound within the cells and are not immediately replaced by other atoms of K from the blood stream. The atoms of K in the second pool are exchangeable and are constantly being replaced by new atoms of K. The K present in the urine has thus originated from the exchangeable K pool.

The exchangeable pool is measured in the following fashion. Each subject is given 5 μc of K^{42} at 9:00 A.M. All urine voided from 9:00 A.M. until 10:00 P.M. is collected in one vessel and the amount of K^{42} eliminated is recorded. The first void the next morning is collected in a second container, and all urine voided thereafter until the subject is measured is collected in a third container. The amount of K^{42} present in the body at any time is thus known. All K^{42} has been mixed into the pool of exchangeable K by the definition of exchangeable K. The ratio of K^{42} to normal K present in the urine the following morning should equal the ratio of K^{42} to normal K present in this so-called exchangeable pool. The amount of K^{42} present in the urine determined by gamma-ray analysis, and the amount of normal K present is measured with a flame photometer. The amount of K present in the exchangeable pool is found by multiplying the K^{42} present in the body by the ratio:

$$\frac{\text{normal K in the urine}}{\text{K}^{42} \text{ in the urine}}$$

This value of K is then compared with the total body potassium as measured by the in vivo gamma-ray techniques.

On the basis of repeated tests performed on five subjects it can be said that the exchangeable potassium is of the order of 90 per cent or more of the total potassium. It seems certain, however, that because of either

instrumentation irreproducibility or more likely, unsuspected diuretic factors, the measured specific K^{42} activity in the urine is likely to vary considerably in the same individual. It is felt, therefore, that greater accuracy in the determination of the exchangeable fraction must await clarification of the apparent irreproducibility of the specific K^{42} activity of the urine.

V. Radium in Humans

C. E. Miller

There is seemingly a tendency to relate the radium burdens of former dial painters to the length of their employment in the radium dial industry. Information elicited during conversations with some of these former dial painters indicates that such a relationship does not exist.

One individual disclosed that some of the girls painted their fingernails, eyebrows, and even streaks in their hair with radium dial paint before going out on social dates. A second subject stated that the painters who taught new employees would eat a quantity of radium dial paint from a spatula to convince them that the material was harmless.

Considering such practices, it should not be considered unlikely to find an individual with a current burden of $2 \mu c$ of Ra^{226} as a consequence of an employment period of only three months (prior to 1930) in the radium dial painting industry. To our collection of cases containing Ra^{226} as a result of either employment or therapy two subjects have been added bearing 2.45 and 1.15 μg respectively.

VI. Radioactivity in the Liver of a Tridacna Clam

C. E. Miller

On June 30, 1956, Dr. Theodore R. Folsom of the Scripps Institute of Oceanography, La Jolla, California, sent two samples from the liver of a Tridacna (killer) clam to this Laboratory for gamma-ray analysis. This clam had been collected May 18, 1956, prior to the tests of that year in the Bikini Lagoon, 1000 feet northwest of the shore of Bikini Island at a depth of 10 feet. The weight of the whole clam meat was estimated at 1.8 to 2.0 kg wet weight. The weight of the part saved for assay (commonly called the liver) was 171 grams wet weight and 29 grams dry weight, of which two samples weighing 5 and 7 grams were used for measurement.

The gamma ray-scintillation spectra obtained from the 7-gram sample is illustrated in Fig. 24 wherein the top curve is the spectrum obtained from the gross sample. The low energy photopeak was identified as Co^{57} and