

## Radioactivity of People and Foods

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The problems of widespread, low-level radioactive contamination from nuclear weapons testing have been increasingly before the public during the past year. The principal concern is the fallout and entry into the biosphere of strontium-90. There is general agreement that present levels of strontium-90 in foodstuffs and in the human body are far below the most conservative permissible amounts; however, the human burden of strontium-90 may be expected to rise as a result of deposition of stratospheric debris from weapons already (and subsequently to be) tested. Predictions based on conservative assumptions indicate that there remains a considerable margin of safety. If the rate of weapons testing continues to increase, however, this margin may eventually disappear.

Although the permissible levels contain inherent safety factors, it is essential that close attention be devoted to all aspects of the fallout problem during the next several years. Only in this way can advance notice of the possible approach to permissible levels be obtained and assurance given that they will not be exceeded inadvertently. Recent reports of the National Academy of Sciences-National Research Council Committee on the Biological Effects of Atomic Radiation (1) support the importance of systematic measurements of general levels of radioactivity in order that information on the rate of accumulation of extraneous radioactivities may be obtained while the latter are still below natural levels.

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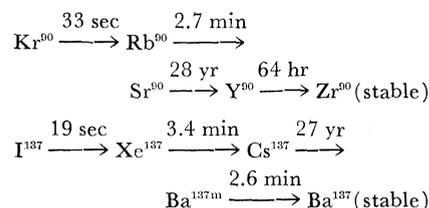
Large-scale production of nuclear power will create problems of a similar nature. A 100-megawatt (heat) reactor will, in one year of operation, produce the same quantity of long-lived fission products as the detonation of a 1-megaton fission bomb. The estimate of the U.S. nuclear power production rate by 1975 is 20,000 to 40,000 megawatts, and the United Kingdom expects to be producing 6000 megawatts by 1965. Reactor-produced fission products constitute a much less immediate problem than those from a bomb test, since more control can be exercised over their immediate fate, but disposal of the fission products must eventually be made.

If disposal is to be simple enough to make nuclear power economically competitive, dispersal by natural means such as ocean burial or other means may have to be resorted to. This will increase the possibility that reactor-produced fission products may ultimately enter the food cycle and reach man. The basic problems of permissible body burdens and distribution mechanisms in the biosphere, therefore, are similar for bomb and reactor debris, and information gathered in the study of the former problems should prove valuable in the latter.

An extensive survey of strontium-90 levels (Project Sunshine) has been underway for several years, and the results have been reported by Libby (2-4) and by Kulp (5). Because strontium-90 and its daughter yttrium-90 emit only beta rays, analysis requires time-consuming and destructive chemical separations. Detailed studies of the temporal and spatial distribution of long-range fallout would be easier if they could be based on a gamma-emitting nuclide. The discovery of the presence of the fission

product cesium-137 in human beings and in foodstuffs by Miller and Marinelli (6) provides a possibility of such an approach.

Similarity of the decay chains of the fission products of mass 90 and mass 137 indicates that distribution of cesium-137 and strontium-90 in bomb debris will be similar:



Both nuclides have two gaseous or volatile predecessors with appreciable half-lives. Strontium-90 and cesium-137 are formed at relatively late times after bomb detonation and are not proportionally included in the larger and more refractory particles which fall out locally. Stratospheric storage and distant deposition will be high for both nuclides, and their ratio in distant fallout should be approximately that calculated from the known fission yields. Once strontium-90 enters the biosphere, its behavior becomes very complex. Its concentrations along the ecologic chain change slowly and reflect a summation of all past fallout. In addition, it enters plants both through the soil (in some relationship with available calcium) and by foliate absorption from direct fallout.

One very important and difficult problem is to determine the fraction of strontium-90 entering the ecologic chain by way of these routes. Cesium-137, however, is apparently poorly taken up from the soil by plants (7) and its biological half-times (8) are comparatively short [140 days in man (9) and 20 days in the cow (10)]. These factors suggest that cesium in people and in milk and other foodstuffs may be a direct and relatively simple measure of fallout rate. One should be able, therefore, to make a direct determination of fallout rate as a function of geographic location and time, as well as of changes in stratospheric storage following test operations, by measuring cesium-137 in biological materials. Cesium-137 measurements on soils might provide a more convenient method than strontium-90 measurements for estimating integrated fallout.

Cesium-137 and strontium-90 also are similar in that they are soluble and closely related to potassium and calcium, respectively, which are normal base exchange cations in soil and essential constituents of living matter. In this they differ from other high-yield fission products such as zirconium-niobium-95, ruthenium-rhodium-106, and cerium-144, which have been observed in rug dirt by Miller and Marinelli at Argonne National Laboratory (11) but which are apparently not ecologically concentrated and have not been detected in the general population and in foodstuffs.

## Potassium-40 and Cesium-137 in

### People and Foodstuffs

After the announcement by Miller and Marinelli of the presence of cesium-137 in people (6), an intensive program of study of this nuclide in people and in foodstuffs was begun at the Los Alamos Scientific Laboratory. Some 1500 measurements were made; preliminary results have been reported previously (12). This article (13) summarizes the data collected during 1956. A compilation of all the primary data is being prepared as an unclassified laboratory report which will include detailed analyses of procedures, sources of error, and other information.

Measurements were made with the Los Alamos "human counter" (14), a large liquid scintillation detector that is capable of counting gamma rays from human subjects and from samples of foodstuffs up to several hundred pounds

in weight with 100 percent geometrical efficiency. Although the energy resolution of this detector is quite limited compared with that of a sodium iodide (thallium) crystal, it is adequate to permit the simultaneous determination of the cesium-137 (0.661 Mev) and potassium-40 (1.46 Mev) gamma rays. Its ultimate sensitivity is 0.0005 microcurie of gamma activity (20 disintegrations per second) for only 100 seconds of counting time. If a 100-kilogram sample is counted, this corresponds to a specific activity of  $5 \times 10^{-15}$  curies per gram, which is far below the natural radioactivity of most materials. The natural potassium-40 radioactivity of man (about 0.013 microcurie as gamma rays) can be measured to a precision of better than 5 percent in less than 2 minutes. The cesium-137 determination has a precision of 0.001 microcurie for the same counting time.

*Potassium-40 in people.* The average potassium content of the adult male is estimated to be about 133 grams (6, 15, 16) (0.19 percent of gross body weight of the standard man), which is equivalent to about 400 potassium-40 gamma disintegrations per second.

Figure 1 gives the natural potassium-40 gamma activity of 164 representative subjects, 81 of which were reported earlier (17), plotted against gross body weight. These data show a pronounced scatter of the points to the right of a limiting line and a definite difference between males and females. Correlation of potassium-40 activity with the fat-free body weight of a select group of these subjects indicated the amount of fat to

be the principal factor causing variation in apparent potassium content of the body (17). The total body potassium expressed as percentage of gross body weight will show considerable variation, therefore, depending on sex, age, weight, body type and physical activity, but it can be accurately calculated from a determination of total body water.

In Fig. 2 the specific activity of potassium-40 (gamma disintegrations per second and pound) is plotted against subject age. These data confirm the general decrease of potassium with age reported by Sievert (16). The solid lines indicate the probable upper limits for uncontaminated male and female subjects, respectively. Not enough children have been measured for us to be certain of the trend below age 15. The dashed lines, therefore, are estimates over this region. Deviation from these curves is an indication of possible surface contamination of individuals during periods of local fallout, since only 0.002 microcurie of hard gamma contamination is sufficient to raise the average adult from the lower to the upper limit of the specific activity distribution.

*Cesium-137 in people.* Libby (2) adopted the procedure of reporting strontium-90 results as strontium-90/calcium ratios because of the metabolic similarity of strontium and calcium and to facilitate the comparison of different types of materials. Our cesium-137 results are reported as cesium-137/potassium-40 ratios for similar reasons. The principal differences in the biological behavior of the two elements can be accounted for in terms of the appropriate biological half-times. The ratios are reported as the ratio of cesium-137/potassium-40 gamma disintegrations (18).

Figure 3 summarizes the measurements of cesium-137/potassium-40 ratios in people for 1956. The triangles represent results in people from various parts of the United States (the distribution is indicated on the map, Fig. 4). Each point is an average for 10 to 20 persons, and the range of values before averaging was 0.1 to 0.9. The circles are averages of measurements on a local control group of 10 laboratory personnel. The scattered high values during the period from June to September are probably the result of surface contamination from tropospheric fallout during Operation Redwing. That they were caused by surface contamination was indicated by their sudden rise and fall, by abnormally high apparent potassium-40 values during the same period, and by the fact that these high apparent potassium-40 values were reduced to normal after bathing in those cases in which remeasurement was possible.

Because of this evidence of external contamination, a line through the more reproducible lower limit of the distri-

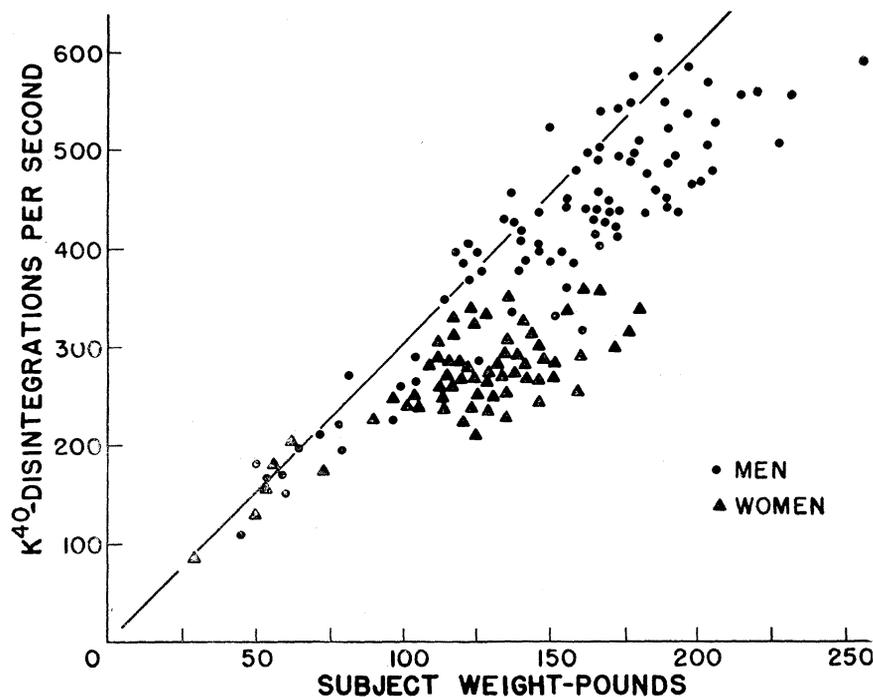


Fig. 1. Potassium-40 gamma activity in people as a function of gross body weight.

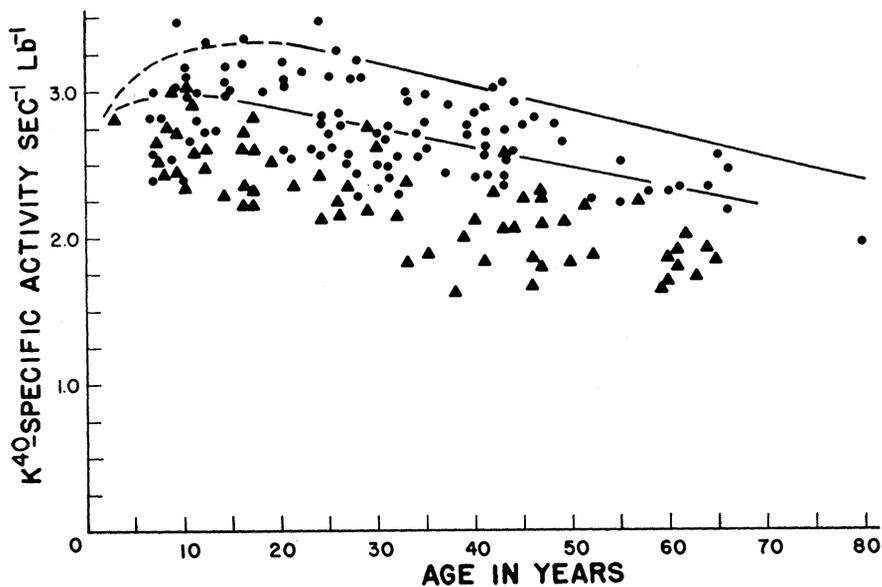


Fig. 2. Potassium-40 specific activity in people as a function of age.

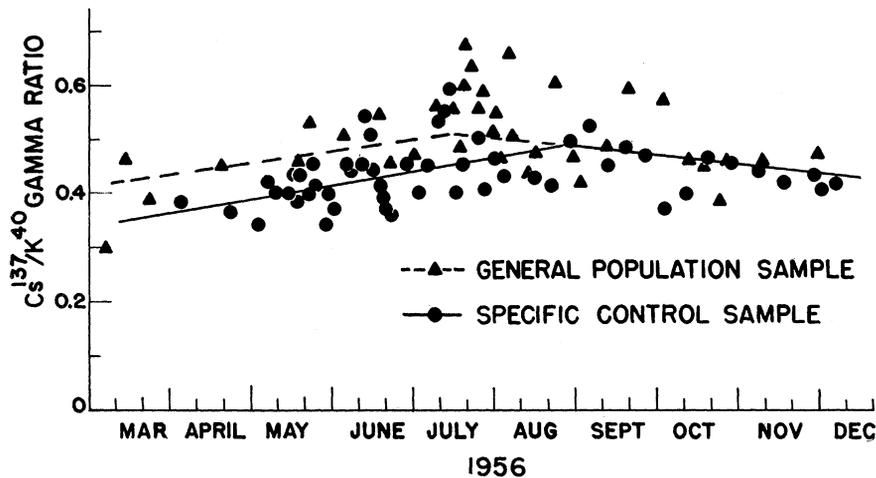


Fig. 3. Cesium-137/potassium-40 gamma ratio in people during 1956.

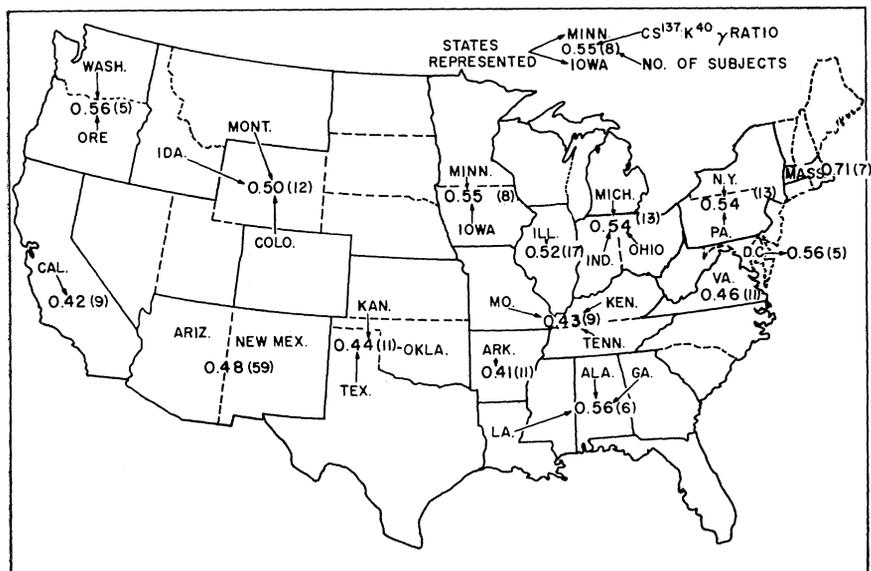


Fig. 4. Geographic distribution of cesium/potassium ratios in people.

tribution is regarded as representing the trend of internal activity. The data from the two groups agree in that they indicate a slight rise during the spring followed by a slow decline during the fall. The control group was apparently somewhat lower in the spring, but in the fall the two groups were indistinguishable.

General 1956 averages of cesium-137/potassium-40 ratios for people from various states are presented in Fig. 4. The results are surprisingly uniform in view of the sizable variations among individuals from the same state. Uncertainty in the averages due to small sample size precludes any deduction of fine structure until more data are available. Within the range  $0.5 \pm 0.2$ , the cesium-137/potassium-40 ratio is essentially uniform over the United States, except during periods of tropospheric fallout.

The frequency distribution of potassium-40 and cesium-137 in the population sample is essentially normal. The potassium-40 frequency curve is given in Fig. 5 as a histogram with a normal error curve fitted to it. The standard deviation of the normal curve is 18 percent. The subsidiary peak outside the normal curve is caused by surface contamination during periods of tropospheric fallout.

Figure 6 shows the corresponding frequency curve of the cesium-137 data for the same population sample. Distribution is again normal, but the width is twice as great as that of potassium-40, the standard deviation being 36 percent. The smaller deviation of the potassium-40 data probably reflects control of the potassium-40 level of the body by a homeostatic mechanism that is not highly dependent on intake. The cesium-137 burden, however, may vary with the dietary habits of the subject and the concentration of cesium-137 in his foodstuffs.

Libby (19) has shown that other trace elements, such as stable strontium, strontium-90 and radium-226, show normal frequency distribution curves with deviations comparable to that observed for cesium-137.

The abnormal subsidiary peak shown in the potassium-40 distribution curve is not present in the cesium data. This indicates merely that the surface contamination distorting the potassium-40 level was present in the cesium-137 channel to a proportional extent and left the cesium/potassium ratio unaffected.

Cesium-137 in milk and other foodstuffs. Figure 7 summarizes the measurements of cesium-137/potassium-40 ratios in milk during 1956. A peak in cesium-137 activity during July, presumably owing to tropospheric fallout from Operation Redwing, is clearly visible in Wisconsin and New Mexico samples, but is absent from Kentucky milk. This observation is consistent with the path of the cloud as estimated in the U.S. Public Health Service air sampling network.

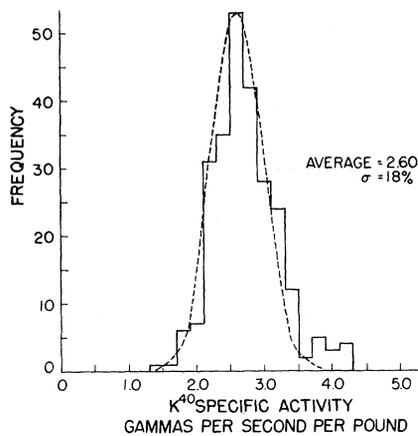


Fig. 5. Frequency distribution of potassium-40 specific activity.

A peak in the activity in Wisconsin milk in October is indicated also; it may be the result of a foreign test.

Data on geographic distribution of the cesium-137/potassium-40 ratio in milk are as yet scanty, but are summarized in Fig. 8. As with the measurements of people, one concludes that distribution is essentially uniform within the limits of the data. The uniformity, of course, applies only to the periods in which tropospheric clouds are not present. It is interesting that the two Australian milk samples (Fig. 7) are in agreement with the general U.S. average, lending support to the assumption that the general levels are derived from the stratospheric reservoir. A sample of American dry milk produced in 1942 showed no detectable cesium-137, the cesium-137/potassium-40 ratio being less than 0.02.

Some preliminary measurements of cesium-137 in foodstuffs other than milk are given in Table 1. During the spring of 1956, beef and lamb showed a ratio comparable to that of people but considerably higher than similar samples collected in the winter of 1956-57. Dur-

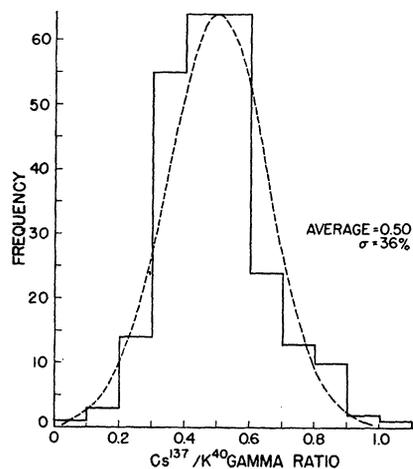


Fig. 6. Frequency distribution of cesium/potassium ratio.

ing both periods, beef and lamb consistently ran higher than pork, which might be expected from the differences in grazing and feeding habits. One sample of dried blood collected in April 1952 showed a ratio less than one-third that of samples collected during the winter of 1956-57.

### Discussion

Measurements of present levels of cesium-137 in people indicate that it is of little significance in the potential hazard of radioactive fallout from weapons testing programs. The present average cesium-137/potassium-40 total disintegration ratio is about 0.05. Taking into consideration their respective energies, the radiation dose from present levels of cesium-137 is only one-twentieth of that from natural potassium-40, or about 1 milliroentgen per year. This is about 1 percent of the average total natural radiation dose and less than  $10^{-3}$  of the dose of cesium-137 given in the Recommendations of the International Commission for Radiological Protection as the maximum permissible level for the general population (20). Interest in cesium-137, therefore, centers on its potential usefulness in the study of fallout mechanisms.

A rough quantitative comparison of the present average strontium-90 and cesium-137 levels in people is of interest. According to Libby (3), the strontium-90 level in children is about 0.001 microcurie. A fractionation factor of about 10 against strontium between primary fallout and human bone is indicated by the stable strontium data (21) (that is to say, the strontium/calcium ratio in soil is 10 times the strontium/calcium ratio in bone), but cesium can be assumed to be quantitatively absorbed by both cow and man. Although strontium will continue to accumulate because of its long biological half-time, the effective accumulation time for cesium will be limited to some 200 days. If stratospheric fallout is assumed to have begun with Operation Castle (1954), strontium-90 has been accumulating for some 2 years, and this factor will cause it to exceed cesium-137 by  $2 \times 365/200$ , or 3.6. Finally, the relative activity yield in the fission process is 1.27 in favor of cesium (assuming fission yields of 0.0510 and 0.0620 for the mass-90 and mass-137 chains, and half-lives of 27.7 years for strontium-90 and 26.6 years for cesium-137). The over-all factor is then  $10 \times 1.27/3.6$ , or about 3 for cesium-137, and the estimated level based on a strontium level of 0.001 microcurie is 0.003 microcurie. Considering the crudity of the several approximations, this is in surprisingly good agreement with the observed average of 0.005 microcurie.

Table 1. Radioactivity in foodstuffs.

Sample	K <sup>40</sup> specific activity (disintegration/sec lb)	Cs <sup>137</sup> /K <sup>40</sup> ratio
<i>Meat, spring 1956</i>		
Beef rounds	3.84	0.53
Lamb, dressed carcass	3.83	0.81
Pork, fresh hams	3.52	0.30
Pork, loins	3.23	0.19
<i>Meat, winter 1956-57</i>		
Beef, sirloins	2.75	0.15
Lamb, dressed carcass	3.67	0.16
Pork, loins	3.55	0.10
Pork, loins	3.26	0.07
<i>Dried blood</i>		
Illinois, Apr. 1952	9.2	< 0.07
California, winter 1956-57	7.3	0.25
Minnesota, winter 1956-57	5.4	0.25
Texas, winter 1956-57	5.9	0.18
<i>Flour, spring 1956</i>		
High-altitude wheat (Colorado)	1.30	0.09
Bleached, enriched (A)	1.70	0.32
Bleached, enriched (B)	1.49	0.27
Whole wheat, graham	7.00	0.11
<i>Potatoes, spring 1956</i>		
Colorado	7.82	< 0.06
Idaho	6.52	< 0.06
<i>Vegetables, spring 1956</i>		
Lettuce	2.34	< 0.07
Cabbage	3.20	0.12
Carrots	6.82	< 0.03
<i>Fruits, spring 1956</i>		
Tomatoes	3.81	0.03
Oranges	2.10	0.38
Grapefruit	3.30	0.25
Watermelon	3.75	< 0.03
<i>Coffee, spring 1956</i>		
	30.00	< 0.06

Table 2. Calculated cesium-137 intake based on per capita food consumption. Diet was based on "Consumption of food in the United States, Supplement for 1954 (22); N.D., not detected.

Source	Consumption (lb/mo)	Cs <sup>137</sup> concn. (mμc/100 lb)	Cs <sup>137</sup> intake (mμc/mo)
<i>Dairy products</i>			
(as dry-milk solids)	5.8	14	0.81
Meats	11.4	3.3	0.38
<i>Flour and cereal products</i>			
Vegetables	13.0	1.0	0.13
Citrus fruits	16.8	N.D.	?
Potatoes	3.2	2.4	0.21
Potatoes	8.8	N.D.	?
Total			1.5

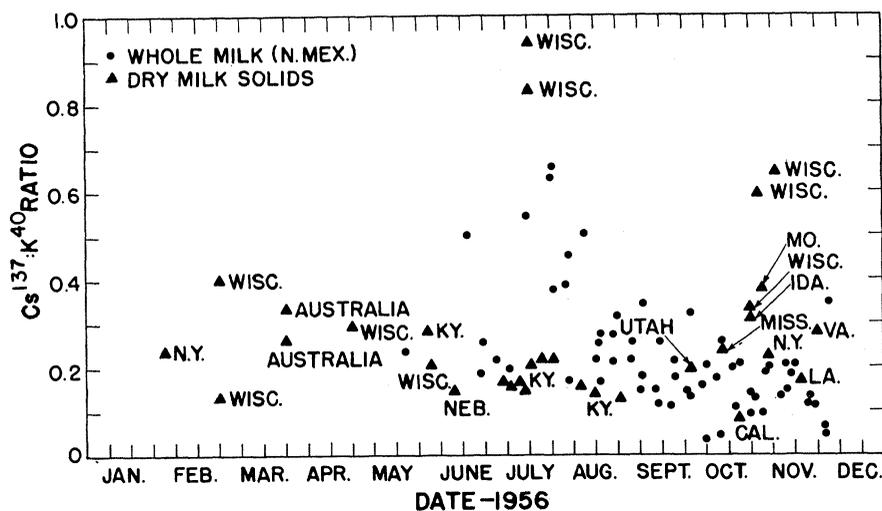


Fig. 7. Cesium-137/potassium-40 gamma ratio in milk during 1956.

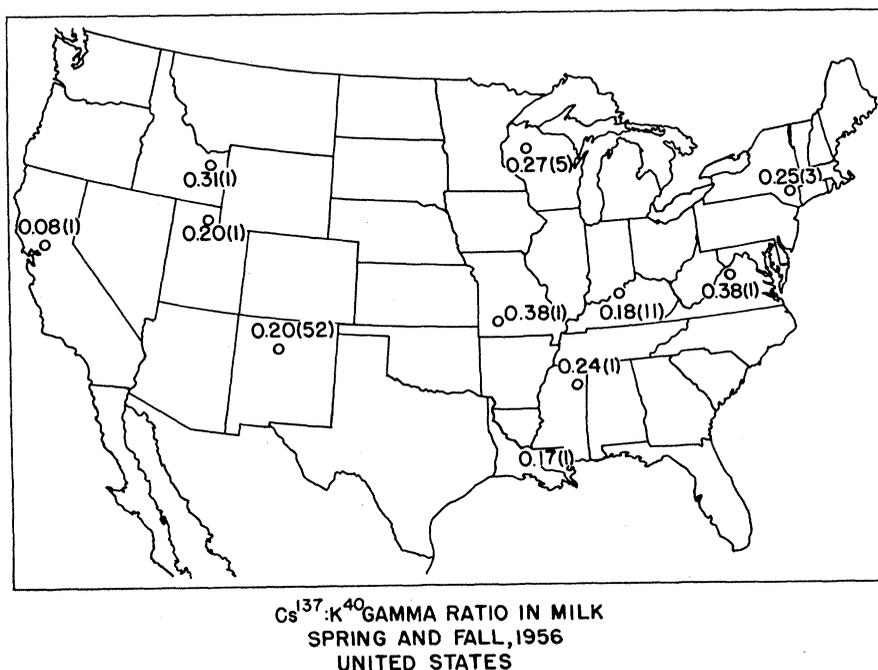


Fig. 8. Geographic distribution of cesium/potassium in milk.

Measurements of cesium-137/potassium-40 ratios in milk during 1956 (Fig. 7) indicated peak activities resulting from periods of tropospheric fallout. The relative effect of such increases in foodstuffs on the cesium-137 level in people can be estimated from the simple model shown in Fig. 9. A step function change in the foodstuff level will be followed by a  $(1 - e^{-\lambda t})$  change in the population level (where  $\lambda$  is the biological elimination rate), and a new equilibrium value will be reached only after an elapsed time of the order of 1 year. If the foodstuffs return to their previous value before equilibrium is attained, the population level will cease rising and will decay back to its previous value with a half-time corresponding to the biological elimination rate.

This model can be applied to the situation during July and August, when the level of cesium-137 in milk rose by about a factor of 3. Since not enough data are available to define completely the shape of the peak, and since milk values are used as representative of all foodstuffs, the actual peak can be replaced with a step function of the same approximate area. This gives a rise of about two times "normal" for a period of 50 days. In this case, the maximum rise in the population level, predicted on the basis of the model in Fig. 9, is 20 percent. Using the average value of the cesium-137/potassium-40 ratio for the control subjects in the spring of 1956 (Fig. 3) of 0.4, their calculated ratio 6 months later is 0.5. The observed summer maximum average was 0.48, in agreement with the model.

An estimate of the biological half-time of cesium-137 in the chronically exposed case was obtained by counting a large urine sample representing 52 man-days of excretion. The sample showed 408 disintegrations per second of potassium-40 (136 grams of potassium) and 40 disintegrations per second of cesium-137. Assuming an average body burden of 0.005 microcurie of cesium-137 for the six subjects who contributed urine samples, the excretion rate is 0.004 per day, which corresponds to a half-time of some 180 days if the excretion is exponential and entirely urinary. If fecal excretion is 25 percent of urinary, the half-time would be 145 days. This is in agreement with the biological half-time of 140 days observed on volunteers who ingested 1 microcurie of radiocesium (9).

Using Bureau of Agriculture statistics for food consumption per capita in the United States (22) and our preliminary values for the average cesium-137 content of foodstuffs, the dietary intake of cesium-137 can be estimated (Table 2). On the basis of these data, it appears that milk contributes about 50 percent and meat about 25 percent of the cesium-137 found in the body. The excretion rate of cesium-137 can also be estimated from these intake data. This method is only an approximation because of uncertainties in diet and in the average cesium-137 level in the various dietary components. According to the data in Table 2, the turnover rate is of the order of 1.5 millimicrocuries per month, compared with the observed value of 0.6 millimicrocurie. Part of the discrepancy may result from using retail weights in computing the diet with no allowance for wastage and loss of minerals in cooking, but the principal source of error is probably the inadequacy of our knowledge about cesium in foodstuffs. For comparison, a similar computation was made for potassium (Table 3). The calculated potassium intake is about 3 grams per day, while the observed urinary excretion was 2.6 grams per day. Elkinton and Danowski (23) reported potassium turnover as falling in the range of 2 to 6 grams per day.

While the spring 1956 average value

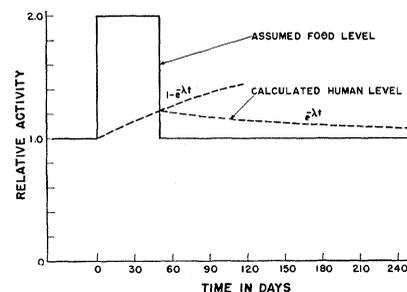


Fig. 9. Calculated effect of increased cesium in diet on human level.

Table 3. Calculated potassium intake based on per capita food consumption.

Source	Con- sump- tion (lb/mo)	Potassium	
		Con- tent (g/lb)	In- take (g/mo)
Dairy products	5.8	6.0	35
Meats	11.4	1.2	14
Flour and cereal	13.0	0.5	6
Vegetables	16.8	1.0	17
Citrus fruits	3.2	1.0	3
Potatoes	8.8	2.0	18
Total			93

for the cesium-137/potassium-40 ratio in milk was 0.25, the average in people for the corresponding period was 0.4. This difference may be explained on the basis of the longer hold-up time of cesium in the body as compared with potassium. If  $q_{\text{cesium}}$  is the amount of cesium in the average daily diet, and  $q_{\text{potassium}}$  is the corresponding amount of potassium, then  $q_{\text{cesium}}/q_{\text{potassium}}$  is the cesium/potassium ratio for the average diet. The milk ratio can be used since it is the most important single factor and is the only one known with any accuracy. The equilibrium amounts of cesium and potassium in the body, on the basis of the simplest model, will be given by the product of  $q\tau$  for each element, where  $\tau$  is the mean life of the element in the body (24). For cesium,  $\tau$  has been determined to be 200 days;  $\tau$  for potassium can be estimated from our data on the potassium content of normal urine as about 58 days. Therefore, cesium should be concentrated relative to potassium by a factor of 200/58, or 3.4. If the average diet ratio is 0.23, the predicted ratio in people is about 0.8. This is too high by a factor of 2.

Libby (4) has estimated stratospheric injection by Operation Redwing at about 6 megatons of fission products in addition to the 18 megatons left from the

previous operations. This would imply a 30-percent increase in the fallout rate from the stratospheric (world-wide) component after the tropospheric component is gone. A comparison of the spring and autumn milk averages indicates no detectable increase in the fallout rate. The spring sampling was inadequate; hence there is considerable uncertainty about the proper average. However, it would appear that, if anything, the cesium levels in the fall were lower. This may be a seasonal variation resulting from the change from pasture to hay feeding of the dairy herds, which would conceal possible small increases.

### Summary

Measurements of the cesium-137 content of people and of foodstuffs indicate that this nuclide is unlikely to be a decisive factor in the long-term hazards from weapons testing and reactor waste disposal. The amount of cesium-137 now present in the population of the United States averages 0.006 microcurie and shows no marked dependence on geographic location. The average radiation dose received from cesium-137 is one-twentieth of that received from natural radiopotassium and 1 percent of the average total dose from all natural sources. Because of the short biological half-life of cesium of about 140 days, it does not accumulate in the body as does strontium-90. The study of the distribution of cesium-137 is being continued to furnish information on the mechanisms of the fallout process and provide a measure of the rate of fallout and of stratospheric storage.

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13. This work was performed under the auspices of the U.S. Atomic Energy Commission. We are grateful to a number of persons who assisted in various phases of this program. We are particularly indebted to L. D. Marinelli and C. E. Miller of Argonne National Laboratory for helpful discussion and for their generosity in making measurements of interest to us on their crystal counter. Their assistance aided materially in the initial development of this program. J. W. Ballow supplied most of the foodstuff samples, and R. J. Remaley of the American Dry Milk Institute has been very helpful in the procurement of samples of dry milk. H. E. Gilbert of the Los Alamos Scientific Laboratory set up the punched card system used for electronic data processing.
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24.  $\tau = 1/\lambda = t_{1/2}/0.693$ , where  $\tau$  is the mean or average time the nuclide remains in the body,  $\lambda$  is the elimination rate, and  $t_{1/2}$  is the time necessary to remove half the body burden.

