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Radionuclides in Man from Nuclear Tests

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The results of recent experimental work in the determination of the stratospheric inventory of fission products—i.e., the fraction of Sr^{90} and Cs^{137} taken up directly from rain—and new measurements of the concentration of Sr^{90} in human bone at this laboratory make possible a more accurate prediction of the future radiation doses from these isotopes to the world population. It is concluded that the U. S. diet has passed its peak concentration of Sr^{90} and Cs^{137}, that Sr^{90} probably will remain the largest contributor to the radiation dose to an individual, and that Cs^{137} measurement can be used to monitor Sr^{90} in milk in emergency situations. These results also suggest that in the event of large-scale nuclear warfare, the general radioactive contamination would not preclude the existence of large populations if short time (6 to 12 months) survival were possible.

In 1953, when the potential seriousness of world-wide fallout was first clearly identified, virtually nothing was known of the mechanisms, or rates of movement of nuclear debris from the point of detonation to the human population. In the intervening years, as a result of a major research effort at many laboratories, the larger aspects of the fallout problem have been solved. The previous speakers and their associates have played a large role in this development and their papers have treated many key problems in the movement of fission products to and through the food chains. It is my lot to summarize briefly the present status of the situation from these papers and other sources, and to report on some of the work at the Geochemical Laboratory of Columbia University on the levels of some of the critical radionuclides in man.

Production and Distribution of Nuclear Debris

A comprehensive study of the stratospheric reservoir has been operative for the past 3 years. By using a meridional net woven by flights of manned aircraft and supplemented by the balloon program of the Atomic Energy Commission, results were obtained which now make it possible to state the rate and mechanism of removal of the debris from the stratosphere (*8*). Figure 1 shows the tropical and polar tropopauses, the discontinuity, and the ground level. Debris injected into the polar stratosphere has a half residence time of about 6 months and essentially all comes out into the Northern Hemisphere. Debris injected by megaton weapons at the equator has a half residence time of about 1 year. These values were determined by actually measuring the change in the stratospheric reservoir with time.

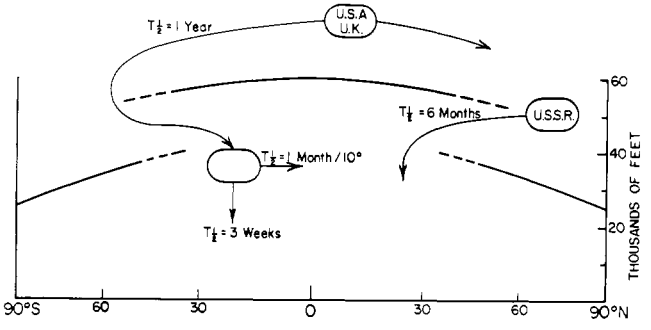

Figure 1. Transfer of fission product debris from stratosphere to surface of earth

The dominant transfer mechanism of stratospheric debris is through the break between the polar and equatorial tropopauses. The quantity of debris which passes through this gap is greater during the winter than at any other time of year, as this is the period of greatest activity of the jet stream which is located in this discontinuity. Once in the troposphere, stratospheric debris is removed by precipitation with a 3-week half life. The debris also moves laterally at the rate of about 10° per month. Since the polar regions have very low precipitation, this volume of the troposphere merely acts as a storage space, and thus, the average specific activity of rain is essentially constant from 30° to 90° N or S. Toward the equator, however, the washout is effective so that a minimum in the deposition occurs in the 0° to 10° S latitude belt.

Figure 2 shows that the specific activity of rain increases in the spring of each year, following the winter stratospheric transfer, and then reaches a minimum in the fall. The fact that W^{185}, a tracer introduced in the U. S. Hardtack series at 10° N in the summer of 1958, also shows a spring peak in 1959, proves that this seasonal pattern has a meteorological origin and is not primarily due to Russian testing.

The distribution of the Sr^{90} on the surface of the earth is also fairly well known. The more recent estimate is shown in Figure 3 (*18*) where the ratio of specific activity on the ground between the latitude zones 40° to 50° N to

40° to 50° S is about 3. The asymmetry would not be as great were it not for the U. S. S. R. testing in the north polar latitudes.

The integrated surface deposition for the past 6 years and the predicted value for July 1960 (*18*) are given in Table I. The absolute values are probably good only to ±20%, but the relative yearly inventories are probably more precise. The total Sr^{90} deposited by April 1960 should be about 4.0 MC. (megacuries). At this same time the residual stratospheric inventory should be only 0.6 MC., with about two thirds of this being in the northern stratosphere. Therefore, the present surface distribution will remain unchanged and that the total cumulative deposit will increase to a maximum in 1961, but this maximum will only be about 10% greater than the surface deposit in early 1960.

The measured specific activity of rain for the zone 30° to 60° N for the past 3 years and the predicted levels for 1960 are given in Table II. Note the sharp reduction in 1960, where the specific activity in the summer months is predicted to be half of 1955–58 and one quarter that of 1959.

The mechanism of removal of the particulate debris is by precipitation. The Sr^{90} and Cs^{137} of stratospheric origin are essentially in water-soluble form (*16*). Some of the Nevada debris is insoluble; this represents less than 5% of the total in the eastern U. S. Further, the Sr^{90}/Cs^{137} ratio is essentially constant so that the more comprehensive Sr^{90} data may be used to calculate Cs^{137} deposition. Pu^{239} is essentially in insoluble form.

When the rain hits the surface of the earth, the isotopes are distributed vertically in the soil according to their chemical behavior. Sr^{90} penetrates most deeply, Cs^{137} much less, and Pu^{239} probably is held in the upper quarter inch. Subsequent to initial deposition and vertical distribution there is little or no vertical movement on a time scale of a few years.

Table III summarizes measurements of Sr^{90} with depth from various localities and laboratories. Detailed work at this laboratory has shown no significant change in the vertical profile from 1953 until the present. At least 70% of the Sr^{90} still appears to be in the upper 2 inches of untilled soil and at least 90% of it remains in the upper 6 inches in the average soil.

Carbon-14, as produced in the nuclear explosion, is mostly in the form of CO_2. As such it probably follows the particulate debris into the troposphere, but there its similarity ends, for the CO_2 is not washed out by rain but rather is mixed with the pre-existing CO_2 of the troposphere and thus enters the CO_2 cycle. Some exchanges with the ocean, some with the biosphere, and some is returned to the stratosphere (*3*).

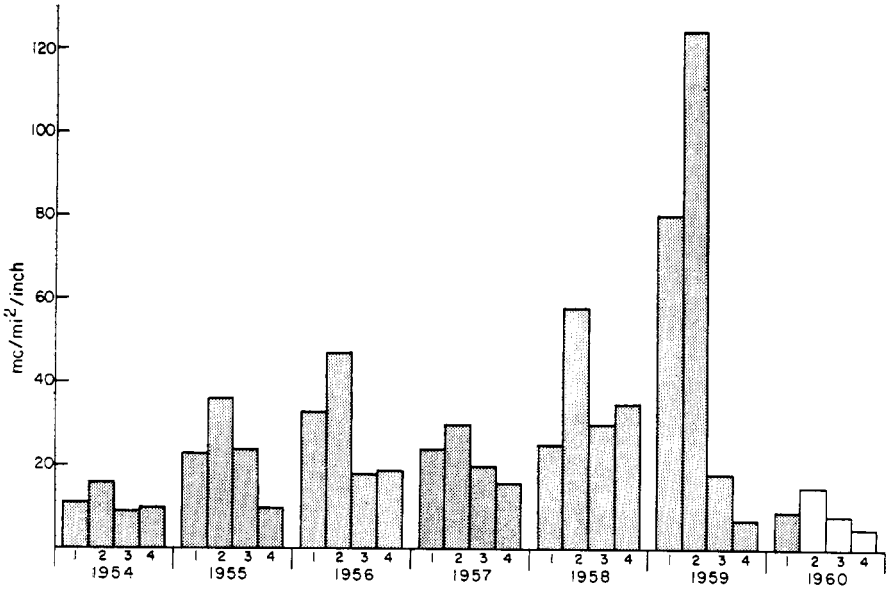

Figure 2. Seasonal variation in specific activity of Sr^{90} in precipitation, 30° to 90° N

Table I. World Surface Inventory of Sr^{90} with Time (*18*)

Date	Megacuries of Sr^{90}
July 1954	0.2
July 1955	0.6
July 1956	0.9
July 1957	1.4
July 1958	2.0
July 1959	3.2
July 1960	4.1[a]

[a] Predicted value.

Table II. Specific Activity of Sr^{90} in Rain 30° to 60° N during Each Quarter from 1954–60

Year	Millicuries/Sq. Mile/Inch/Quarter			
1954	0.11	0.16	0.09	0.10
1955	0.23	0.36	0.24	0.10
1956	0.33	0.47	0.18	0.19
1957	0.24	0.30	0.20	0.16
1958	0.25	0.58	0.30	0.35
1959	0.80	1.24	0.18	0.07
1960	0.14[a]	0.20[a]	0.11[a]	0.04[a]

[a] Predicted values.

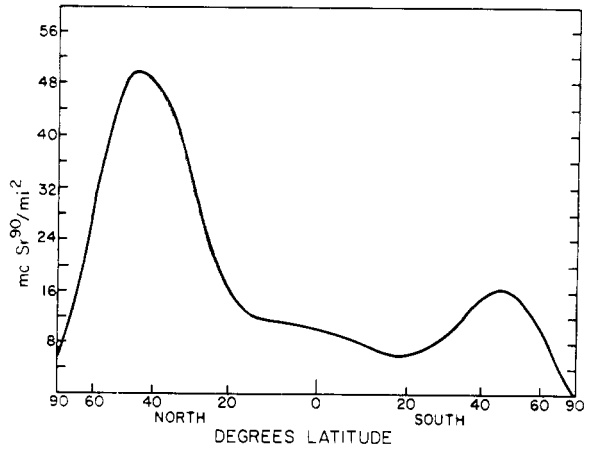

Figure 3. Ground level inventory of Sr^{90} in late 1959, after Walton, 1960

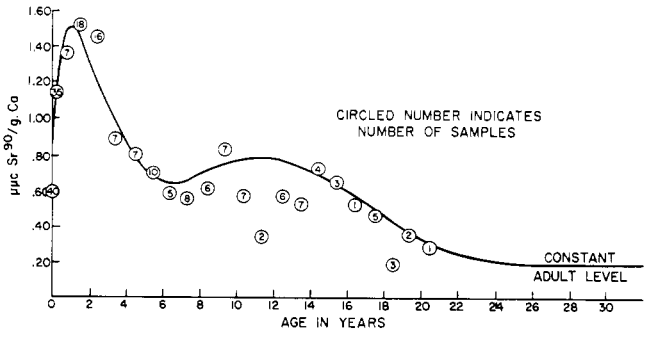

Figure 4. Theoretical curve and experimental data showing relation of specific activity of Sr^{90} in skeleton with age of individual for 1957–58 in Western culture, 30° to 90° N

Table III. Summary of Experiments on Vertical Penetration of Sr^{90} in Soil

Set of Samples	Reference	No. of Samples	Date of Collection	% Sr^{90} in 0–2 Inches
Lamont campus	[a]	3	Aug. 1953	80
Midwest farms	(9)	6	1953, 1954	80
England and Wales	(1)	4	July 1956	≥80 (0–4 in.)
U. S. collection	(9)	17	Oct. 1956	74
Utah and Montana	[a]	5	Aug. 1957	74
U. S. collection	(10)	17	Oct. 1957	73
Ecuador and Venezuela	[a]	5	Nov. 1957	80
Rome and Lausanne	[a]	3	Mar. 1958	88
New York City area	[a]	4	May 1958	76
Mandan, N. D.	[a]	1	Aug. 1958	75
U. S. collection	(10)	23	Oct. 1958	62
			Weighted av.	72

[a] This laboratory.

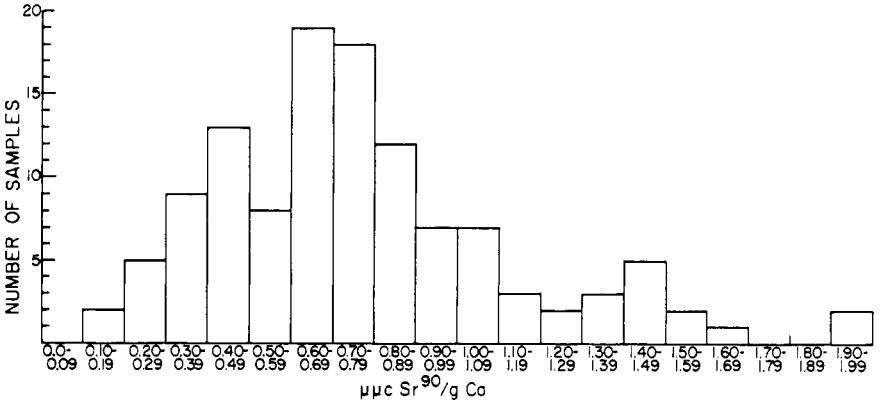

Figure 5. Histogram of New York and Chicago fetuses, 1957–58

Concentration of Sr^{90} and Cs^{137} in Plants

The basic question as to what determines the quantity of a fission product in a plant is more complex and was one of the major unknowns in the fallout equation until recently. The weight of evidence now points to direct absorption as more important than uptake through the roots of plants. Or, put another way, the concentration of, say, Sr^{90} or Cs^{137} in grass, is primarily dependent on the rate of fallout (specific activity of rain × quantity of rain per month) rather than the cumulative deposit in the soil.

Langham (13) has shown that in 1959 the Cs^{137} level in milk did not climb with the cumulative ground deposit, but after the spring peak it dropped to levels below that even in 1957.

Christenson and Fowler (7) have shown that the plants get most of their Ca and Sr from soil levels well below those containing most of the Sr^{90}.

Nishita, Romney, and Larson (15) showed that the uptake of fission products by plants from contaminated soils from the Nevada test site is small, and Tukey, Wittwer, and Bukovac (17) demonstrated that aboveground parts of plants readily absorb radionuclides from external spray applications.

By field experiments using Sr^{89} to simulate the distribution of Sr^{90} in the soil, Burton, Milbourn, and Russell (6) could predict the quantity of fallout Sr^{90} in the soil that would move into rye grass. This proved to be only 10 to 20% of the observed Sr^{90} from fallout. Thus they attributed at least 80% of the uptake to the rate factor, that is, to direct fallout, and less than 20% to the cumulative factor.

An experiment is in progress which will make it possible to obtain a fairly accurate estimate of the relative importance of these two factors for all of North America. The procedure will be to compare the Sr^{90} concentration in milk from all stations, including the 40 powdered milk plants by the Los Alamos Scientific Laboratory, the domestic liquid milk stations maintained by the Public Health Service in about a dozen cities, and other U. S. and Canadian localities for the summers of 1958, 1959, and 1960. The cumulative deposit by the summer of 1960 will be double that of 1958, but the rate of fallout will only be one third. Thus, if Russell's experiment (6) holds for the average North American milk farm, it would be expected that in 1960 the Sr^{90} concentration in milk will drop to about one half the 1958 level, or to about 4 μμc. of Sr^{90} per gram of Ca, whereas in 1959 it averaged 12 μμc. of Sr^{90} per gram of Ca. It might level off at about 2 μμc. of Sr^{90} per gram of Ca in 1961–62.

Table IV. Micromicrocuries of Sr^{90} per Gram of Ca Found in North America

Sample	1957	1958	1959
Milk	5.7	8.0	12.2
Diet	6.6	9.2	13.7
New bone, cal.	1.6	2.3	3.4
0 to 4 years	1.0	1.7	..
Fetus, cal.	0.8	1.2	1.7
obs.	0.5	0.7	0.9

Since there is reasonably comprehensive coverage of the Sr^{90} concentration in North American milk, it is useful to obtain the relation between the level in milk and the total diet. Individual plant foods vary greatly in their Sr^{90} content and their average concentration of Sr^{90} is somewhat higher than milk. Weighting each food by the average contribution of that food to the diet, several investigators have estimated the total Sr^{90} in the diet of the U. S. and the U. K. (5, 12). The average diet appears to be 15 to 20% higher than milk.

Earlier in this symposium, Wasserman (19) has reviewed the situation on discrimination factors and has pointed out that on normal diets the Sr/Ca Observed Ratio, $O.R._{diet\text{-}human\ bone}$, is 0.25. Further, the placental discrimination presents another factor of 2 against strontium for a net $O.R._{diet\text{-}fetus}$ of 0.12 if the mother's skeleton is not involved.

Table IV gives the experimental milk values for the last 3 years and the computed diet levels for the U. S. From the latter the concentration of Sr^{90} in newly depositing bone tissue can be estimated as shown. The observed average values for bones of young children are in excellent agreement when the newborn child has a relatively low Sr^{90} concentration in his skeleton and those more than one year old have lived on diets with lower specific activity of Sr^{90} in previous years. The bone level for 1-year-olds approaches the theoretical value for new bone.

The concentrations of Sr^{90} in fetuses from New York and Chicago are lower than the calculated value assuming an $O.R._{mother\ diet\text{-}fetus} = 0.12$. The reason for this is probably dilution of the dietary calcium in the mother's blood with calcium from her bones which would have much less Sr^{90}.

Age Effect

As a result of the changing dietary levels since 1953, the average concentration of Sr^{90} in the bones of growing children is a complex function of age. Figure 4 gives the theoretical curve of the Sr^{90} concentration in bone in the U. S. in 1957–58 as a function of age of

the individual. This plot was first suggested by Langham (*13*) and is based on the following parameters: the calculated diets for North American children since 1953, an $O.R._{diet\text{-}bone}$ of 0.25, the average Ca deposited each year after Mitchell *et al.* (*14*), and $O.R._{diet\text{-}fetus}$ of 0.12 all for 1957–58. The points are the experimental averages of all samples measured. Bryant (*4*) obtained a similar result but with a lower curve for the same period for children's bone in Australia. This suggests that the assumptions on which the theoretical curve is based are correct.

The Sr^{90} concentration in the average adult skeleton from a given geographical area is independent of the age of the individual at death. The concentration of Sr^{90} in the adult skeleton depends on the dietary level and the rate of exchange. This latter is probably a function of time with the most rapid exchange occurring in the first few years. At present the results are consistent with a turnover rate of about 3% per year.

Geographical Distribution and World Average

The geographical distribution of activity levels in human bone is essentially the same as for the gross fallout. For a given type of diet—for example, the Western-type diet, which encompasses all of North America above 30° N and most of Europe—the average bone content of Sr^{90} is the same everywhere. At 30° to 60° N in the oriental rice-diet areas there is evidence that the bone levels may be somewhat higher. The bone levels in the Northern Hemisphere are about twice those of the Southern Hemisphere. The ratio of bone levels between the hemispheres is greater than the ratio of the absolute fallout as a result of the exchange of food between the hemispheres.

Some comparative data are given for one station near Bonn, Germany, for which reasonably large numbers of samples are available (Table V). The average values for the world population in April 1960 are probably about 0.3 picocuries (pc.) of Sr^{90} per gram of Ca for adults and 2.5 pc. of Sr^{90} per gram of Ca for 1-year-olds.

Distribution Curves

The average bone level for people at any age, in large population groups at any location on earth, at any time in the future may now be predicted on the basis of the information discussed above. The distribution around the mean of the population is a more difficult problem. Each group has its distinct problems. Adults, for example, do not have an age dependency but their bones have had such low activities that fairly large experimental errors were encountered. One-year-olds might be considered, but the great individual differences in the rate of development would cause such a large apparent variation that the true equilibrium distribution would be narrower. Fetuses will vary as a result of the mother's dietary habits in a more complex way than the adult population. On the other hand, a reasonable sample of fetuses will permit a fair estimate of the distribution curve for fetuses at least over the large fraction of the fetus population.

Table V. Sr^{90} in Human Bone Near Bonn, Germany[a]

(Micromicrocuries of Sr^{90} per gram of Ca)

Age	1955–56	1957–58	1958–59
Adult	0.15 (90)	0.35 (61)[b]	0.46 (87)
0–4 years	...	1.47 (34)	1.80 (17)

Std. dev., ±40% Std. error on mean, ±5%

[a] No samples available for 1956–57.
[b] Values in parentheses are number of samples.

Figure 5 shows a histogram for the New York–Chicago fetuses. This curve is slightly skewed to the right. It can be fitted roughly by a log normal plot or by an infinite number of other functions. What these data do provide is a good estimate of the distribution among fetuses in the U. S. and western Europe within a factor of three of the mean. It does not permit reliable extrapolation far to the right. These results suggest that about 95% of the fetuses in North America will be within a factor of two of this mean. Similar figures were obtained for the analyses of the 200 to 300 whole body ashes of New York cadavers.

Little can be said about the proper extrapolation to the right. One thing appears certain, few people in modern society can maintain a high level for any sustained period unless they eat a limited abnormal diet. The total effect of different food sources with time is to produce uniformity in the population. As the concentration of Sr^{90} in the diet levels off, variations due to different rates of metabolism among the population will also be reduced. Thus, the present empirical distribution curves all represent an exaggerated situation.

Estimates of the small fraction of people who live on special diets or a food from restricted geographical areas such as the primitive Indians of the upper Amazon must be made by analyzing the suitable foods. This has been done at this laboratory. The maximum level that has been found for a group of people was from the jungle of Ecuador where one Indian village in late 1957 was consuming a major calcium source that carried 40 to 50 pc. of Sr^{90} per gram of Ca when the U. S. diet was about 7 pc. of Sr^{90} per gram of Ca. It is unlikely that people in such an environment exceed 0.001% of the world population. Since this occurred in an area of

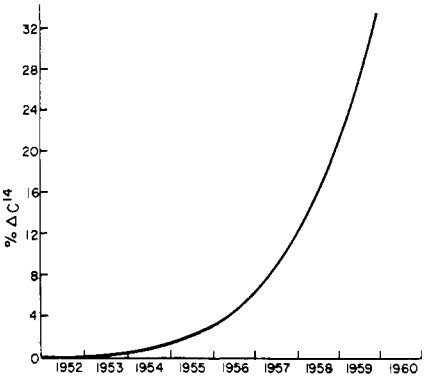

Figure 6. Increase in specific activity of C^{14} in ground level CO_2 as a result of nuclear detonations

200 inches of mean annual rainfall, the concentration of Sr^{90} in this diet should drop very rapidly with the depletion of the stratospheric reservoir. Present data do permit conservative prediction of the distribution curve for 95% of the population, but are not adequate to predict the upper end of the world population histogram.

Predictions

If the fraction of Sr^{90} in food due to the cumulative soil deposit in 1959 is taken as 10 to 20% (*6*), the U. S. diet has already passed its peak and it should level off toward the end of 1960 at about 2 $\mu\mu$c. of Sr^{90} per gram of Ca. This is equivalent to a new bone level of 0.5 $\mu\mu$c. of Sr^{90} per gram of Ca. It should then decay slightly faster than the radioactive half life. This would yield a maximum radiation dose of 1% of natural background. The skeletons of young children have also, therefore, reached their peak and will decrease in all cases. Adults will increase slightly to equilibrium.

The Cs^{137} level in 1958 was about 54 $\mu\mu$c. of Cs^{137} per gram of K. Since little Cs^{137} uptake from the soil is expected based on Langham's report (*13*), the internal concentrations should drop by the end of 1960 to about 10 $\mu\mu$c. of Cs^{137} per gram of K and to 1 $\mu\mu$c. of Cs^{137} per gram of K by the end of 1963. The long-term hazard of this isotope would, therefore, be entirely due to its contribution to external gamma radiation and would yield a dose of 3 to 4

Table VI. Carbon-14 Inventory

(10^{27} atoms on July 1)

Reservoir	Natural Steady-State	Bomb-Produced C^{14} [a]		
		1958	1960	2000
Stratosphere	7	8.4	6	0
Troposphere	28	3.5	12	3
Biosphere	8	0.2	1	1
Ocean	1960	0.8	3	18

[a] Through 1959.

milliroentgens (mr.) per year beginning in 1960, from a geometrically planar surface, or if the usual factor of 10 for shielding is accepted, 0.3 to 0.4 mr. per year to average man. This will probably decay away slightly faster than the radioactive half life due to mixing in the soil, surface washing, etc. This level is about 0.3% of the natural background dose rate.

Pu^{239} is a potentially hazardous isotope, but its insolubility permits very great discrimination against it from soil to plant or plant to man, as Langham (*13*) and Nishita (*15*) have shown. It may be inhaled, however. Krey (*11*) has reported finding Pu^{239} in human tissue at levels up to 1% of maximum permissible concentration (M.P.C.). This suggests inhalation of direct fallout, and thus, the Pu^{239} levels will also decrease as the rate of fallout.

Broecker and coworkers (*2*, *3*) and others have examined the increase in specific activity of C^{14} in the lower atmosphere and biosphere. Figure 6 shows the increase with time. The atmosphere toward the end of 1959 was 30% higher than prebomb background. This level should increase somewhat in 1960, and if C^{14} follows the fission products out of the stratosphere, it will begin to decrease in 1961 as it exchanges with the carbon in the oceans with a half time of 7 years. The time lag for equilibration with the human body is about 1 year. The perturbation that the bomb C^{14} has given the natural reservoir can be readily seen in Table VI. In 1958 the stratospheric concentration of C^{14} had doubled. By 1960 this concentration should already be lower than it was in 1958, the troposphere held near its maximum representing a 40% increase over the natural background, but by the year 2000 the bomb C^{14} would largely be in the ocean.

The contribution to the body dose from natural C^{14} is about 1 mr. per year, hence in 1960 this would have increased to about 1.4 mr. per year, an increase of about 0.3% of the natural background dose. This will disappear more rapidly than the Sr^{90} or Cs^{137}, but the integrated dose to the world population may be larger.

Conclusions and Observations

The evidence suggests that the greatest dose rate from weapon debris produced to date has already been experienced. During 1960 the contributions of Sr^{90}, Cs^{137}, and C^{14} may be comparable. Thereafter, Cs^{137} will contribute only to the external gamma dose and C^{14} will be gradually taken up by the sea at a somewhat faster rate than the radioactive decay of Sr^{90} and Cs^{137}. In all cases after 1960 the total radiation dose rate from all of these isotopes should contribute less than 1% of the natural background dose.

Two other observations that result from the work reported in this symposium may be in order.

Since concentration of both Sr^{90} and Cs^{137} appear largely to depend on the rate of fallout, the analyses of Cs^{137} in milk should permit a reasonable estimation of the Sr^{90} content. Indeed some hundreds of powdered milk samples that were run at the Los Alamos Scientific Laboratory for Cs^{137} and at Lamont for Sr^{90} show a standard deviation in the ratio of less than a factor of 2. Thus, since the Cs^{137} analysis can be made in a matter of a minute or two and the Sr^{90} radiochemical assay takes several weeks, it is clear that Cs^{137} assay should be used and, indeed, is probably adequate for monitoring in an emergency situation.

Much has been said about the effect on the world population of a nuclear war. From the data presented here today, it can be calculated that nuclear war using 3000 megatons of bombs, a common estimate if most major cities in the U. S. and U. S. S. R. are considered targets, the average human bone level at equilibrium from world-wide fallout would only increase by a factor of 15 over that which would be present in 1961 as a result of weapon tests to date. This in time would yield a total annual radiation dose of about 15% of the natural background. The radiation hazard in a nuclear war is local and acute. The long-range effects to noncombatants is therefore relatively small. Even in the areas of intermediate fallout, it appears unlikely that the long-term dietary levels from food grown on this soil would exceed the maximum permissible dose for industrial workers. This is not to minimize the gravity of the acute situation for some months after the event but does serve to clarify the problem of survival.

Acknowledgment

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