Some Considerations of Present Biospheric Contamination by Radioactive Fallout

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Strontium-90 and Cs^{137} fallout from nuclear weapons tests through October 1958 and entry of these radionuclides into the food chain are summarized as an introduction to the ACS Symposium on Radioactive Fallout in Relation to Foods. Total production of Sr^{90} and Cs^{137} has been about 9 and 14 MC., respectively. Surface deposition levels will reach a maximum in about 1961. Both radionuclides have entered the food chain and man. Levels in man are lower, however, than predicted by ecological models based on 100% entry via plant uptake from the soil. It is not possible presently to estimate equilibrium levels in man and his foods with respect to integrated surface deposition levels. Calculation of radiation doses from Sr^{90} plus Cr^{137} to children born at the time of maximum fallout (assuming ecological equilibrium at approximately twice the 1958 levels) indicates respective maximum 30-year genetic and 70-year bone and bone marrow exposures that will be about 2, 9, and 4% of that from average natural background. If a linear dose-response relationship is accepted, fallout may increase the incidence of radiation-induced genetic, bone, and bone marrow disease in this generation by 2, 9, and 4%, respectively, of that resulting from natural background exposure.

TESTING of nuclear weapons releases radioactive debris into the atmosphere, and its deposition over the earth's surface is termed fallout. Fallout may be divided into three categories—local, intermediate or tropospheric, and stratospheric or world-wide—on the basis of time and place of deposition (*3, 4*).

Local fallout is deposited within the first 24 hours and confined to the immediate environs of the detonation. Since it is not widely distributed and much of it is deposited in the ocean, such deposition has not contributed significantly to general contamination of the biosphere.

Tropospheric fallout takes place in 20 to 30 days after a weapon test and occurs essentially in a poorly defined band around the world in the general latitude of the detonation. Small detonations contribute all their fission products to local and tropospheric fallout. Although tropospheric fallout is widely distributed, it has not contributed greatly to general long-range contamination.

Weapon detonations equivalent in energy release to 1 million tons (megaton) or more of high explosive carry their fission products into the stratosphere, where they are widely dispersed because of stratospheric diffusion and mixing. This material returns slowly to earth (average return time about 6 months to 3 years) and is referred to as stratospheric fallout. Stratospheric fallout has made the greatest contribution to long-range or world-wide contamination, since over 90% of all fission products produced by nuclear weapon tests has been from weapons with yields greater than 1 megaton.

The moratorium on large-scale nuclear weapon tests makes it appropriate at this time to summarize the present status of Sr^{90} and Cs^{137} biospheric contamination. These radionuclides are considered the most important in long-range fallout because of their long physical half lives and their chemical similarities to natural body constituents, calcium for skeletal formation and potassium for cellular function, which result in relatively high uptake in man and animals.

World Inventory of Sr^{90} and Cs^{137} Contamination

Weapon tests by all nations have produced fission products from about 90 megatons of fission energy release. (The recent French tests were so small that their contribution to the world inventory was insignificant.) Total energy release from fission plus fusion (thermonuclear energy does not produce fission products, but it contributes energy to carry the debris into the stratosphere) has been about 170 megaton equivalents (*3*). One megaton of fission energy produces about 100,000 curies of Sr^{90} (0.1 megacurie, MC.) and about 160,000 curies (0.16 MC.) of Cs^{137}. Total production of these two potentially harmful radionuclides has been about 9 and 14 MC., respectively. If distributed instantaneously and uniformly over the entire earth's surface (2×10^8 sq. miles), the Sr^{90} contamination level would be about 45 mc. per square mile of the earth's surface and Cs^{137} contamination would be about 70 mc. per sq. mile. These numbers, however, are misleading. The debris has not returned to earth instantaneously and, since both radionuclides have radiological decay half times of about 28 years, they have been disappearing at the rate of about 2% per year since their times of production. About 30% of their total production was deposited as local fallout (much of it in the ocean), where it does not contribute to general biospheric contamination. Furthermore, fallout is not uniform but occurs preferentially in the temperate latitudes and predominantly in that hemisphere in which the tests are held. This, of course, means that the highest deposition levels occur in the north temperate latitudes, where ironically about 80% of the world's population lives.

Distribution of Sr^{90} and Cs^{137} Contamination

The general distribution of fission products (as of November 1958, just after the last U.S.S.R. and U. S. test series but prior to significant fallout from these tests) was summarized in the 1959 Congressional Subcommittee Hearings (*3*). A select study group consisting of the most knowledgeable investigators in the field estimated that the total Sr^{90} and Cs^{137} production (9 and 14 MC., respectively) was, at that time, approximately equally partitioned as world-wide fallout already deposited on the surface of the earth, tropospheric and

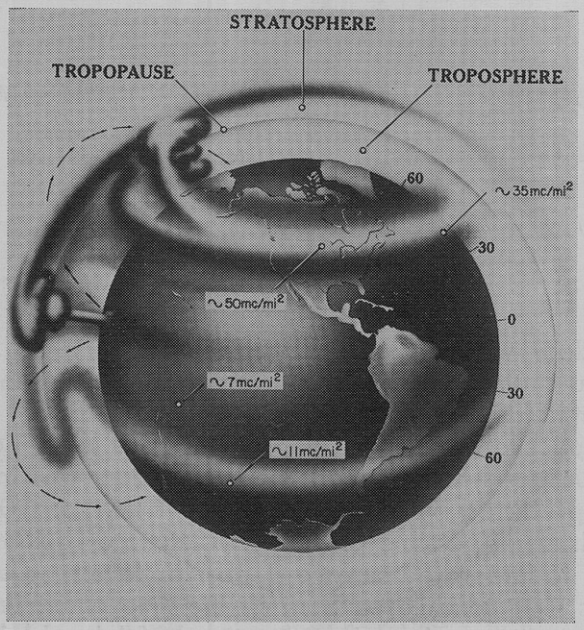

Figure 1. Mechanism of stratospheric fallout in relation to U.S. and U.S.S.R. test site locations

Surface deposition levels are for Sr^{90} as of November 1958

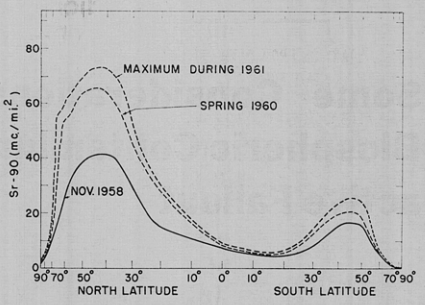

Figure 2. Predicted present and future Sr^{90} surface deposition levels as a function of latitude

stratospheric contamination yet to be deposited, and material that had been deposited at or near test sites where it would not make a significant contribution to general biospheric contamination.

Before the detailed distribution pattern of world-wide contamination can be understood, it is necessary to consider the mechanism of stratospheric fallout. Above the earth's surface is a region of more or less constant air temperature. This region is the tropopause; the atmosphere below it is the troposphere, and that above is the stratosphere. Near the equator the height of the tropopause is about 55,000 feet and over the polar regions its height is 30,000 to 35,000 feet. In the temperate latitudes (about 40° N and S), the tropopause is ill-defined or discontinuous. Masses of warm air rising above the tropopause near the equator may diffuse poleward, where they contact areas of intense cold in the stratosphere above the winter pole and upon cooling descend again into the troposphere. Return to the troposphere is preferential in the temperate latitudes because of the stratospheric circulation and the poorly defined or discontinuous troposphere. There is, in essence, a tropospheric-stratospheric air circulation system which results in fission products injected into the stratosphere being returned to the troposphere preferentially in the regions of 40° to 50° N and S latitudes. Once they are returned to the troposphere, they are deposited on the earth's surface in about 30 days, more or less in relation to area rainfall. Since most weapon tests have been held north of the equator, fallout has been much higher in the north temperate latitudes than in comparable regions of the southern hemisphere. Material injected at far northerly latitudes (as in the case of the large U.S.S.R. test series, October 1958) returns to the earth very fast (half time, 4 to 6 months) and all in the northern hemisphere. This mechanism of stratospheric fallout and the influence of U.S.S.R. and U. S. test site locations on world-wide distribution of radioactive contamination are illustrated in Figure 1.

On the basis of the above model of stratospheric deposition, one would expect maximum fallout levels to occur at about 40 to 50° N latitude, minimum levels near the equator, and a small peak in the south temperate latitudes. During 1958, a world-wide soil sampling program was conducted by the Department of Agriculture. When these samples were analyzed for Sr^{90} and the results averaged over 10° latitudinal bands, the surface concentration pattern was quite compatible with the general deposition model (*3*). Normalization of these data to November 1958 and integration with regard to world surface area showed that indeed approximately one third (3 MC.) of the total Sr^{90} produced was already on the earth's surface as of that date. Assuming no more weapon tests, this information and estimates of stratospheric fallout rate may be used to make crude predictions of present Sr^{90} and Cs^{137} surface contamination levels and the maximum level that will be reached when the rate of fallout from the stratosphere is just enough to compensate for radioactive decay of that already deposited. Such crude predictions of Sr^{90} surface deposition levels are shown in Figure 2. Assuming no fractionation of Sr^{90} and Cs^{137} during production and fallout, the Cs^{137} levels would be about 1.6 times the Sr^{90} values for the same latitude. These data show that the maximum Sr^{90} surface deposition level in November 1958 in the north temperature population belt (about 40° N) was about 40 mc. per sq. mile. By the spring of 1960, the maximum may be about 60 to 65, and during 1961 (at the time the level will begin to decrease) it will be about 70 to 75 mc. per sq. mile, assuming no more large-scale weapon tests. Because of the proximity of the Nevada Test Site, deposition levels in certain areas of the U.S. may be 10 to 15 mc. per sq. mile higher than the average for the general latitude. The disproportionate increase in the northern hemisphere between November 1958 and the present is a result of rapid fallout from the large U.S.S.R. test series at 73° N latitude in October 1958.

Incorporation of Sr^{90} and Cs^{137} into Man and His Ecological Environment

If Sr^{90} and Cs^{137} are widely distributed over the earth, they will enter the ecological cycle from which man gets his food and be taken into his body as radioactive contaminants of calcium and potassium, respectively, thereby increasing his radiation exposure. Much effort has been expended on working out and postulating ecological transport of Sr^{90} and Cs^{137} (*3, 4*). One would expect discrimination between Sr^{90} and calcium and between Cs^{137} and potassium as they are transported along the food chain from soil to man. Such discrimination is frequently expressed as the change in the radioisotope concentration in the required element as a result of passing through a particular step in the cycle. For example, the concentration of Sr^{90} in the calcium of milk is about 13% of its concentration in the calcium of the cow's diet, or $\frac{(Sr^{90}/Ca)_{milk}}{(Sr^{90}/Ca)_{forage}} = 0.13$. From such discrimination factors, the soil deposition levels, and the average diets of the population, one can compose ecological models which can be

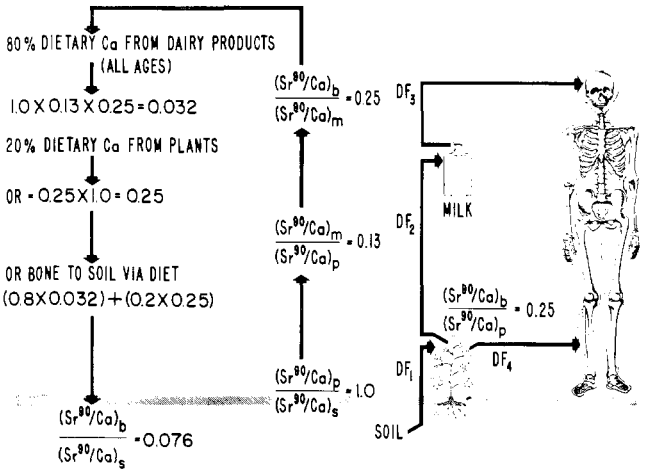

Figure 3. A model for ecological transport of Sr^{90} from soils to man (U.S. diet)

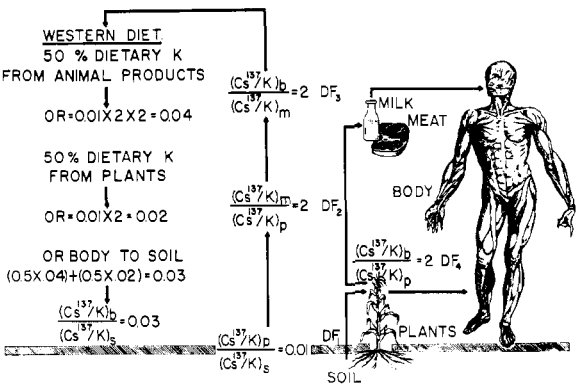

Figure 4. A model for ecological transport of Cs^{137} from soils to man (U.S. diet)

used to make crude postulations of the Sr^{90} and Cs^{137} levels in man when he and his ecological environment are in equilibrium with the soil contamination level. Such an ecological model for Sr^{90} is shown in Figure 3, and for Cs^{137} in Figure 4. These projections indicate that the Sr^{90} concentration in the bone calcium of the U.S. population (at ecological equilibrium) should be about 7% of its concentration in the available soil calcium, and that the Cs^{137} concentration in body potassium should be about 3% of its concentration in the available soil potassium.

Present indications are that Sr^{90} and Cs^{137} equilibrium levels in man may be much lower than predicted on this basis. There are so many inherent uncertainties in the basic parameters of such models that their principal value lies with their qualitative features in pointing out the routes of ecological transport. Only where entry of Sr^{90} and Cs^{137} into plants is predominantly through root absorption will their levels in man and his foodstuffs bear a consistent relationship to integrated fallout and conform to a true ecological model. Even in this case, a direct relationship between integrated fallout and biospheric contamination may be transient, since diffusion below the root feeding zone may be expected eventually.

The most certain way of estimating fallout contamination levels in man and his food chain is by direct analysis. Direct analysis leaves no doubt that Sr^{90} and Cs^{137} are finding their way into the biosphere. Strontium-90 surface deposition levels in the New York area and in milk from the same region have been measured by the Atomic Energy Commission's New York Health and Safety Laboratory since mid-1954 (*3*). Although the Sr^{90} content of milk has been increasing generally with the integrated soil deposition level since the beginning of large-scale weapon tests, it fluctuates seasonally and may actually decrease while the deposition levels are always increasing. These data may be interpreted as meaning that present Sr^{90} concentrations in milk are reflecting both uptake from the soil and via direct fallout contamination of forage. Cesium-137 in milk fluctuates even more widely than Sr^{90}, suggesting that a much greater fraction is entering the food chain via direct contamination of vegetation. These observations are important because of the dependence of future Sr^{90} and Cs^{137} concentrations in foods and in man on their modes of entry into the ecological chain. If entry is largely via direct contamination of vegetation, contamination of man and his food chain should decrease in relation to their decreasing fallout rates as the stratospheric reservoir becomes depleted. If entry is largely via uptake from the soil through plant roots, then levels in man and his foods will follow the integrated concentration in the soil. The relative extent to which Sr^{90} and Cs^{137} is entering the ecological cycle via these two routes has not been established unequivocally. Undoubtedly, the ratio will vary with time, soil type, available soil cations, plant species, and other factors. The United Kingdom's Agricultural Research Laboratory (*1*) has estimated that only 20% of the Sr^{90} in milk produced in England and Wales in 1958 was derived from soil uptake and about 80% from direct contamination of forage. The ratio will, of course, change as the stratosphere is depleted of fallout, and eventually 100% will be derived from soil uptake. The equilibrium level attained in man's food cycle (and subsequently in his bones), however, may be less than the maximum predicted by the ecological cycle shown in Figure 3 by as much as a factor of five. The actual equilibrium level of Cs^{137} may be lower than that predicted in Figure 4 by an even larger factor.

Analyses of other foods, meats, cereals, fruits, and vegetables, show the presence of Sr^{90} and Cs^{137}. Approximately 90% of Sr^{90} in the diet of the U.S. population is derived from milk, vegetables, and cereals, their respective contributions being about 60, 20, and 12% of the total. In the case of Cs^{137}, about 80% is coming from two dietary sources—namely, milk and meat products.

Table I. Average Concentration of Sr^{90} and Cs^{137} in People as a Function of Time

Year	Strontium,^{90} μμc./G. of Ca: Children, 0 to 4 yr.	Strontium,^{90} μμc./G. of Ca: Adults	Cesium^{137}, All Ages, μμc./G. of K
1954	...	0.07	..
1955	0.5	0.11	..
1956	0.7	0.10	48
1957	1.2	0.13	49
1958	1.7	0.21	60
1959	2.3	0.31	74

Since the principal purpose of fallout studies from the biological viewpoint is the assessment of the potential hazard to man, direct measurements of Sr^{90} and Cs^{137} levels in people are extremely important. Measurements of the actual levels of Sr^{90} in human bone have been carried out since 1954 by laboratories in the U.S. and the U.K. The most recent report from the Lamont Geological Observatory (*2*) summarizes the results of several hundred bone analyses collected from various parts of the world but rather heavily weighted in favor of the north temperate latitudes and of Western populations. The average Sr^{90} concentration in the bones of children (0- to 4-year age group) and adults as a function of time is shown in Table I (*2*). Bone analyses indicated an average Sr^{90} skeletal burden in 1-year-old children from Western cultures during 1959 of 2.1 μμc. per gram of calcium. One-year-old children are nearest to being in equilibrium with Sr^{90} in the diet and, therefore, have the highest bone levels. Variation of aver-

age Sr^{90} burden with age has been shown to be in agreement with predictions based on skeletal growth and fallout rates. Adult bones which contain only that Sr^{90} laid down by exchange and bone remodeling show no age dependence and during 1959 averaged about 0.3 $\mu\mu$g. of Sr^{90} per gram of calcium. The average value for the adult population was about 8 to 10% of what it would have been had the entire skeleton been in complete equilibrium with the Sr^{90} in the diet. Lack of equilibrium is a result of the long growth period of the human skeleton (20 years) relative to the period of weapon testing and the low rate of exchange and remodeling of old bone (3 to 4% per year). It is not possible to say to what extent the rate of increase in Sr^{90} bone concentration shown in Table I was a result of increasing integral fallout and soil uptake, since the rate of fallout was increasing also over a large portion of the observation period.

The Sr^{90} values given in Table I are averages only. One of the most important controversies regarding estimation of the potential hazards of fallout to man concerns the distribution of bone values about the mean. Just how many people may accumulate Sr^{90} bone levels 2, 5, or possibly 10 times the average cannot be estimated until the nature and shape of the population distribution curve are established adequately. The data certainly do not show a simple normal distribution. Attempts have been made (*2, 3, 4*) to estimate the fractions of the population that may be expected not to exceed specific multiples of the mean value. Present data are inadequate, however, to provide a basis for completely acceptable estimates.

Cesium-137 levels in people are easily measured by in vivo counting in very sensitive whole body counters. Total body potassium is determined simultaneously by measuring the gamma rays from the K^{40} isotope of natural potassium. Since cesium is metabolically similar to potassium, which is proportional to the body's protoplasmic mass, Cs^{137} levels are expressed as $\mu\mu$c. per gram of body potassium. Such measurements have been carried out on several hundred people per year (largely from the U.S.) since the first part of 1956. Yearly average Cs^{137} levels are given in Table I. When expressed as $\mu\mu$c. of Cs^{137} per gram of potassum, there were no significant differences between children and adults. Unlike Sr^{90}, the biological half time of Cs^{137} is relatively short (about 120 days), and equilibrium with the diet is established rather rapidly regardless of age. Detailed consideration of the data shows much less variation in Cs^{137} levels in people than is the case with Sr^{90}. The population distribution curve appears essentially normal with a standard deviation of about 36%.

Table II. Average Doses from Sr^{90} plus Cs^{137} Fallout Compared with Natural Background Radiation

(U. S. population at equilibrium)

Organ or Tissue	Natural Bkgd., Rads	Sr^{90} plus Cs^{137} Rads	Sr^{90} plus Cs^{137} % of bkgd.
Gonads[a]	3	0.05	2
Bone marrow[b]	7	0.3	4
Bone[b]	10	0.9	9

[a] Average end of reproductive age = 30 years.
[b] Average lifetime = 70 years.

Various attempts have been made to predict average equilibrium levels of Sr^{90} and Cs^{137} in children at the time of maximum fallout (about 1961) from weapon tests to date. The purpose of such predictions has been to provide a basis for estimating genetic, bone, and bone marrow radiation doses to the next generation and to postulate the potential genetic and somatic hazards of continued weapon testing and nuclear war to the world population. Most of these predictions have been based on a number of rather conservative and tenuous assumptions, paramount among which is that Sr^{90} and Cs^{137} body burdens will be proportional to the integrated surface deposition levels, and that these radioisotopes have entered the food chain largely via plant absorption from the soil. Kulp *et al.* (*2*) recently made predictions of Sr^{90} bone levels based on the assumption of a 20% soil uptake. Their estimate is approximately a factor of five lower than previous ones. Continuation of the analytical programs for another two or three years will give a more definitive answer to this very important problem.

Radiation Exposure from Fallout and Its Potential Biological Significance

Strontium-90 is potentially a somatic hazard and since it concentrates in bone and emits only β-rays the biologically significant exposures will be the 70-year integrated radiation doses to the bone and bone marrow from material fixed in the skeleton. The hazards to the population from bone and bone marrow exposure are possible increases in the incidence of bone sarcoma and leukemia, respectively. Cesium-137, however, emits both β- and γ-radiation and contributes, therefore, to radiation exposure both internally and externally. Although it will contribute also to the bone and bone marrow exposure, the 30-year integrated dose to the gonads is considered, for genetic reasons, biologically more significant. Estimates of the average 30-year gonadal dose and 70-year bone and bone marrow doses for the U. S. population (born near the time of maximum fallout) resulting from Sr^{90} plus Cs^{137} (including external radiation from the latter) are given in Table II, where they are compared with the doses expected from natural background radiation. These estimates are predicated on the assumption that level of both Sr^{90} and Cs^{137} in the diet and in people will be proportional to the integrated soil levels and that ecological equilibrium will result in the radiation dose rate decreasing in proportion only to the rate of radioactive decay. The nature of these assumptions is such that the dosage estimates are maximized, and it is quite possible that they are too high by as much as a factor of two to five.

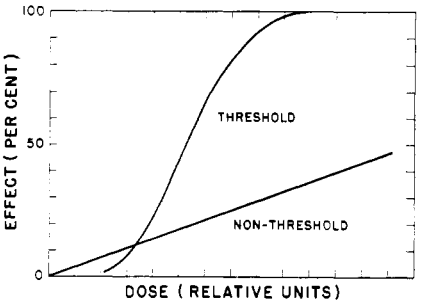

Figure 5. Threshold and nonthreshold responses to radiation exposure

Comparison of the estimated doses with the doses resulting from natural background (Table II) is important for it provides a basis for a relative measure of the potential hazards of fallout. Since man's appearance on the earth, he has lived in a radioactive environment. This so-called natural radiation background is of both cosmic and terrestrial origin—i.e., cosmic rays from outer space and their associated phenomena, and radiations from naturally occurring radionuclides distributed in the earth's crust. Carbon-14 and tritium produced by cosmic ray interactions with the earth's atmosphere and K^{40} from the soil are radioactive isotopes of essential elements in the biosphere. Other radioactive isotopes (Th^{232}, U^{238}, and their associated decay products) are widely distributed in the environment and have found their way into man as trace contaminants. Man's existence is ample proof of his ability to tolerate low levels of radiation exposure. The net result of fallout is a small increase in the radiation background to which all life is exposed. This raises the most difficult problem of all—i.e., that of trying to ascertain the significance of this small increase to the general health and well-being of the world population.

Genetic response to radiation is generally believed to be linear with dose (Figure 5) and, when delivered at conventional rates, 30 to 50 roentgens is believed to be the dose required to double the natural mutation frequency (*4*). At least with fruit fly spermatogonia, linear response has been established for

doses ranging from several thousand down to 25 roentgens. If the dose-response function continues linear and passes through the origin, any amount of radiation (no matter how small) will carry a certain probability of producing a detrimental effect and a certain fraction of the natural mutation rate must be attributed to unavoidable background exposure. The fraction of natural frequency that results from background radiation is not known. The most outstanding recent results regarding the potential genetic effects of background and fallout radiation are those reported by Russell (*3*), who found that mutation frequency in spermatogonia and oöcytes of mice was influenced by dose rate. From this extremely important discovery, it may be concluded that the 30-year dose from background and from fallout may have less genetic consequences than had been predicted previously. The discovery that mutation frequency is dose-rate dependent, however, does not invalidate the linear hypothesis, and it cannot be assumed from present knowledge that further lowering of the dose rate will result in still less potential genetic risk.

In any event, the predicted dose rate and the integrated 30-year gonadal dose from present levels of biospheric contamination are between 1 and 2% of that from natural background; therefore, the potential genetic hazard to the world population from weapon tests to date will not be more than 1 to 2% of the unavoidable burden from natural sources of radiation.

Estimation of the potential somatic consequences of biospheric contamination from weapon testing is even more difficult and uncertain than the estimation of genetic effects. The absolute somatic hazards imposed on the world population by the bone and bone marrow doses estimated in Table II are critically dependent on the shape of the dose-response curves for leukemia and bone sarcoma and, as with mutation frequency, on the extent to which they may be dose-rate dependent. If somatic response shows a curvilinear or threshold relationship with dose (Figure 5), a factor (or factors) other than 70-year integrated exposure must be important, and biological effect may drop to insignificant levels at the very low doses and dose rates received from natural background radiation and fallout. Although existence of an absolute threshold may never be proved empirically, existence of a practical one may be inferred from a curvilinear dose-response relationship. If, however, the linear proportionate hypothesis is accepted, any amount of radiation to the bone and bone marrow carries a finite probability of producing bone sarcoma and leukemia, and a fraction of the natural population incidence of these diseases must be attributed to natural background. In this case also, the controversy over population distribution of Sr^{90} bone concentrations about the mean loses much of its significance, as it is necessary to average the potential hazards over the population at risk. Only in the event of a threshold in the dose-response relationship is it of primary importance to know the fraction of the population that exceeds the mean by 2, 5, or 10 times. Figure 5 presents the alternative possibilities in a controversy which cannot be resolved at present and which may not be resolved unequivocally for many years. The answer is not known, even by the most vocal opponents or proponents of nuclear weapon testing. At present it is possible only to say from the data in Table II that if a fraction of the normal incidence of leukemia and bone cancer indeed is caused by natural radiation, weapon tests to date may increase the incidence of these diseases in the generation born during the time of maximum biospheric contamination by about 5 to 10% of that due to unavoidable natural background exposure.

Literature Cited

The proceedings of the two Congressional Subcommittee Hearings on Fallout from Nuclear Weapon Tests (referenced below) contain 4683 pages of testimony and documented material furnished for the record by dozens of participants and investigators. The reader is referred to these proceedings for additional information, for support material, and for specific credit to the individuals cited. No attempt is made to cite individual references except in those cases where publication has occurred since the hearings.

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Received for review July 14, 1960. Accepted December 15, 1960. Division of Agricultural and Food Chemistry, 137th Meeting, ACS, Cleveland, Ohio, April 1960.

PLANT UPTAKE OF RADIONUCLIDES

Availability of Exchangeable and Nonexchangeable Strontium-90 to Plants

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A PORTION of the strontium-90 in soils is not readily exchangeable with neutral salts, such as ammonium acetate or strontium nitrate (*2, 12, 14*). Since the strontium-90 in world-wide fallout is essentially water-soluble (*6*), the portion that is not now exchangeable has become so by reactions in the soil. Soluble and exchangeable cations in soils are usually readily available for uptake by plants (*10*); therefore, it is of interest to attempt to determine whether or not the nonexchangeable portion of strontium-90 in soils contributes significantly to plant uptake.

Discrimination Factor

Discrimination factors are a measure of the relative availabilities of two similar ions to plants grown in a nutrient medium. For the uptake of strontium-90 and calcium from soils, the discrimination factor is defined as:

$$\frac{\mu\mu\text{c. available } Sr^{90}}{\text{grams available Ca in soil}} \Big/ \frac{\mu\mu\text{c. } Sr^{90}}{\text{grams Ca in plant}}$$

While the measurement of strontium-90 and calcium in plants is unequivocal, the measurement of available cations in