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## **Entry of Radioactive Fallout into the Biosphere and Man**

*By Wright Langham and E. C. Anderson*

### *Introduction*

Discussion of the potential hazard of world-wide radioactive fallout from nuclear weapons tests may begin with the consideration of three basic facts.

1. The world population is receiving small exposure to radioactive materials originating from nuclear weapons testing. Fission products from bomb detonations have been and are being deposited over the surface of the earth, increasing the external gamma radiation background and finding their way into the human body through inhalation, direct contamination of food and water, and by transmission along ecological cycles from soils-to-plants-to-animals and to man.

2. Enough radiation, either from an external source or from radioactive isotopes deposited in the body, will produce deleterious effects. These effects may result in an increase in genetic mutations, shortening of life expectancy, and increased incidence of leukemia and other malignant and nonmalignant changes.

3. Radiation exposure is not a new experience for the world population. All life has been exposed to radiation since the beginning. Radiation from cosmic rays, from radioactive minerals in the earth's crust and from radium, potassium 40, carbon 14, and thorium deposited in the body constitute this so-called natural background. The amount of natural background radiation is such that persons living to an age of 70 years receive an average total dose of about 7 rem, while their skeletons (as a result of radium and other radioactive materials deposited in the bones) receive an average dose equivalent to about 10 to 12 rem. The natural background dose to some segments of the population may be at least three times the average because of variations in cosmic ray intensity and composition of the earth's crust with geographic location.

The net result of fallout is a small increase in the radiation background to which all life is exposed. The problem of the potential hazard of world-wide fallout then becomes one of trying to ascertain the magnitude and significance of this increase in background dose with regard to its potential risk to man's health and well-being.

Contamination from nuclear weapons testing may be divided on the basis of local and distant (world-wide) fallout.

Local fallout is of primary significance in the event of war in which weapons with a high fission component may be detonated at or below the surface to maximize surface contamination. In this case, fission products of short and intermediate half-life are of major concern since local fallout occurs within a few hours after detonation.

Distant (world-wide) fallout is of significance both with regard to continued weapons testing and in the event of nuclear war. Since months and even years are required for fission products to deposit over the earth's surface, only the long-lived radionuclides are important.

External exposure from environmental deposition of gamma-emitting fission products is of concern primarily because of the potential production of genetic changes. Internal exposure is of significance primarily with regard to the potential production of somatic effects in the tissues in which the various fission products deposit upon entering the body.

This report is restricted mostly to the potential internal hazard of distant (world-wide) fallout, with emphasis on  $\text{Sr}^{90}$ .  $\text{Sr}^{90}$  is believed to be the most important radionuclide because of its similarity to calcium (resulting in a high rate of uptake by plants and animals), long physical and biological half-life, and high relative fission yield. These factors lead to high incorporation in the biosphere and a long residence time in bone. General contamination will result in the bones of the population eventually reaching an equilibrium state with  $\text{Sr}^{90}$  in the biosphere.

#### *Production of Biologically Important Radionuclides from Weapons Tests*

A crude estimate of production of biologically important radionuclides from past nuclear weapons tests would be helpful in assessing the potential hazard of present biospheric contamination and in extrapolating to future levels in the event of continued testing or nuclear war.

Statements during the Subcommittee Hearings of the Joint Committee on Atomic Energy, Congress of the United States (1) assumed a constant nuclear weapons test rate of 10 megatons (MT) of fission yield per year, beginning in the spring of 1952. This leads to a total testing by all nations of about 55 MT of fission yield by mid-1957. The total estimate

may be reasonably realistic; however, the assumption of a constant test rate is highly questionable (2).

One megaton of fission energy release results in the production of about 100,000 curies of  $\text{Sr}^{90}$  (3), which suggests a total  $\text{Sr}^{90}$  production of 5.5 megacuries (MC) from weapons tests by all nations to mid-1957. From the fission yield curve (thermal neutron fission of  $\text{U}^{235}$ ) and the appropriate decay constants, it is possible to make a crude estimate of the total production of other radionuclides of potential biological importance. Table 1 shows estimates of total production (in terms of MC of initial activity) and other pertinent data for the more important intermediate- and long-lived components of weapon debris. The values for total yield are crude approximations only because it was necessary to use the fission yield curve for thermal neutron fission of  $\text{U}^{235}$ , and isotopic abundance varies with the fissionable material and the neutron energy. None of the values, however, are incorrect by more than a factor of about 2.

Table 1  
Potentially Hazardous Radionuclides in Fallout from Nuclear Detonations

Radio-nuclide	Type of radiation	Fission* abundance (%)	Radiological half-life	Total** production (MC)	Abs. on ingestion (%)	Body MPL ( $\mu\text{c}$ )
$\text{Pu}^{239}$	$\alpha$	—	24 000 years	0.3	$3 \times 10^{-3}$	0.037
$\text{Sr}^{90}$	$\beta$	5.0	27.7 years	5.5	30	1.0
$\text{Cs}^{137}$	$\beta, \gamma$	6.2	26.6 years	7.2	100	54
$\text{Pm}^{147}$	$\beta$	2.6	2.64 years	30	$1 \times 10^{-2}$	60
$\text{Ce}^{144}$	$\beta, \gamma$	5.3	285 days	200	$1 \times 10^{-2}$	5
$\text{Zr}^{95}$	$\beta, \gamma$	6.4	65 days	1100	$1 \times 10^{-2}$	26
$\text{Y}^{91}$	$\beta$	5.0	58 days	1500	$1 \times 10^{-2}$	5
$\text{Sr}^{89}$	$\beta$	4.6	51 days	950	30	4
$\text{Nb}^{95}$	$\beta, \gamma$	6.4	35 days	2000	$1 \times 10^{-2}$	76
$\text{Ba}^{140}$	$\beta, \gamma$	6.0	13 days	5000	5	4
$\text{I}^{131}$	$\beta, \gamma$	2.8	8 days	4000	100	0.7

\* Slow neutron fission of  $\text{U}^{235}$ ; abundance in weapon debris is somewhat different.

\*\* Total initial activity in megacuries produced by all weapons test to mid-1957.

The total production of  $\text{Pu}^{239}$  was estimated from the report of *Stewart, Crooks and Fisher* (4), who postulated from analysis of bomb debris that one  $\text{Pu}^{239}$  atom was formed per fission by neutron interaction with bomb components. Since 1 KT of fission yield is produced by  $1.4 \times 10^{23}$  fissions (3), each of which results in the production of a  $\text{Pu}^{239}$  atom, 55 MT of fission would produce 0.2 MC of  $\text{Pu}^{239}$ . Other isotopes of plutonium, when converted to equivalents of  $\text{Pu}^{239}$ , bring the total production to



about 0.3 MC equivalents. The production values given in table 1 are not a measure of the relative biological importance of the various nuclides, but merely provide some general idea of the relative initial activities produced by all weapons tests through mid-1957. Development of sufficiently sensitive detectors should result eventually in detection of most of these radionuclides in foods and man.  $\text{Sr}^{90}$  (5, 6),  $\text{Cs}^{137}$  (7), and  $\text{I}^{131}$  (8) have been measured quantitatively in the human body, and the presence of  $\text{Ce}^{144}$  in pooled urine samples has been reported (9). In addition,  $\text{Ba}^{140}$  (10) and  $\text{Sr}^{89}$  (11) have been observed in milk, and other radionuclides have been detected in air and other materials composing man's environment. The extent to which they pose a potential threat to man's health and well-being depend on their rate of production and on their individual physical and biological properties.

### *Distribution of Fallout from Nuclear Detonations*

#### *Postulated Mechanisms of Distribution*

Libby (5, 12) was first to propose a model explaining fallout and distribution of atomic debris from nuclear weapons detonations. His model is based on three kinds of fallout—local, tropospheric, and stratospheric.

Local fallout is deposited in the immediate environs of the explosion during the first few hours. This debris consists of the large particles from the fireball and includes partially or completely vaporized residues from the soil and structures which are swept into the cloud.

Tropospheric fallout consists of that material injected into the atmosphere below the tropopause which is not coarse enough to fall out locally. This debris is sufficiently fine that it travels great distances, circling the earth from west to east in the general latitude of the explosion, until removed from the atmosphere (with a half-time of 20 to 30 days) by rain, fog, contact with vegetation, and other meteorological and/or physical factors.

Stratospheric fallout is composed of fission products that are carried above the tropopause and can result only from large weapons (of the order of 1 MT and greater). Libby (13a, b) has postulated that atomic debris, once it is injected above the tropopause, is mixed rapidly throughout the stratosphere and falls back uniformly into the troposphere with a half-time of about 7 years. As it returns to the troposphere, it is deposited over the earth's surface in relation to meteorological conditions. He attributed the higher  $\text{Sr}^{90}$  soil concentrations in the United States to meteorological conditions and to local and tropospheric fallout as a result of the proximity of the Nevada Test Site. The generally higher

concentrations in the north temperate latitudes were attributed to prevailing meteorological conditions and their effects on tropospheric fallout from tests in the USSR and at the United States Pacific Proving Grounds.

*Machta* (14) proposed a model of stratospheric fallout which differs in some respects from *Libby's*. He postulated that stratospheric mixing is slow and that stratospheric distribution of fission products is still non-uniform. He feels that a major portion of the nuclear debris is still in the northern portions of the northern hemisphere, rather than uniformly spread over the entire globe or even uniformly dispersed in the northern hemisphere itself. He feels also that stratospheric movement of the fission products is largely by direct transport from west to east in the general latitude of the point of injection with very slow vertical mixing. Slow polewards circulation of stratospheric air from equatorial regions provides some mixing towards the poles. The higher concentration of fallout in the temperate latitudes is explained on the basis of air exchange between the stratosphere and troposphere through the break in the tropopause in the vicinity of the jet streams. A large part of the higher concentration of  $\text{Sr}^{90}$  found in the northern part of the United States may result from preferential stratospheric leakage in the vicinity of  $30^\circ \text{ N}$  to  $40^\circ \text{ N}$  latitude instead of the proximity of the Nevada Test Site. Qualitatively, both models predict the same general distribution of fallout. Quantitatively, the *Machta* model predicts a greater degree of nonuniformity of fallout over the earth with higher deposition of fission products in the north and south temperate latitudes from nuclear debris still in the stratospheric reservoir. Fig. 1 shows the essential features of the *Machta* model and the present general world-wide surface distribution pattern of  $\text{Sr}^{90}$ .

#### *Average Maximum Surface Deposition Levels*

*Present Levels (1956-1957).*—A crude indication of latitudinal distribution of the integrated  $\text{Sr}^{90}$  surface deposition levels as of June 1956, derived from soil data, is shown by the lower curve in fig. 2. This curve is essentially the same as the one given by *Machta* (14) except a few points have been added and the peak concentration in the north temperate latitudes is drawn slightly higher to allow some weighting for average  $\text{Sr}^{90}$  levels in United States soils. These data suggest a level of about  $13 \text{ mc/mi}^2$  for the north temperate latitudes. No soil data are available yet for mid-1957. Fallout data from pot collections in New York and Pittsburgh, however, showed that cumulative  $\text{Sr}^{90}$  fallout increased by about 50 per cent from June 1956 to June 1957 (15). The upper curve in fig. 2 represents estimated latitudinal fallout distribution

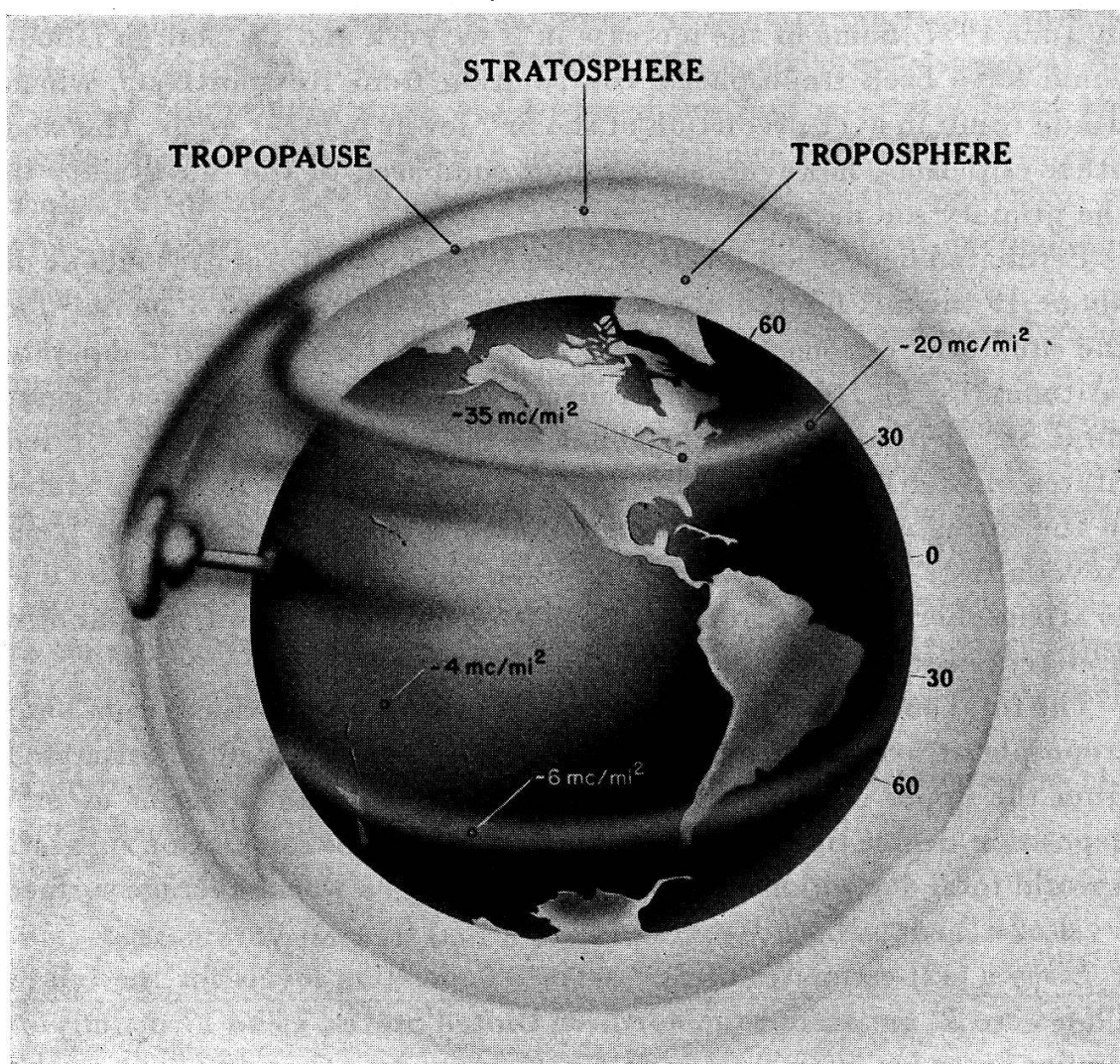


Fig. 1. Mechanism of distribution of world-wide fallout.

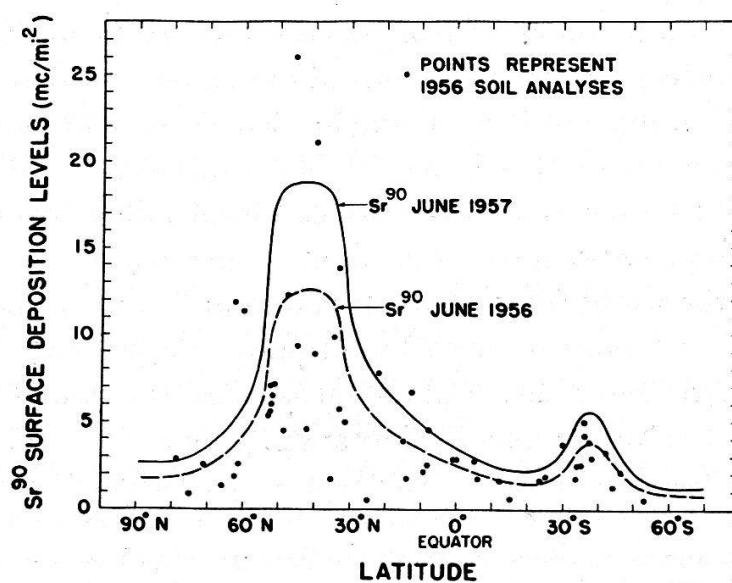


Fig. 2. Surface deposition levels of  $\text{Sr}^{90}$  from soil analyses.

in June 1957. Some of the increase in New York and Pittsburgh fallout could have been tropospheric contribution from Russian tests, which would result in over-prediction of the  $\text{Sr}^{90}$  levels in other areas. This and other criticisms, however, seem minor compared to the uncertainty in the primary soil data.

Estimated deposition levels in June 1957 show a total  $\text{Sr}^{90}$  fallout of about 19 mc/mi<sup>2</sup> for the north temperate latitudes, 3 to 4 mc/mi<sup>2</sup> for the equatorial regions, and about 5 to 6 mc/mi<sup>2</sup> for the south temperate latitudes (fig. 1). Data from pot collections in the New York area suggest total  $\text{Sr}^{90}$  deposition levels of about 35 mc/mi<sup>2</sup> in the northern United States in mid-1957. The rapid build-up of  $\text{Sr}^{90}$  in the northern states in the spring of 1957 cannot be attributed to tropospheric fallout from Nevada tests, since Operation Plumbbob had not begun. It may be due to tropospheric fallout from spring test operations in the USSR and to preferential stratospheric fallout from past tests.

The total amount of  $\text{Sr}^{90}$  deposited over the earth's surface (from both tropospheric and stratospheric fallout) as of mid-1957 can be estimated from the upper curve in fig. 2 by replotting the data in terms of  $\text{Sr}^{90}$  deposition/degree times the earth's area/degree. This calculation suggests a world total deposition of 1.64 MC, which gives a world average surface level of 8.2 mc/mi<sup>2</sup>.

Libby's (12) estimates of  $\text{Sr}^{90}$  surface deposition levels for the fall of 1956 were 22 mc/mi<sup>2</sup> for the northern United States, 15 to 17 mc/mi<sup>2</sup> for similar latitudes elsewhere<sup>1</sup>, and 3 to 4 mc/mi<sup>2</sup> for the rest of the world. These values are in good agreement with those estimated from the lower curve of fig. 2. He also estimated the stratospheric reservoir at about 2.4 MC (24 MT equivalents of fission yield). His predictions were based on 1955 soil analyses, his model of tropospheric and stratospheric fallout, and a general knowledge of the megatons of fission devices detonated during the spring and summer of 1956. Using his deposition values and the world  $\text{Sr}^{90}$  production up to that time, 24 MT equivalents still in the stratosphere would be possible only provided local fallout from surface detonation of megaton weapons was about 25 per cent.

Values for the world total production (about 5.5 MC to mid-1957) and deposition of  $\text{Sr}^{90}$  may be used to estimate the present magnitude of the stratospheric reservoir. Fallout measurements from United States megaton detonations in the Pacific suggest that  $50 \pm 17$  per cent of fission debris falls out locally. The rest is partitioned between tropo-

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<sup>1</sup> The north temperate fallout band was indirectly defined as the region between 60° N and 10° N latitude. It is assumed that the surface deposition of 16 to 17 mc/mi<sup>2</sup> applies to this area.



spheric and stratospheric fallout. Since the fallout time of tropospheric debris is of the order of 20 to 30 days, the material not accounted for by local fallout plus total world-wide deposition must still be in the stratospheric reservoir. Such a material balance calculation estimates the stratospheric  $\text{Sr}^{90}$  content at  $1.11 \pm 0.93$  MC, or the equivalent of  $11.1 \pm 9.3$  megatons of fission yield. The average is about one-half the value estimated by *Libby* (12).

High altitude air sample measurements suggest that considerable specific fractionation of fission debris is occurring. If, however, it is assumed that serious fractionation of  $\text{Sr}^{90}$ ,  $\text{Pu}^{239}$ , and  $\text{Cs}^{137}$  does not occur (12), it is possible to estimate the general distribution of these nuclides in relation to  $\text{Sr}^{90}$ . Since their radiological half-lives are long compared to the period of testing and the stratospheric storage time, their general distribution should be in direct ratio to their total production relative to that of  $\text{Sr}^{90}$  (table 1). On this basis, average maximum surface deposition levels of  $\text{Pu}^{239}$  and  $\text{Cs}^{137}$  in the north temperate latitudes (by mid-1957) would be about 1.2 and 25 mc/mi<sup>2</sup>, respectively.

Present world-wide distribution of  $\text{Cs}^{137}$  and  $\text{Pu}^{239}$ , estimated on the above basis, are compared with  $\text{Sr}^{90}$  in table 2.

Table 2  
Comparison of World-Wide Distribution of  $\text{Sr}^{90}$ ,  $\text{Cs}^{137}$ , and  $\text{Pu}^{239}$  from Nuclear Detonations\*

Region	Mid-1957		
	$\text{Sr}^{90}$	$\text{Cs}^{137}$	$\text{Pu}^{239}$
Northern U.S.A.	35 mc/mi <sup>2</sup>	46 mc/mi <sup>2</sup>	2.1 mc/mi <sup>2</sup>
North temperate latitudes	19 mc/mi <sup>2</sup>	25 mc/mi <sup>2</sup>	1.2 mc/mi <sup>2</sup>
South temperate latitudes	5-6 mc/mi <sup>2</sup>	7 mc/mi <sup>2</sup>	0.3 mc/mi <sup>2</sup>
Rest of world	3-4 mc/mi <sup>2</sup>	5 mc/mi <sup>2</sup>	0.2 mc/mi <sup>2</sup>
World average	8 mc/mi <sup>2</sup>	10 mc/mi <sup>2</sup>	0.45 mc/mi <sup>2</sup>
Total surface deposition	1.64 MC	2.1 MC	0.10 MC
Stratospheric reservoir	1.10 MC	1.4 MC	0.06 MC

\* Assuming no fractionation.

The estimated levels are general averages only and assume no fractionation and uniform distribution within the respective areas. Actually, this general picture is greatly oversimplified. Some fractionation is indicated by air sampling data and once fission products are suspended in the troposphere (either directly from the detonation or from stratospheric leakage, regardless of mechanism), meteorological conditions play a major role in their surface distribution. *Libby* has stressed the importance of rainfall, snow, fog, and mist (5, 12). Within any major area

fluctuations in levels of surface deposition may occur which correlate with local meteorological conditions. *Machta* (14) has guessed that areas as large as milk-sheds may not have more than two to three times the average deposition for the latitude. He points out, however, that desert areas where there is practically no rainfall may have almost zero fallout.

#### *Future Levels (Assuming No More Tests)*

Fallout of  $\text{Sr}^{90}$  and other long-lived radionuclides from the stratospheric reservoir will continue even if weapons tests are stopped. Whether the integrated surface deposition levels continue to build up will depend on whether the rate of stratospheric fallout more than compensates for the rate of decay of material already on the ground.

From the surface deposition levels in table 2 and the value of  $1.1 \pm 0.9$  MC  $\text{Sr}^{90}$  for the stratospheric reservoir, estimation of future deposition levels, assuming no more weapons tests, is possible.

If  $M(t)$  is the surface deposition level and  $Q(t)$  is the stratospheric storage in mc/mi<sup>2</sup> at any time, the rate of change of the surface deposition level is:

$$\frac{dM(t)}{dt} = -\lambda M(t) + kQ(t)$$

where  $\lambda$  is the radioactive decay constant of  $\text{Sr}^{90}$ , and  $k$  is the stratospheric fallout rate constant (assumed to be first order). If  $\lambda M(t) = kQ(t)$ ,  $dM/dt$  is zero. In this case, additional stratospheric fallout just compensates for radioactive decay, and  $M(t)$  does not change. Such an equilibrium state is transitory, since  $Q(t)$  is constantly decreasing (both by decay and by fallout). Loss by radioactive decay in  $M(t)$ , therefore, soon exceeds gain from  $Q(t)$ , and  $M(t)$  falls. If  $\lambda M_0$  is greater than  $kQ_0$  (where  $M_0$  and  $Q_0$  are the concentrations at  $t = 0$ , the time of cessation of tests), the latter situation already exists and the ground level begins to fall when testing stops. Only if  $\lambda M_0$  is less than  $kQ_0$  will additional fallout from the stratosphere exceed the decay of the ground contamination and the surface deposition level continue to rise. If the mean times of decay and fallout are 40 and 10 years, respectively,  $Q(t)$  must be at least  $\frac{1}{4} M(t)$  for surface deposition to increase.

Future levels, in the event of no more testing, can be estimated if it is assumed that fallout in the future will have the same degree of non-uniformity as in the past. In this case, the effective stratospheric storage ( $Q(t)_e$ ) for a given area is related to the average stratospheric storage ( $Q(t)_{av}$ ) by the equation:

$$Q(t)_e = \frac{M(t)}{M(t)_{av}} Q(t)_{av}$$



where  $M(t)$  is the observed ground concentration in the area in question, and  $M(t)_{av}$  is the averaged world-wide ground concentration. On the basis of this assumption, the soil levels increase everywhere by the same ratio and reach a maximum about 1963, which is some 10 per cent higher than present levels.

Assuming uniform stratospheric fallout, some areas do not increase since the additional stratospheric fallout is insufficient to compensate for radioactive decay. The time of maximum ground concentration (where it does occur) varies also with location, being about 1966 in the south temperate latitudes and 1969 elsewhere.

Neither method of estimation is strictly correct. The assumption of uniform fallout may underestimate build-up in the northern latitudes, and the assumption of nonuniformity of future fallout according to the past may tend to overestimate build-up in those areas where some of the material deposited in the past came from tropospheric fallout. As stated by *Machta* (14), it is hoped that the truth lies somewhere in between. It must also be kept in mind that the stratospheric reservoir may well be 2.4 MC as estimated by *Libby* (12).

Future  $\text{Cs}^{137}$  levels, assuming no fission product fractionation and no more tests, will be about 1.3 times higher than the corresponding  $\text{Sr}^{90}$  levels since their radiological half-lives are essentially the same.  $\text{Pu}^{239}$  levels will continue to rise for several years because of its 24 000-year half-life. In this case,  $\lambda M_0$  will be less than  $kQ_0$  until the stratospheric reservoir is essentially depleted. However, surface deposition levels will not increase more than 0.6, which is the ratio of the present total surface deposition to the estimated stratospheric reservoir.

Predicted average maximum surface deposition levels of  $\text{Sr}^{90}$ ,  $\text{Cs}^{137}$ , and  $\text{Pu}^{239}$  (assuming nonuniform fallout and cessation of tests) are given in table 3.

Table 3  
Predicted Average Maximum Surface Deposition Levels of  $\text{Sr}^{90}$ ,  $\text{Cs}^{137}$ , and  $\text{Pu}^{239}$   
(assuming no more weapons tests after mid-1957)

Region	$\text{Sr}^{90}$	$\text{Cs}^{137}$	$\text{Pu}^{239}$
	(mc/mi <sup>2</sup> )*	(mc/mi <sup>2</sup> )*	(mc/mi <sup>2</sup> )**
Northern U.S.A.	39	51	3.3
North temperate latitudes	21	27	2.0
South temperate latitudes	6	8	0.5
Rest of world	4	5	0.3
World average	9	12	0.8

\* Maximum will be reached in about 1965.

\*\* Maximum will be reached essentially in about 30 years.

Surface deposition levels of other biologically significant isotopes, which all have half-lives short compared to the stratospheric storage time and for which  $\lambda M_0$  is already greater than  $kQ_0$ , will begin decreasing immediately when weapons tests are stopped.

#### *Future Levels (With Continued Testing)*

If weapons tests continue at a constant rate (in terms of fission yield), the decay of radionuclides in the biosphere will eventually equal the rate of production, and continued testing will result in no further increase in deposition levels. At the present rate of testing (assumed to be 10 MT of fission per year for the past 5 years), equilibrium  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$  levels will be reached in about 100 years. Isotopes with shorter half-lives will reach equilibrium sooner.  $\text{Pu}^{239}$  obviously will continue to increase essentially in proportion to its total production.

*Campbell* (16) and *Stewart et al.* (4) have estimated surface deposition levels of  $\text{Sr}^{90}$  at equilibrium with a uniform test rate, and their calculations suggest levels about 30 times the present values. Their equations are derived, however, from stratospheric fallout and apply to ground levels due to the stratospheric component only.

*Libby* (17) estimated surface build-up on the basis of total levels on the ground at  $t = 5$  years and predicted equilibrium levels 11 times the present values. His calculations have been checked by *Neuman* (18) and others. *Libby* also assumed that about 30 per cent of the  $\text{Sr}^{90}$  (over the long period required for equilibrium) would become unavailable to plants and the available equilibrium levels would be only about 8 times the present values (1). Attempts are being made to obtain actual yearly fission product production rates to refine further predictions of surface levels under continued testing. Until then, an equilibrium build-up factor for  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$  of about 10 with a continued average test rate of 10 MT of fission yield per year seems reasonable. Table 4 shows future average maximum surface deposition levels of  $\text{Sr}^{90}$ ,  $\text{Cs}^{137}$ , and  $\text{Pu}^{239}$  calculated, on the above basis, from the data in table 2.

Others have made similar estimates of  $\text{Sr}^{90}$  surface deposition levels. *Libby* (19) estimated equilibrium levels for the United States at 400 to 600 mc/mi<sup>2</sup>. *Neuman* (18) estimated a United States deposition level of about 400, and *Machta* (14) 350 to 850 mc/mi<sup>2</sup>.

#### *Incorporation of Nuclear Debris into the Biosphere and Man*

Radionuclides from fallout may enter the body through inspiration of the contaminated atmosphere and by ingestion of contaminated food and water.

Table 4  
Average Maximum Surface Deposition Levels of  $\text{Sr}^{90}$ ,  $\text{Cs}^{137}$ , and  $\text{Pu}^{239}$   
(assuming a continuing test rate of 10 MT of fission yield per year)

Region	$\text{Sr}^{90}$	$\text{Cs}^{137}$	$\text{Pu}^{239}$
	(mc/mi <sup>2</sup> )*	(mc/mi <sup>2</sup> )*	(mc/mi <sup>2</sup> )**
Northern U.S.A.	350	460	40
North temperate latitudes	190	250	24
South temperate latitudes	55	70	6
Rest of world	35	50	3
World average	80	100	11

\* At equilibrium in about 100 years.

\*\* In about 100 years, not at equilibrium.

*Stewart et al.* (4) estimated the mean  $\text{Sr}^{90}$  and  $\text{Pu}^{239}$  concentrations in air at ground level in England during 1952 to 1955 as  $4 \times 10^{-16}$ \* and  $3 \times 10^{-17}$   $\mu\text{c/cc}$ , respectively. Assuming the ratio of  $\text{Cs}^{137}/\text{Sr}^{90}$  in air is the same as their ratio of total production, the mean  $\text{Cs}^{137}$  concentration in air during the same period would be  $5 \times 10^{-16}$   $\mu\text{c/cc}$ . The respective occupational maximum permissible air concentration of  $\text{Sr}^{90}$ ,  $\text{Pu}^{239}$ , and  $\text{Cs}^{137}$  recommended by the International Commission on Radiological Protection (21) are  $2 \times 10^{-10}$ ,  $2 \times 10^{-11}$ , and  $2 \times 10^{-7}$   $\mu\text{c/cc}$ . The estimated mean values are 5 to 8 orders of magnitude lower than the maximum permissible air concentrations recommended for the general population.

Since the tropospheric fallout time is 20 to 30 days, the mean air concentration values during 1952 to 1955 probably approximate equilibrium conditions with the past 5-year rate of biospheric contamination from stratospheric fallout (20). In this case, continued weapons tests at the past rate should not increase the mean air concentrations greatly. As suggested by *Stewart et al.* (4) and *Bryant et al.* (11), inhalation of nuclear debris is not a major factor in the potential hazards of world-wide fallout.

Comparison of measured and estimated concentrations of the principal long-lived radionuclides in water with the maximum permissible concentrations recommended by the International Commission on Radiological Protection (21) suggest also that ingestion of contaminated drinking water is relatively unimportant (11).

Ingestion of food contaminated through soil integration and plant uptake of long-lived radionuclides seems to pose the major potential hazard.

When nuclear debris is deposited on the earth's surface and incorpo-

\* Their calculated value agrees reasonably well with the average measured value of  $3 \times 10^{-15}$   $\mu\text{c/cc}$  (for the same period at Washington, D.C.) reported by *Martell* (20).

rated in the soil, the individual nuclides are taken into plants through the root system according to their individual soil-plant relationships. That which settles directly on vegetation may remain as surface contamination or may enter the plant through foliate absorption. When plants are eaten by animals, the radioactivity incorporated in the plants or deposited on their surfaces is absorbed and retained by the animal according to the specific metabolic characteristics of the individual nuclides. When plant and animal products are eaten by man, the radioelements they contain are absorbed and incorporated into his tissues, again in accordance with their individual metabolic properties.

A few of the long-lived radionuclides in nuclear debris will be considered individually, since their accumulation in the soil and ecological transport to man appear to be the major concern.

### *Strontium 90*

*Ecological Incorporation and Discrimination.*— $\text{Sr}^{90}$  is chemically and metabolically similar to calcium. Therefore, it is incorporated into the biosphere along the same ecologic chain. It is taken into plants through the root system in relation to available soil calcium and absorbed and deposited in human bone in relation to the  $\text{Sr}^{90}/\text{Ca}$  ratio in the diet.

It is reasonable to assume that strontium may be discriminated against with respect to calcium in passing along the ecological chain. For example, the  $\text{Sr}^{90}/\text{Ca}$  ratio of human bones may be expected to be lower than that of soil. Attempts have been made to determine the over-all  $\text{Sr}^{90}/\text{Ca}$  discrimination ratio in going from soils to human bone by determining the individual discrimination factors ( $DF$ ) that occur at the various steps along the ecological cycle. *Menzel* (22) obtained a soil-to-plant discrimination factor ( $DF_1$ ) of 0.7 for four widely different soil types using both radioactive and stable strontium. *Larson* (23) and *Bowen and Dymond* (24) obtained comparable values.

A discrimination factor ( $DF_2$ ) of 0.13 in going from plants-to-milk has been reported by *Alexander et al.* (25) and *Comar* (26), and the discrimination factors ( $DF_3$ ) from plants-to-bone and from milk-to-bone ( $DF_4$ ) have been estimated at 0.25 (27, 28).

The over-all discrimination ratio ( $OR_{\text{bone-soil}}$ ) in going from soil-to-human bone via the diet may be estimated from the various discrimination factors and the fraction of dietary calcium derived from dairy products and from other sources. For example, for the United States population the amount of dietary calcium derived from dairy products is estimated at about 80 per cent. The remainder is derived from cereals, vegetables, meats, etc. On the basis of the above generalizations,

$$\begin{aligned}
 (OR_{bone-soil}) &= (0.8 \times DF_1 \times DF_2 \times DF_3) \\
 &+ (0.2 \times DF_1 \times DF_4) = (0.8 \times 0.7 \times 0.13 \times 0.25) + \\
 &+ (0.2 \times 0.7 \times 0.25) = 0.05
 \end{aligned}$$

and indicates that the average equilibrium concentration of  $Sr^{90}$  in bone calcium for the United States population will be about 5 per cent of the concentration in the available soil calcium (fig. 3).

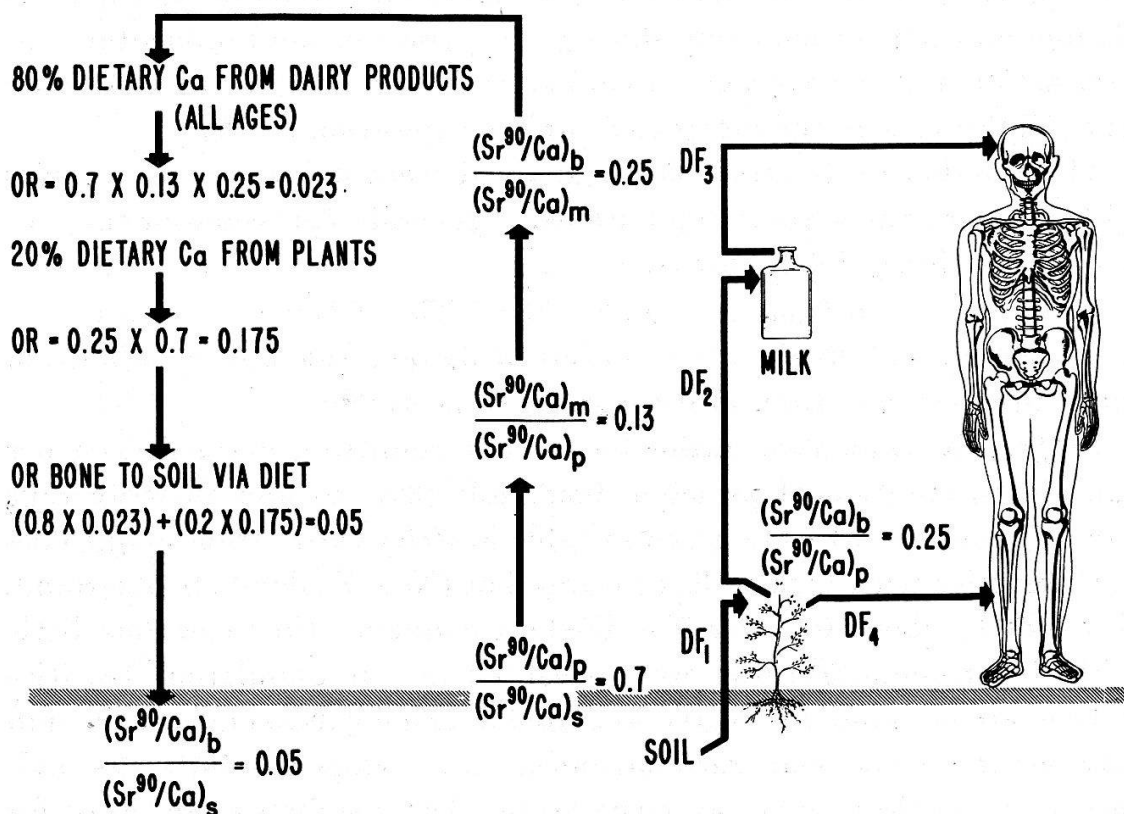


Fig. 3. Ecological discrimination against  $Sr^{90}$  with respect to calcium (United States).

It should be emphasized that over-all discrimination ratios derived in the above manner apply only to passage along the ecological chain. The ecological discrimination ratio automatically assumes that calcium and strontium are uniformly mixed in soil to the average depth of the plant feeding zone. No allowance is made for direct foliar contamination, for dilution with a greater reservoir of available soil calcium through plowing, for the possibility that it may become less available with time through soil binding and leaching, or for differences in uptake by different plant species.

**$Sr^{90}$  Levels in Bones of the Population.**—Present and future average maximum  $Sr^{90}$  equilibrium levels in bones of the population can be estimated from the soil-to-bone discrimination ratio, the ratio of milk to other sources of calcium in the diet, and the present and predicted average maximum surface deposition levels given in the previous sections.



Assuming an average of 20 g available Ca/ft<sup>2</sup> of soil to a depth of 2.5 inches, 1 mc of Sr<sup>90</sup>/mi<sup>2</sup> is equivalent to 1.8  $\mu\mu\text{c}$  Sr<sup>90</sup>/g available soil calcium. If all of the Sr<sup>90</sup> is in available form, multiplication of the surface deposition levels by 1.8 gives the Sr<sup>90</sup> activity per gram of available soil calcium. Multiplication of the specific activity of available soil calcium by the Sr<sup>90</sup> discrimination ratio should give the average maximum specific activity of calcium laid down in the adult skeleton through exchange and bone remodeling during the period of environmental contamination and the average maximum Sr<sup>90</sup> concentration in a skeleton at equilibrium with the integrated surface deposition levels.

The fraction of dietary calcium derived from dairy products varies widely among the various populations. A general expression for the ecological discrimination factor is:

$$(OR_{\text{bone-soil}}) = (M_f \times 0.025) + (R_f \times 0.175)$$

in which  $M_f$  and  $R_f$  are the fractions of dietary calcium derived from dairy products and from other sources, respectively.

Sr<sup>90</sup>/Ca discrimination ratios for various countries, derived from per capita consumption of principal foodstuffs (29, 30) and their average calcium content (31), are given in table 5. ( $OR$ ) varies from about 0.04 for countries with high milk consumption (New Zealand, Switzerland, Sweden) to about 0.15 for Far Eastern countries that consume little milk. Discrimination ratios were weighted for the population densities of the various countries to give weighted average values of 0.1, 0.06, and 0.12 for the north temperate latitudes, south temperate latitudes and, rest of the world population, respectively. The discrimination ratios for the various areas are only superficially adjusted for differences in population dietary habits and make no allowance for individual variations in calcium metabolism and for the fraction of Sr<sup>90</sup> entering the food chain through direct fallout on vegetation. They may be conservative, however, because they are derived on the basis of complete availability of the deposited Sr<sup>90</sup> and on the assumption that all of man's dietary calcium comes from the top 2.5 inches of the soil.

Table 5  
Sr<sup>90</sup>/Ca Discrimination Ratios for Various Countries Derived from  
*per capita* Consumption of Principal Foodstuffs

Country	$M_f$	$R_f$	$OR$
Algeria	0.69	0.31	0.060
Argentina	0.79	0.21	0.055
Australia	0.82	0.18	0.050
Austria	0.85	0.15	0.046
Belgium-Luxembourg	0.75	0.25	0.061



Table 5 (Continued)

Country	M <sub>f</sub>	R <sub>f</sub>	O <sub>R</sub>
Brazil	0.60	0.40	0.084
Bulgaria	0.54	0.46	0.085
Burma	0.33	0.67	0.125
Canada	0.85	0.15	0.046
Chile	0.67	0.33	0.072
China	0.23	0.77	0.140
Columbia	0.67	0.33	0.072
Cuba	0.67	0.33	0.072
Czechoslovakia	0.71	0.29	0.067
Denmark	0.79	0.21	0.055
Egypt	0.57	0.43	0.088
Finland	0.84	0.16	0.047
France	0.75	0.25	0.061
Germany	0.74	0.26	0.063
Greece	0.63	0.37	0.079
Hungary	0.53	0.47	0.094
India	0.51	0.49	0.097
Indochina	0.16	0.84	0.151
Indonesia	0.11	0.89	0.158
Italy	0.62	0.38	0.081
Ireland	0.75	0.25	0.061
Israel	0.73	0.27	0.064
Japan	0.18	0.82	0.148
Malaya	0.19	0.81	0.146
Mexico	0.56	0.44	0.090
Morocco	0.75	0.25	0.061
Netherlands	0.83	0.17	0.049
New Zealand	0.88	0.12	0.041
Norway	0.86	0.14	0.044
Pakistan	0.72	0.28	0.066
Peru	0.41	0.59	0.113
Philippines	0.18	0.82	0.148
Poland	0.55	0.45	0.091
Portugal	0.30	0.70	0.123
Rhodesia	0.41	0.59	0.113
Rumania	0.59	0.41	0.085
Spain	0.50	0.50	0.099
Sweden	0.87	0.13	0.043
Switzerland	0.87	0.13	0.043
Thailand	0.55	0.45	0.091
Turkey	0.37	0.63	0.119
Union of South Africa	0.71	0.29	0.067
United Kingdom	0.81	0.19	0.052
United States	0.80	0.20	0.053
Uruguay	0.82	0.18	0.050
Venezuela	0.75	0.25	0.061
Yugoslavia	0.67	0.33	0.072

Average maximum Sr<sup>90</sup> equilibrium bone levels in the world's population postulated from ecological considerations are given in table 6.

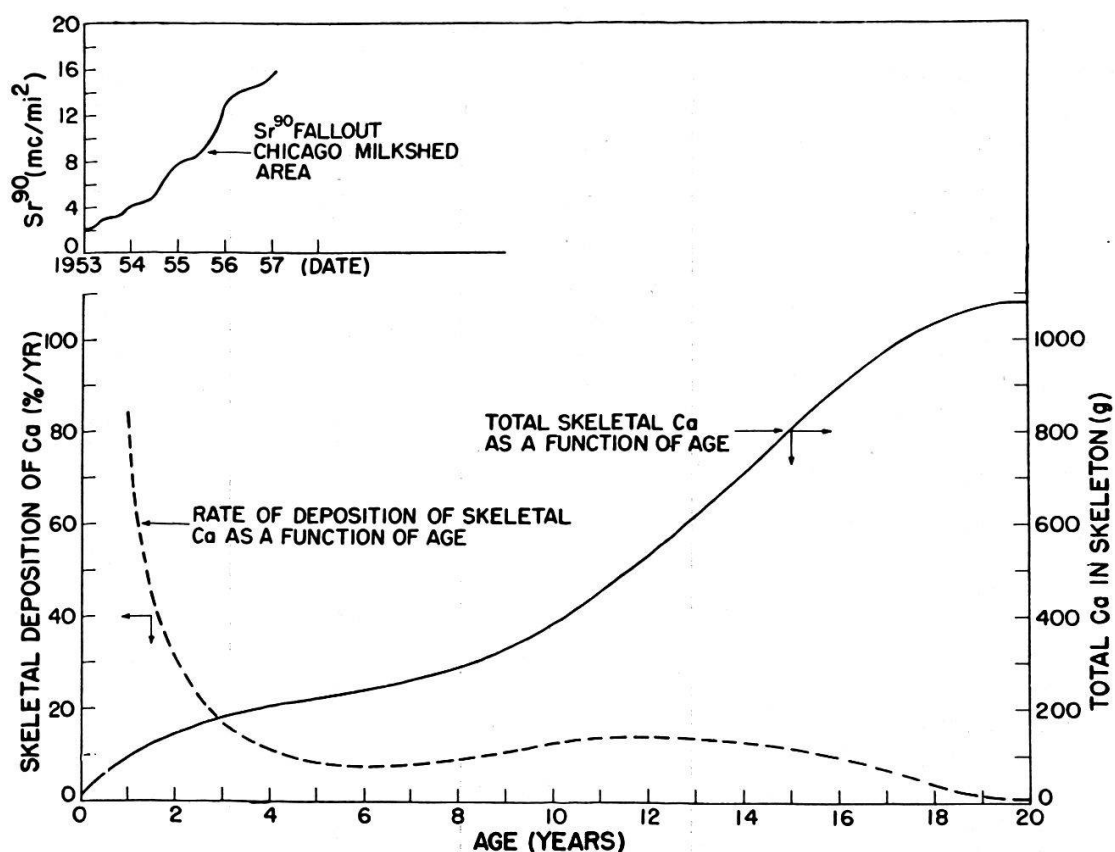


Fig. 4. Rate of skeletal accretion in relation to rate of environmental contamination.

These data suggest the average maximum level of  $\text{Sr}^{90}$  in the bones of the population of the United States would be about  $3.1 \mu\mu\text{c/g Ca}$ , if they were in ecological equilibrium with the 1957 soil deposition levels. The average of the north temperate population belt would be about the

Table 6  
Postulated Average Maximum Equilibrium  $\text{Sr}^{90}$  Bone Levels in the World Population ( $\mu\mu\text{c/g Bone Ca}$ )

	Mid-1957		About 1963*		About 2050**	
	Ecol. data	Bone data	Ecol. data	Bone data	Ecol. data	Bone data
United States	3.1	1.7	3.5	1.9	31	17
North temperate latitude	3.2	1.7	3.6	1.9	32	17
South temperate latitude	0.6	0.5	0.7	0.6	6	5
Rest of world	0.8	0.3-0.5	0.9	0.5-0.8	8	3-5
World average***	(2.8)	(1.5)	(3.1)	(1.7)	(28)	(15)

\* Assuming no more weapons tests.

\*\* At equilibrium with a continued test rate of 10 MT equivalents of fission per year.

\*\*\* Population weighted average.

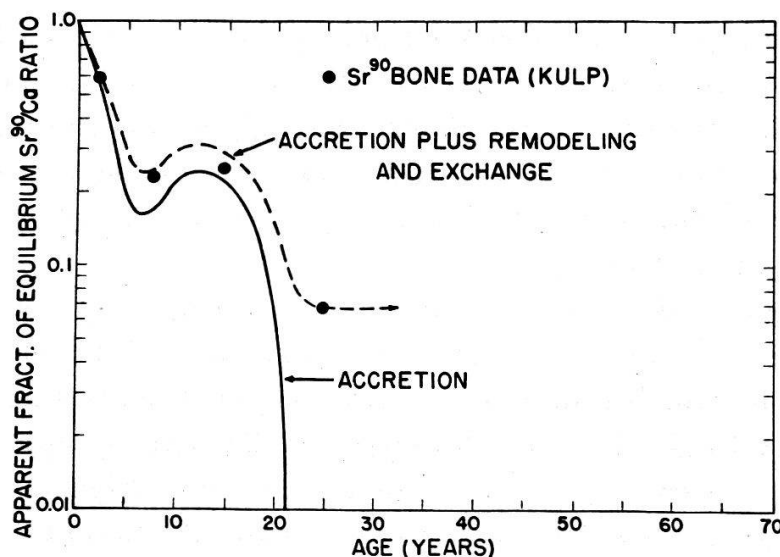


Fig. 5. Apparent fraction of equilibrium  $\text{Sr}^{90}/\text{Ca}$  ratio in relation to skeletal age.

same as the United States because of the lower ratio of milk to cereals in the diet of the heavily populated countries of the Far East. The population weighted world average is only slightly lower than the average for the north temperate latitude, which is not surprising since over 80 per cent of the world's population lives in that region.

An alternative method of estimating average maximum equilibrium bone levels involves the use of current  $\text{Sr}^{90}$  bone analyses, by adjusting the data for the pronounced variation in  $\text{Sr}^{90}/\text{Ca}$  ratio in bone as a function of skeletal age. *Langham* and *Anderson* (32) estimated the fraction of  $\text{Sr}^{90}/\text{Ca}$  skeletal equilibrium from the rate of skeletal accretion (33) and the rate of increase in integrated fallout shown in fig. 4. It was assumed that each yearly increment of skeletal growth contains  $\text{Sr}^{90}$  at a concentration corresponding to the  $\text{Sr}^{90}$  build-up in the biosphere for that year. For a first approximation, the skeleton was regarded as a unit and the  $\text{Sr}^{90}$  burden averaged over the entire skeleton.

Calculated values for the apparent fraction of equilibrium  $\text{Sr}^{90}/\text{Ca}$  ratio as a function of age, based on skeletal growth rate alone and a yearly doubling time of the  $\text{Sr}^{90}$  level are shown by the solid curve of fig. 5. The points represent *Kulp's* 1955–1956 data (6) normalized to the 0- to 4-year age group as representing 59 per cent of equilibrium  $\text{Sr}^{90}$  concentration.

At age 24 (4 years beyond the age at which skeletal growth stops) these data show that 7 to 10 per cent of the skeletal calcium was involved in bone remodeling plus exchange during the period of environmental contamination. If an equivalent fraction of the skeletal calcium of growing subjects is involved in exchange plus remodeling, then the  $\text{Sr}^{90}$  levels in children would be proportionally higher (dashed line, fig. 5)

than the curve based on skeletal calcium accretion alone. This indeed appears to be the case and indicates that the major factors have been considered in constructing the model. The upper curve in fig. 5 permits the use of adequate bone data from any age group to predict the average maximum equilibrium  $\text{Sr}^{90}$  bone level and indicates a value of  $0.9 \mu\mu\text{c/g Ca}$  by the end of 1955.

$\text{Sr}^{90}$  content of skeletons of stillborns (12) during 1955 averaged about  $0.5 \mu\mu\text{c/g Ca}$ , which gives an average maximum equilibrium level of 1.0 when the placental discrimination factor of 0.5 (34) is considered. *Bryant et al.* (35) in England reported analyses of 28 bone samples from subjects of all ages collected about January of 1956. Eight samples from persons ranging from 3 months to  $3\frac{1}{2}$  years old (average  $1\frac{1}{3}$  years) averaged  $0.9 \mu\mu\text{c Sr}^{90}/\text{g Ca}$ , and 11 subjects ranging from 20 to 65 years of age (average 36 years) averaged  $0.07 \mu\mu\text{c Sr}^{90}/\text{g Ca}$  (after dividing all rib results by 2 (6)). The predicted average maximum  $\text{Sr}^{90}$  equilibrium levels about January 1956, based on these age groups, are 1.0 and  $0.9 \mu\mu\text{c/g Ca}$ , respectively.

On the assumption that surface deposition levels had a doubling time of 1 year, an average maximum bone equilibrium level of  $1.8 \mu\mu\text{c/g Ca}$  was predicted for the north temperate latitudes for the fall of 1956 (32). Data on  $\text{Sr}^{90}$  fallout from pot collection samples in New York and Pittsburgh (5) show, however, that fallout did not double but increased by only about 50 per cent. On this basis, the predicted level for the north temperate population belt in the fall of 1956 would be  $1.4 \mu\mu\text{c/g Ca}$ . *Kulp* (28) applied the same age weighting method to 1956–1957 milk data (assuming a bone-to-diet discrimination ratio of 0.25) and estimated average maximum equilibrium bone levels (by the end of 1956) of 1.1, 0.9, 1.1, and 0.5 for North America, Europe, Asia, and the rest of the world, respectively. A crude estimate of present and future average maximum equilibrium bone levels can be made from the 1956 data by assuming they will be proportional to the predicted average maximum surface deposition levels given previously. These estimates are compared in table 6 with those postulated from ecological considerations. Values postulated from ecological discrimination and from bone analyses differ by a factor of about 2. This discrepancy results mostly from the weighted influence of the Far Eastern countries, with low milk consumption and large populations, on the discrimination ratios. Although bone data are probably more reliable than ecological predictions, they may be low since they were predicted on the assumption that the analyses represented the average bone levels for the various regions. Since the number of samples from the United States and Europe ex-

ceeded the number from countries with large populations and low relative milk consumption, it is unlikely that they are weighted adequately for population density and dietary habits.

Another troublesome feature of such estimates is that they are average maximum equilibrium levels and make no allowance for local variations of fallout due to meteorological factors, variations in available soil calcium, dietary patterns and habits, nutritional state of segments of the population, individual metabolic condition, etc.

Frequency distribution patterns have been reported for stable strontium (36), natural radium (37), and  $\text{Cs}^{137}$  (7) in man. All these nuclides show essentially normal distributions with standard deviations of about 35 per cent. *Libby* (13) has stated that (at steady state among people living in a given locality) only one person in about 700 will have more than twice the average  $\text{Sr}^{90}$  burden, and the chances of anyone having as much as three times the average will be about one in 20 million. At present, the  $\text{Sr}^{90}$  measurements of bone samples from subjects of all ages show a much greater scatter than indicated by a standard deviation of 35 per cent. The greater scatter of the observed values is due largely to the fact that samples came from many localities and (because of the relatively short period of environmental contamination and the age dependence of  $\text{Sr}^{90}$  deposition) represent varying degrees of equilibrium conditions. The spread may be expected to decrease as equilibrium is approached (13, 28).

Local meteorological conditions will result in increased intensity of fallout in certain localities. The worst possible situation that could come about would be for these "hot spots" to coincide with localities of low available soil calcium in which the population grew up and lived in provincial isolation. *Libby* (13) has considered this problem in view of the general averaging which occurs in food distribution systems and has postulated that a factor of 5 encompasses the total variation due to all factors.

The question as to the applicability of the normal distribution curve to  $\text{Sr}^{90}$  equilibrium levels in bone has been raised (38, 39). The observed distribution of stable strontium in bone (36) appears to be log normal rather than normal; in fact, the former is rather common for geochemical distribution (40). The great fundamental difference in the mechanisms of distribution of stable strontium and  $\text{Sr}^{90}$ , however, greatly weakens arguments based on the analogy. Whether the distribution of equilibrium levels in the bones of the population will be normal or log normal can probably be decided only by more extensive experimental evidence.



## Cesium 137

*Ecological Incorporation and Discrimination.*—Cesium is chemically and metabolically similar to potassium, an essential body constituent. If it enters the food chain from the soil (rather than by direct fallout on plants), its uptake via the ecological cycle and incorporation into man should be in relation to the exchangeable or available soil potassium. It is reasonable, therefore, to consider incorporation of  $\text{Cs}^{137}$  into the biosphere in terms of  $\text{Cs}^{137}/\text{K}$  ratios. Like  $\text{Sr}^{90}$ ,  $\text{Cs}^{137}$  may be incorporated through direct fallout on vegetation and through soil accumulation and uptake by plants. When  $\text{Cs}^{137}$  comes in contact with soil, it is rapidly fixed. Leaching studies (41) show essentially all of the  $\text{Cs}^{137}$  remains in the top inch of soil, even after 200 inches of simulated rainfall. The extent of fixation, as with potassium, is probably proportional to the colloidal content of the soil, being greatest in clays and clay loams and least in light sands and sandy loams.

Plants discriminate heavily against  $\text{Cs}^{137}$  with respect to potassium, even when the cesium is in an exchangeable form. *Auerbach* (42) reported uptake of  $\text{Cs}^{137}$  by corn grown in a lake bed once used for the disposal of reactor wastes. He found that the  $\text{Cs}^{137}/\text{K}$  ratio in the plants was about 1 per cent of the exchangeable  $\text{Cs}^{137}/\text{K}$  ratio in the soil. *Menzel* (43) obtained a discrimination factor of about 0.04 between  $\text{Cs}^{137}/\text{K}$  in barley and corn and the ratio in available soil potassium, and definitely showed that plant uptake of  $\text{Cs}^{137}$  was inversely proportional to exchangeable and available soil potassium. Without considering exchangeable soil potassium, others (44, 45) have studied the ratio of  $\text{Cs}^{137}$  per gram of dry plant materials to the concentration per gram of soil and obtained values of 0.006 to 0.18 (average 0.07). These data suggest that the average  $\text{Cs}^{137}$  concentration in the potassium of plants should be about 0.04 times the exchangeable  $\text{Cs}^{137}$  concentration in exchangeable soil potassium.

Exchangeable soil potassium, to a depth of 2.5 inches, may vary from about 25 to 400 lbs/acre. About 100 lbs/acre is a reasonable average for the agricultural soils of the United States. This is equivalent to about  $3 \times 10^7$  g of exchangeable K/mi<sup>2</sup>. Deposition and mixing to a depth of 2.5 inches of 1 mc of  $\text{Cs}^{137}$ /mi<sup>2</sup> gives a total concentration of about 30  $\mu\mu\text{c/g}$  of exchangeable soil potassium. *Larson et al.* (46) added  $\text{Cs}^{137}$  to three different types of soils and determined the amount that could be extracted with N  $\text{NH}_4\text{Ac}$ . The exchangeable  $\text{Cs}^{137}$  ranged from 13 to 33 per cent with an average of 25. Assuming that 75 per cent of the  $\text{Cs}^{137}$  is fixed in a form unavailable to plants, the discrimination factor ( $DF_1$ ) in going from soils-to-plants would be equal to 0.01 and the con-



centration of  $\text{Cs}^{137}$  in plant potassium from fallout of 1 mc/mi<sup>2</sup> would be about 0.3  $\mu\mu\text{c/g}$ .

The  $\text{Cs}^{137}$  deposition levels in the northern United States (mid-1957) is estimated at about 46 mc/mi<sup>2</sup>, which suggests 15  $\mu\mu\text{c Cs}^{137}/\text{g}$  of plant potassium, or a  $\text{Cs}^{137}/\text{K}^{40}$  gamma ratio of 0.18. The calculated ratio is in reasonable agreement with values measured in the Los Alamos large-volume liquid scintillation counter (7). Measured  $\text{Cs}^{137}/\text{K}^{40}$  ratios in 1957 dried milk samples from the northern United States (47) averaged about 30  $\mu\mu\text{c/g}$ , giving an estimated discrimination factor ( $DF_2$ ) of about 2 in favor of  $\text{Cs}^{137}$  in going from plants-to-milk.

Tracer studies on man (48) show that  $\text{Cs}^{137}$  and  $\text{K}^{42}$ , upon ingestion, are absorbed essentially 100 per cent and that they are excreted with mean times of about 150 and 50 days, respectively. These data suggest a discrimination factor of about 3 in favor of  $\text{Cs}^{137}$  in going from diet ( $DF_3$  and  $DF_4$ ) to man<sup>1</sup>. Since 50 per cent of the potassium in a western diet comes from milk and dairy products (7), the over-all ratio ( $OR$ ) of  $\text{Cs}^{137}/\text{K}$  in going from soils-to-man equals  $0.5(0.01 \times 2 \times 3) + 0.5(0.01 \times 3)$ , or 0.045. In other words, the  $\text{Cs}^{137}$  concentration per gram of body potassium should be about 4.5 per cent of the total  $\text{Cs}^{137}$  concentration per gram of exchangeable soil potassium.

Anderson et al. (7) suggested that  $\text{Cs}^{137}$  may be entering the biosphere and man largely through direct fallout on vegetation and not by plant uptake from the soil. This suggestion was based on the following considerations: 1. The high fixation of  $\text{Cs}^{137}$  in soil and its very slow leaching rate make it unlikely that the  $\text{Cs}^{137}$  can be in equilibrium with exchangeable soil potassium to the depth of the plant feeding zone. 2. The  $\text{Cs}^{137}$  to  $\text{K}^{40}$  ratio of people does not seem to be increasing in relation to integrated  $\text{Cs}^{137}$  fallout. 3.  $\text{Cs}^{137}/\text{K}^{40}$  ratios in milk show sharp increases during periods of weapons testing, after which they rapidly return to near their previous levels, suggesting the possibility of a quasi-equilibrium condition with the rate of stratospheric fallout. The relatively small effect of a sharp increase in the  $\text{Cs}^{137}$  content of foods during periods of tropospheric fallout on the  $\text{Cs}^{137}$  content of people can be explained by the simple model shown in fig. 6 (7). A step function change in the food-stuff 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 about 1 year. If the food-stuffs return to their previous value before equilibrium is attained, the

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<sup>1</sup> A value of 3 for the discrimination factor from the milk-to-man ( $DF_3$ ) is not confirmed by measurements on people and milk from the same areas (47). These data strongly suggest a discrimination factor of approximately 1.

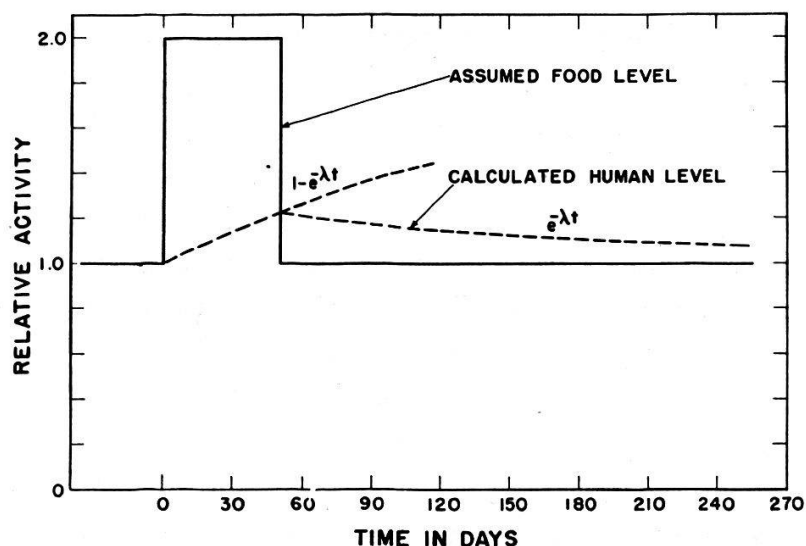


Fig. 6. Calculated effect of temporary increase in  $\text{Cs}^{137}$  level of the diet on the  $\text{Cs}^{137}$  in people.

population level will cease rising and will return to its previous value with a half-time corresponding to the biological elimination rate.

**$\text{Cs}^{137}$  Levels in the Population.**—Concentrations of  $\text{Cs}^{137}$ /g of body potassium can be estimated from predicted average maximum surface deposition levels given in tables 2, 3, and 4 and the ecological considerations discussed previously. One millicurie  $\text{Cs}^{137}/\text{mi}^2$  gives a specific activity of  $30 \mu\mu\text{c/g}$  of exchangeable soil potassium. The specific activity times the surface deposition levels times the over-all discrimination ratio (0.045) gives the specific activity per gram of body potassium for the population of any fallout area, assuming no equilization between areas through food distribution channels. Present and future levels of  $\text{Cs}^{137}$  in the population of various regions, estimated from ecological considerations, are shown in table 7.

Measurements of  $\text{Cs}^{137}/\text{K}^{40}$  gamma ratios of the United States population during 1956 averaged about 0.5 (7), which corresponds to  $41 \mu\mu\text{c}$   $\text{Cs}^{137}/\text{g}$  of body potassium or about  $0.0055 \mu\text{c}$  of  $\text{Cs}^{137}$  in the total body assuming 133 g of potassium in a 70-kg man. Measurements during 1957 (47) gave average  $\text{Cs}^{137}$  concentrations of 45 and  $50 \mu\mu\text{c/g}$  of body potassium for the general United States population and the population of the northern states, respectively. Levels in the United States population might be expected to show little variation because of the equalizing effect of general food distribution systems.

The value of  $62 \mu\mu\text{c/g K}$  for the population of the northern United States, estimated from ecological considerations, agrees very well with the average value of  $50 \mu\mu\text{c/g K}$  derived from measured  $\text{Cs}^{137}/\text{K}^{40}$  ratios. The agreement may be purely coincidental and could result from direct

Table 7  
Estimated Present and Future Concentrations of  $\text{Cs}^{137}$  in the Population of the Basis  
of Ecological Considerations

Region	$\text{Cs}^{137}$ concentration		
	Mid-1957 ( $\mu\mu\text{c/g K}$ )	1965 No more tests ( $\mu\mu\text{c/g K}$ )	Continued tests* ( $\mu\mu\text{c/g K}$ )
Northern United States	62	69	620
North temperate latitudes	34	37	340
South temperate latitudes	10	11	100
Rest of world	7	8	70
World average**	(32)	(36)	(320)

\* Assuming equilibrium with continued testing at the past 5-year rate (equivalent to 10 MT of fission per year).

\*\* Population weighted average on the basis of present world population distribution.

fallout on vegetation, fortuitously making up for non-equilibrium of  $\text{Cs}^{137}$  with exchangeable soil potassium.

The estimates of future levels given in table 7 are predicated on the assumption that  $\text{Cs}^{137}$  is entering the biosphere largely through the soil and that the contribution of direct fallout on vegetation is negligible. In this case,  $\text{Cs}^{137}$  levels in people might be expected to rise somewhat in accordance with the estimated values. If present levels represent a quasi-equilibrium with direct fallout, population levels (with cessation of testing) might be expected to start dropping immediately with a half-time comparable to the half-time of stratospheric fallout. In this case, continued testing at the past 5-year rate will produce little or no increase in the average  $\text{Cs}^{137}$  of the population. Present levels in the biosphere actually may be a result of significant contribution from both direct fallout and ecological integration, in which case the truth will be somewhere in between. It should be possible to decide among these alternatives within the next few years.

### *Plutonium 239*

*Ecological Incorporation and Discrimination.*—Although the presence of naturally occurring  $\text{Pu}^{239}$  in pitchblende concentrate has been reported (49), its existence in the biosphere can be attributed entirely to the detonation of nuclear weapons. Unlike  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$ , it is chemically unrelated to any essential constituent of plants or animals.

When plutonium is deposited in soil it is extremely tightly bound, and the establishment of uniform distribution to the depth of the plant feeding zone may require years. A plutonium deposition level of 1 mc/mi<sup>2</sup>

would be equivalent to about  $5 \times 10^{-3} \mu\mu\text{c/g}$  of soil when uniformly mixed to a depth of 2.5 inches. Absorption of plutonium by barley from a sandy soil was studied by *Rediske* (44), who found that the ratio of plutonium concentration in dry plant material to the concentration in the soil was  $9 \times 10^{-4}$ . When ingested by man and domestic animals, absorption of plutonium is only about 0.01 per cent. Once it is absorbed, about 85 per cent is fixed in the skeleton and largely retained throughout the life-time of the animal. The apparent half-time of plutonium elimination by man is about 200 years, which means it is essentially cumulative on absorption. Its high fixation in the skeleton of domestic animals, however, provides an additional discrimination factor of about  $10^{-2}$  in meat and dairy products. The over-all discrimination ratio in going through the ecological cycle from soils to man is at most  $5 \times 10^{-8}$ . The estimated present average maximum plutonium deposition level for the north temperate population belt would lead to a plutonium uptake of the order of  $10^{-7}$  of the recommended maximum permissible level from the consumption of a 3 000-calorie diet for 70 years. With such a large discrimination, it is quite unlikely that incorporation of plutonium fallout into man via the ecological chain can be of any consequence. Incorporation via inhalation and direct fallout on vegetation, although insignificant also, probably would be much greater than incorporation via ecological transport.

### *Iodine 131*

Radioactive iodine from weapons tests has been reported in human thyroids (50, 51) and in the thyroids of domestic animals (50–54). Because of its 8-day half-time,  $\text{I}^{131}$  cannot integrate in the biosphere and its concentration in thyroids fluctuates in relation to tropospheric fallout during periods of nuclear testing. *Van Middlesworth* (50) reported the analysis of 175 human and 1044 cattle thyroids collected from the Memphis, Tennessee, area during November 1954 to March 1956, and *Comar et al.* (51) reported analysis of 1 165 human and 853 cattle thyroids collected from several countries during the period from January 1955 to December 1956. These data show that the concentration of  $\text{I}^{131}$  in cattle thyroids is about 18 to 200 times that of man. The average concentration in cattle thyroids during the period from November 1954 to December 1956 appears to be about 0.5 (54) and the average peak level in man about 0.005  $\text{m}\mu\text{c/g}$  (51).

The principal mode of entry of  $\text{I}^{131}$  into domestic animals seems to be through ingestion of direct fallout on forage. Grazing animals show a much higher thyroid uptake than do lot fed. The mode of entry of  $\text{I}^{131}$

into man is believed to be via direct inhalation with ingestion of contaminated milk as a secondary route. Following oral feeding to cattle, about 6 per cent of the ingested  $I^{131}$  appears in the first week's milk production (55). The average milk concentration during the 1955 period of high level fallout was estimated at about 0.2 m $\mu$ c/liter, which is a factor of 500 below the value chosen as unsafe for public consumption during the recent United Kingdom Windscale reactor accident (56).

Since  $I^{131}$  does not accumulate in the biosphere, the above values may be considered crude average maximum equilibrium levels with the present rate of testing. Although large local fluctuations may be expected from time to time as a result of tropospheric meteorological variations and proximity to test sites, the average  $I^{131}$  content of the thyroids of man and livestock should not increase materially with continued testing at the past 5-year rate.

One m $\mu$ c of  $I^{131}$  per gram of thyroid delivers a radiation dose of about 10 mrad/day (53). The average  $I^{131}$  concentrations during the 1955 peak period of fallout delivered about 35 and 0.3 mrad/week to livestock and man, respectively. The integrated dose received during 1955 was actually much lower (51).

If the 1955 peak levels are maintained in people and livestock, the yearly integrated dose to the thyroid will be about 15 and 1,500 mrad per year, respectively. For man, this is about 1 per cent of the recommended maximum permissible level for continuous exposure of large segments of the population.

The external radiation dose to the neck area in infants and children that possibly has caused later thyroid malignancy is estimated at 200 to 750 r (57), and about 900 rad to the thyroids of sheep chronically fed  $I^{131}$  over a 6-year period failed to produce any observable damage (58).

In the event of nuclear war, it is conceivable that  $I^{131}$  could constitute a significant acute danger in localized areas. However, there seems to be very little probability that  $I^{131}$  levels introduced into the biosphere by continuation of weapons tests at the past rate will pose any general hazard to man and domestic animals.

### *Significance of $Sr^{90}$ and $Cs^{137}$ Levels in the Population*

#### *Strontium 90*

The potential significance of present and predicted  $Sr^{90}$  levels in bone can be evaluated only in relation to human experience, which is indeed inadequate. Bone sarcoma has resulted from a fixed skeletal burden of 3.6  $\mu$ c of pure  $Ra^{226}$ , and non-deleterious bone changes have been observed in persons having only 0.4  $\mu$ c for a period of 25 years (59). Necrosis



Table 8  
Average Natural Background Radiation to the Skeleton (65)

Source of radiation	Skeletal dose rate	Total dose to age 70
	(mrem/year)	(rem)
K <sup>40</sup> (internal)	8	0.56
Ra <sup>226</sup> (internal)	12	0.84
MsTh (internal)	12	0.84
RaD (internal)	12	0.84
Cosmic rays (external)	30	2.10
Local gamma rays (external)	60	4.20
<b>Total</b>	<b>134</b>	<b>9.40</b>

and tumors of the bone have occurred also several years after large doses of X-rays (60), and consideration of human experience with leukemogenic effects of X and gamma radiation (61, 62, 63) suggests that about 80 rads may double the incidence of leukemia.

The only other human experience with which present and predicted levels of Sr<sup>90</sup> may be compared is that arising from natural background radiation. Natural background dose to the bone (during a 70-year lifetime) may vary from about 8 to 38 rem (64). The major contribution to background variation is differences in the radium levels of soils and minerals. The average natural skeletal radiation dose rate was carefully evaluated by *Dudley and Evans* (65) and their data are shown in table 8.

Fig. 7 shows a general summary of estimated skeletal radiation doses

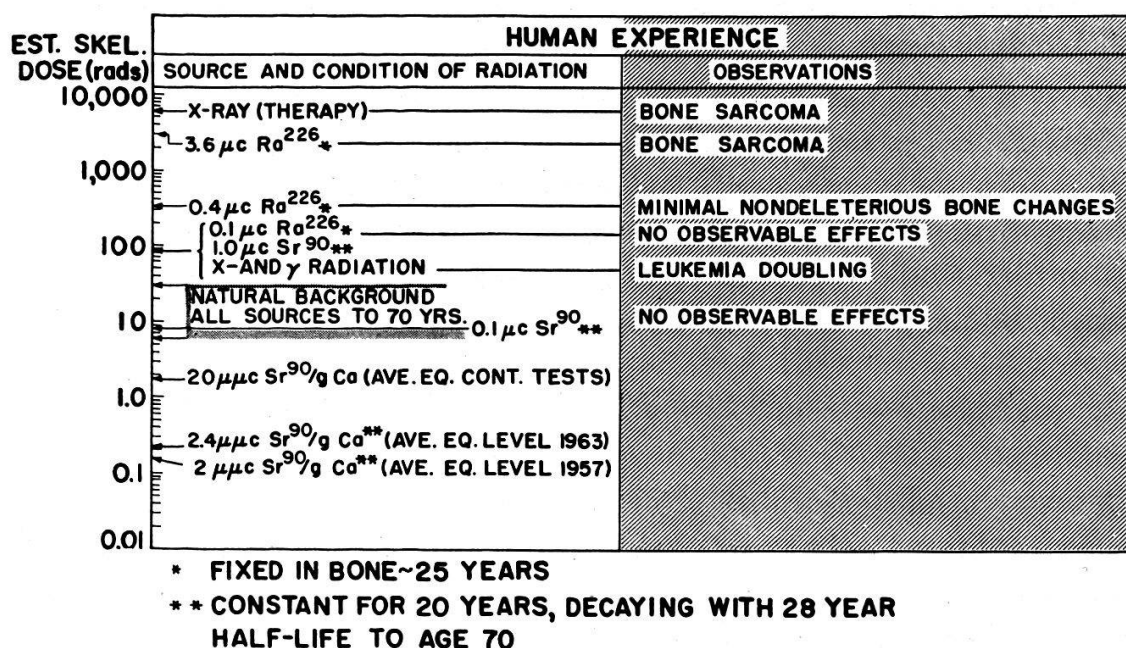


Fig. 7. Estimated Sr<sup>90</sup> skeletal radiation dose in relation to human experience.



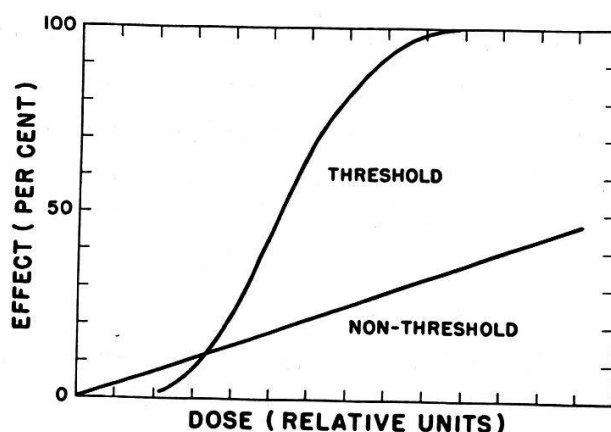


Fig. 8. Threshold and nonthreshold response as a function of radiation dose.

from accepted maximum permissible levels and from present and predicted  $\text{Sr}^{90}$  burdens in relation to human experience. The maximum permissible level of  $\text{Sr}^{90}$  ( $100 \mu\mu\text{c/g Ca}$ ) is estimated to deliver about 8.5 rads<sup>1</sup> to the skeleton during a 70-year life-time. This is comparable to the average natural background dose to the bone for the same time period and a factor of about 4 below the maximum natural background dose to which small segments of the general population may be exposed as a result of differences in altitude and natural radium content of soils and minerals. It is a factor of 40 below the lowest skeletal dose which has produced minimal nondeleterious bone changes and a factor of about 10 below the leukemia doubling dose. These data suggest that the present average maximum  $\text{Sr}^{90}$  equilibrium level will result in a life-time radiation dose of 1 to 2 per cent of the accepted maximum permissible level for the general population. With continued biospheric contamination indefinitely at the past 5-year rate, the average maximum radiation dose may approach about 20 per cent of the presently accepted maximum permissible level.

*Threshold versus Nonthreshold Response.*—If chronic effects of radiation are threshold phenomena,  $100 \mu\mu\text{c Sr}^{90}/\text{g Ca}$  must be looked upon as a true maximum permissible level and not as an average for large segments of the population. If chronic effects are nonthreshold phenomena and linear with dose (fig. 8), the maximum permissible level of  $\text{Sr}^{90}$  in the bones may be expressed in terms of a population or group average, and a portion of the natural population incidence of the effect in question must be attributed to natural background radiation. In this

<sup>1</sup> Eight and five-tenths rads is the calculated dose assuming incorporation to age 20 and decay to age 70 with no more incorporation. If equilibrium were maintained, the calculated skeletal dose would be about 21 rads. Since some but not all of the skeleton undergoes remodeling plus exchange, somewhere between 8 and 21 rads is probably more correct.

case, the potential hazard should be established on the basis of probability of risk averaged over the entire population or group.

At present it is impossible to say whether leukemogenic and sarcomagenic responses to chronic radiation dosage are threshold or nonthreshold relationships. The recent Congressional Hearings (1) failed to produce any degree of unanimity of opinion among the experts. Argument for a linear relationship between incidence of leukemia and radiation dose was presented recently by *Lewis* (62). His argument was based on all major sources of human data and included a consideration of the Japanese atomic bomb survivors, the British cases of X-ray treated spondylitis, X-ray treated cases of thymic enlargement, practicing radiologists, and spontaneous incidence of leukemia in Brooklyn, New York. The validity of his conclusions was questioned by *Warren*, *Brues*, and others during the Congressional Hearings (1). Radiation as a carcinogenic agent has been discussed at length by *Brues* (66), who stated that the relation between radiation dose and carcinogenic effect is not easy to find and a critical experiment has yet to be done which will clearly indicate, even in a single instance, what the relation is over more than a small range of dosages. While admitting that it is not known, he proposes that a threshold relationship between radiation dose and tumor incidence does exist (67).

Without adequate scientific basis but for the purpose of presenting the worst possible potential hazard from  $\text{Sr}^{90}$  biospheric contamination, a comparison may be made between the radiation dose from present  $\text{Sr}^{90}$  bone levels and the postulated leukemia doubling dose (62). Assuming a nonthreshold response and that 10 per cent of the natural incidence of leukemia in the population (6/100,000) is a result of natural background radiation, the average maximum  $\text{Sr}^{90}$  equilibrium bone level for the north temperate population belt would be equivalent to about 1.2 leukemia cases per 10 million population. Averaged over the world population of 2.6 billion, this would produce an increased leukemia burden of 300 cases per year. A world average of  $100 \mu\mu\text{c Sr}^{90}/\text{g Ca}$  would be equivalent to about 16,000 cases.

The above analogy assumes that  $\text{Sr}^{90}$  beta radiation induces leukemia of bone marrow origin at the same rate (per unit of absorbed dose) as X and gamma rays. Much of the beta radiation from  $\text{Sr}^{90}$  will be absorbed in the bone and not reach the hematopoietic tissues at all. Experiments by *Bruce et al.* (68) suggest that  $\text{Sr}^{89}$  (half-life 55 days,  $E_{\beta} = 1.5 \text{ Mev}$ ) administered to mice is relatively more spectacular as an osteosarcogenic agent than a leukemogenic agent. Furthermore, leukemia was not a significant finding in the radium dial painters (69, 70) or in the radium-injection cases (59).

Bone sarcoma is more apt to result from  $\text{Sr}^{90}$  than is leukemia. Human data on radiation-induced osteogenic sarcoma are not adequate to provide even the crudest estimate of the dose response relationship, the population doubling dose, or the fraction of normal population incidence (about 2/100,000) attributable to natural background.

Under the same conditions, the potential risk to the population from bone sarcoma, however, would be less than that calculated for leukemia, since its natural incidence in the population is lower than that of leukemia.

### *Cesium 137*

The present average  $\text{Cs}^{137}$  level in the population of the United States is about 45  $\mu\mu\text{c/g}$  body potassium. This is equivalent to 0.006  $\mu\text{c}$  per person.  $\text{Cs}^{137}$ , like potassium, is concentrated in muscle and the radiation dose it delivers is essentially whole body. The dose delivered is equivalent to approximately 1 mr/year. Taking into consideration the respective energies of their radiations, the dose from the present level of  $\text{Cs}^{137}$  is about one-twentieth of that from natural  $\text{K}^{40}$ , or about 1 per cent of the average natural background. If  $\text{Cs}^{137}$  is entering man largely through the ecological cycle, continued testing at the past 5-year rate may result in an average radiation dose to the United States population of about 10 mr/year, or about 10 per cent of natural background, and a weighted world population average of about 7 per cent of background. Because of nonhomogeneities in fallout and uptake, a few persons may receive doses about 5 times the average. If  $\text{Cs}^{137}$  is entering man largely through direct fallout on vegetation and not through ecological integration, continued testing may not increase the average  $\text{Cs}^{137}$  dose significantly above the present levels.

Concern has been expressed (71) over the possible genetic implications of selective concentrations of  $\text{Cs}^{137}$  in the gonads. The data in table 9

Table 9  
Concentration of  $\text{Cs}^{137}$  in the Gonads of Rats

Days after administration	Concentration gonads/muscle*
Testes	
2	0.70
5	0.71
10	0.52
Ovaries	
2	0.82

\* Average of 3 animals per point.

show the ratio of  $\text{Cs}^{137}$  concentration in the gonads of rats to that in muscle. The testes and ovaries concentrate  $\text{Cs}^{137}$  to the extent of about 70 and 80 per cent of muscle, respectively, and the elimination time from the testes appears shorter. Therefore, the radiation dose delivered to the gonads is comparable to that delivered to muscles, or about 2 mr/year at present United States average  $\text{Cs}^{137}$  levels.

### Discussion

Past testing of nuclear weapons has produced between 5 and 6 MC of  $\text{Sr}^{90}$  (equivalent to 50 to 60 MT of fission energy). About 90 per cent of the production has occurred since 1952 from testing of weapons in the megaton class. United States Pacific tests have been held under conditions that maximized local fallout (where  $\text{Sr}^{90}$  is of no concern because of the vast calcium reservoir of the ocean) and minimized world wide contamination. Soil data suggest that about 1.6 MC of  $\text{Sr}^{90}$  have been distributed as long-range fallout. The present stratospheric reservoir is estimated at about 2.4 MC by *Libby* (12), and at  $1.1 \pm 0.93$  in this report. Present integrated surface deposition levels are such that the rate of  $\text{Sr}^{90}$  decay on the ground is almost equal to the rate of stratospheric fallout. If weapons tests were to stop, integrated surface deposition levels in the north temperate latitudes would probably increase by no more than 10 to 20 per cent, reaching a maximum in about 1963 to 1970.

Unfortunately, because of the locations of the United States and USSR test sites and tropospheric and stratospheric meteorological phenomena, long-range fallout is maximized in the north temperate latitudes where over 80 per cent of the world's population lives. The present average soil level in the northern United States is about 35 mc/mi<sup>2</sup>, and the average level elsewhere in the same general latitudes may be about 20 mc/mi<sup>2</sup>. Deposition levels elsewhere in the world are not potentially important with regard to general world health because of population distribution.

Estimates of average maximum  $\text{Sr}^{90}$  equilibrium bone levels for the northern United States and the north temperate population belt (from weapons tests to date) vary from about 1 to 4  $\mu\text{mc/g}$  Ca. Controversy over the issue of stopping or continuing bomb tests has resulted in greater apparent public confusion over the potential hazard of world-wide  $\text{Sr}^{90}$  fallout than seems justified by the factor of 4 difference in estimates of average maximum equilibrium bone levels. This confusion has resulted largely from differences in choice of reference as to average maximum permissible  $\text{Sr}^{90}$  levels applicable to the general population and differ-

ences in opinion as to an appropriate factor of allowance for nonhomogeneity of fallout and bone uptake.

*Libby* (5, 12) and *Kulp* (72), before any authoritative statements regarding a  $\text{Sr}^{90}$  MPL for the general population had been issued, used the occupational MPL ( $1,000 \mu\mu\text{c/g Ca}$ ) as a reference. Later the National and International Commissions for Radiological Protection recommended that the MPL for large segments of the general population should be one-tenth ( $100 \mu\mu\text{c/g Ca}$ ) that for occupational exposure. The U.S. National Academy of Sciences-National Research Council report (73) inferred that  $50 \mu\mu\text{c/g Ca}$  might be considered as a safe level for the general population. The British Medical Council report (61), while acknowledging that the maximum allowable concentration of  $\text{Sr}^{90}$  in the bones of the general population should not be greater than  $100 \mu\mu\text{c/g Ca}$ , stated that immediate consideration should be required if the concentration in human bones showed signs of rising greatly beyond one-hundredth ( $10 \mu\mu\text{c/g Ca}$ ) of that corresponding to the maximum permissible occupational level. *Lapp* (74) has stated also that the MPL for the general population perhaps should be one-hundredth of the occupational value. All of these numbers have been brought to public attention during the controversy over continued weapons tests (1).

Confusion has been increased also by the use of various safety factors for nonhomogeneity of fallout and bone uptake. Articles have appeared in which no factor was used (5, 6, 12), and others have appeared in which factors of 5 (13) and 10 (74, 75) were recommended.

The effect of choice of values for the acceptable general population MPL and the choice of safety factors for nonhomogeneity of distribution and uptake are shown by the data in table 10. These data were derived by simple proportionality (Maximum Bone Level from Present Tests: 50 MT :: Acceptable MPL: X) and show the MT of fission energy release (over a short period) required to bring the average maximum equilibrium bone levels of the population to the various permissible values that have been called to public attention. The table also indicates the effect of various nonhomogeneity factors on the world average population level. These data show a variation of a factor of about 1 000 in the megaton equivalents of fission that could be detonated, depending on whether one wishes to be ultraconservative and use the highest safety factor for nonuniformity and the lowest value recommended for the general population, or be the opposite and use the occupational MPL and no safety factor for nonuniformity. The most important point to the data is that they explain the principal reasons for public confusion and show that the major areas of uncertainty are, 1. the maximum permissible level



Table 10  
Allowable MT of Fission Energy Release as a Function of Various  
General Population MPL's

Source of equilibrium bone level estimate and region	Ave. Max. Bone Level (no more testing) ( $\mu\mu\text{c/g Ca}$ )	Allowable MT of fission yield			
		10 $\mu\mu\text{c}^*$	50 $\mu\mu\text{c}^*$	100 $\mu\mu\text{c}^*$	1,000 $\mu\mu\text{c}^*$
United States					
Libby (13) – Ecological data	3.9–1.7	130–300	650–1500	1300–3000	13000–30000
Kulp (6) – Ecological data	2	250	1250	2500	25000
Kulp (28) – Bone data	1.5	300	1500	3000	30000
Eisenbud (76) – Milk data	4	120	600	1200	12000
This paper – Ecological data	3.5	140	700	1400	14000
This paper – Bone data	1.9	250	1250	2500	25000
North temperate latitudes					
This report – Ecological data	3.6	140	700	1400	14000
This report – Bone data	1.9	250	1250	2500	25000
World average					
Kulp (6) – Ecological data	1.3	380	1900	3800	38000
This report – Ecological data	3.1	160	800	1600	16000
This report – Bone data	1.7	150	1500	3000	30000
World average (no factor for distribution)	2	250	1250	2500	25000
Average $\times 1/5$ (for non-uniformity)		50	250	500	5000
Average $\times 1/10$ (for non-uniformity)		25	125	250	2500

\* Per gram Ca.

for  $\text{Sr}^{90}$  as applied to the general population, and 2. the deviation of equilibrium bone values from the mean.

The most important question regarding the potential hazard of long-range  $\text{Sr}^{90}$  fallout is in relation to future weapons testing. If there is an upper limit to the amount of  $\text{Sr}^{90}$  in the bones of the population that can be safely tolerated, then the megaton equivalents of fission products that can be contributed per year to the biosphere by all nations is limited.

If  $\text{Sr}^{90}$  contamination from weapons testing by all nations continues at the same rate as has occurred during the past 5 years, equilibrium will be reached in about 100 years. At equilibrium the amount of  $\text{Sr}^{90}$  which will disappear each year from the environment, due to radioactive decay, will equal the amount that is being produced, and continuing weapons tests will not result in any further increase in the population bone levels.

*Libby* (13, 17) and others (18) have predicted that soil and bone levels at equilibrium with the present test rate will be 8 to 13 times the present values. On the basis of present average maximum equilibrium  $\text{Sr}^{90}$  bone levels postulated from the considerations set forth in this paper, the bones of the United States population will reach a steady state with the present testing rate at a value of 17 to 31  $\mu\mu\text{c/g Ca}$ . The equilibrium value for the weighted average world population will be 15 to 28  $\mu\mu\text{c}$  per g Ca.

*Libby* (13) has stated that something between 5 and 20  $\mu\mu\text{c/g Ca}$  would be the average maximum  $\text{Sr}^{90}$  concentration in the bones of the United States population if testing continued indefinitely at the average rate of the past 5 years. *Kulp* (28) predicted an equilibrium level will be approached in the North American population of about 8  $\mu\mu\text{c/g Ca}$  in about 50 years, and *Neuman* (18) in testimony before the Congressional Subcommittee suggested equilibrium bone levels of about 90  $\mu\mu\text{c/g Ca}$  may be reached in the northern United States. The values given above show disagreement by a factor of about 10. If, however, we accept as a reasonable average the values developed in this paper, the average  $\text{Sr}^{90}$  radiation dose to the bones of the population of the northern United States, at equilibrium with continued testing at the past rate, may be about 20 to 30 per cent of the average radiation dose from natural background, or about 20 to 30 per cent of the maximum permissible level adopted by the National and International Commissions. Since individual variations may result in a small number of people accumulating  $\text{Sr}^{90}$  burdens that are 5 times the average, the radiation dose to these few individuals may approach as an upper limit 100 to 150 per cent of the recommended maximum level. If testing is continued at the present rate for 30 years, the average level of  $\text{Sr}^{90}$  in the population of the northern United States may be about 10 to 15 per cent of natural background. This may result in a few people approaching body burdens about 50 to 75 per cent of the recommended maximum.  $\text{Sr}^{90}$  burdens in the weighted world average population will be essentially the same.

The average  $\text{Cs}^{137}$  levels presently in the population of the United States is about 45  $\mu\mu\text{c/g K}$ . This amount of  $\text{Cs}^{137}$  is delivering a radiation dose of about 1 mr/year, or about 1 per cent of the natural background dose. The present population weighted world average may be about 32  $\mu\mu\text{c/g}$ . Continued testing at the past 5-year rate until equilibrium may result in an average world population  $\text{Cs}^{137}$  radiation dose of about 7 per cent of background, depending on whether  $\text{Cs}^{137}$  is entering the biosphere largely via ecological transmission from the soil or by direct fallout on vegetation. In either case,  $\text{Cs}^{137}$  appears to be relatively less

important than  $\text{Sr}^{90}$  as a potential internal hazard from world-wide fall-out. Other long-lived radionuclides, including  $\text{Pu}^{239}$ , appear to be orders of magnitude less significant than  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$ .

These considerations suggest that the past rate of weapons testing, if continued for several years, will not produce internal radiation levels that will exceed the general population maximum permissible levels recommended by the National (77) and International (21) Commissions on Radiological Protection. Although this leads to the conclusion that the present rate of biospheric contamination poses no serious potential somatic hazard to world health, the great uncertainties involved make it imperative that the problem be kept under constant scrutiny if weapons tests are to continue. Fortunately, present levels are not critical and the slow rate of biospheric build-up affords time for continued intensive and extensive study.

### *Summary*

Testing of atomic weapons through mid-1957 has produced about 5.5 MC of  $\text{Sr}^{90}$  and varying amounts of other long- and intermediate-lived isotopes. Of these,  $\text{Sr}^{90}$  poses the greatest potential hazard to world health.  $\text{Cs}^{137}$  and  $\text{Pu}^{239}$  are the other principal long-lived activities in weapons debris.  $\text{Sr}^{90}$  and  $\text{Cs}^{137}$  have been measured quantitatively in people, milk and other foodstuffs, and the intermediate-lived  $\text{I}^{131}$  has been detected in the thyroids of man and domestic animals.

Two theories have been proposed for distribution of weapons debris throughout the world. The Libby model postulates uniform, rapid mixing of weapons debris throughout the stratosphere with equal leakage back through the tropopause, after which it is deposited over the earth's surface in relation to tropospheric meteorological conditions. The Machta model proposes slow, unequal stratospheric mixing with preferential leakage through the tropopause in the vicinity of the jet streams. Qualitatively, the two models give the same distribution pattern. Quantitatively, the Machta model predicts more nonuniform fall-out with higher surface deposition levels at approximately  $40^\circ$  N. and  $40^\circ$  S. latitudes.

Entry of fission debris into the biosphere and man is a function of the soil-plant relationships and metabolic properties of the individual radionuclides. All radionuclides may enter the biosphere through direct deposition on vegetation and through the soil-to-plant-to-milk-to-man ecological cycle. Ecological discrimination may be expected to occur, and it is possible to estimate overall discrimination ratios for various countries from per capita consumption of principal foods and the dis-

crimination factors at the various steps along the ecological cycle. From these ratios it is possible to estimate average maximum equilibrium levels in the population under conditions of no more weapons tests and continued tests at the past 5-year rate.

Average maximum equilibrium  $\text{Sr}^{90}$  bone levels may be estimated from  $\text{Sr}^{90}/\text{Ca}$  ecological discrimination factors and from current bone analyses data. On the basis of ecological discrimination, present population weighted  $\text{Sr}^{90}$  equilibrium bone levels might be expected to be about  $2.8 \mu\mu\text{c/g Ca}$ . Estimates from bone analyses data suggest equilibrium levels of about  $1.5 \mu\mu\text{c/g Ca}$ . With cessation of tests, these estimates may be expected to increase by about 10% as a result of continued fall-out from the stratospheric reservoir. If weapons tests are continued until equilibrium (in about 100 years), average maximum equilibrium bone levels may be a factor of 10 greater than present levels.

Isotopes of plutonium produced by nuclear detonations appear to pose no potential hazard because of the very great discrimination factors operating against its transmission through ecological cycles. With continued tests, plutonium will never reach an equilibrium level because of its 24 000-year half-life. Its build-up in the biosphere, however, will not be great.

The present population weighted world average  $\text{Cs}^{137}$  level in man is about  $32 \mu\mu\text{c/g K}$ . It is not clear at present the extent to which cesium is entering the biosphere via direct fall-out and via the ecological cycle. If it is entering through direct fall-out, present levels may represent a quasi-equilibrium with the rate of stratospheric fall-out. In this case, continued tests will not result in further increases of  $\text{Cs}^{137}$  in people. If it is entering via the ecological cycle, it will integrate in the biosphere, and continued testing at the present rate for about 100 years may result in population levels about 10 times present values.

$\text{I}^{131}$ , because of its 8-day half-life, can enter the biosphere only by direct inhalation and contamination of foods by direct fall-out.  $\text{I}^{131}$  levels in thyroids of domestic animals have averaged from 0.01 to  $0.5 \text{ m}\mu\text{c/g}$  of thyroid. Levels in human thyroids are lower than those of cattle by a factor of about 100.  $\text{I}^{131}$  levels, although highly dependent on variations in tropospheric meteorology and the proximity to nuclear test sites, may be regarded as a quasi-equilibrium condition with the past test rate, and continued testing should not result in further increase in the general average population level.

Only two radionuclides from fission debris ( $\text{Sr}^{90}$  and  $\text{Cs}^{137}$ ) appear likely as potential hazards to man's general health and well-being. The biological significance of present and future levels of these isotopes is

discussed. These considerations suggest that the past rate of testing, if continued for several years, will not produce internal radiation levels that will exceed the recommended general population maximum permissible levels set forth by the National and International Commissions on Radiological Protection. Although this leads to the conclusion that the present rate of biospheric contamination poses no serious potential somatic hazard to world health, the great uncertainties involved make it imperative that the problem be kept under constant scrutiny, if weapons tests are to continue. Fortunately, present levels are not critical and the slow rate of biospheric build-up affords time for continued intensive and extensive study.

### *Zusammenfassung*

Kernwaffenversuche Mitte 1957 haben etwa 5,5 mC  $\text{Sr}^{90}$  und unterschiedliche Mengen anderer langlebiger und mittellanglebiger Isotope hervorgebracht. Von diesen stellt  $\text{Sr}^{90}$  für die Weltgesundheit die größte potentielle Bedrohung dar.  $\text{Cs}^{137}$  und  $\text{Pu}^{239}$  sind die anderen hauptsächlichen langlebigen radioaktiven Substanzen in Atomwaffentrümmern.  $\text{Sr}^{90}$  und  $\text{Cs}^{137}$  wurden bei Menschen, in der Milch und in anderen Nahrungsmitteln mengenmäßig bestimmt und das mittellanglebige  $\text{J}^{131}$  in der Schilddrüse von Menschen und Haustieren ermittelt.

Zwei Theorien der Verteilung der Zerfallsprodukte von Kernwaffen über die ganze Erde wurden vorgeschlagen. Das Libby-Modell postuliert ein einheitliches rasches Vermischen der Zerfallsprodukte in der ganzen Stratosphäre mit gleichmäßigem Zurücksickern durch die Tropopause; nachher erfolgt die Verteilung über die Erdoberfläche entsprechend den meteorologischen Bedingungen der Troposphäre. Das Machta-Modell verfißt eine langsame ungleichmäßige Vermischung in der Stratosphäre mit einem Niederschlag durch die Tropopause, der hauptsächlich in der Nähe von Jet-Strömungen erfolgt. Qualitativ zeigen beide Modelle das gleiche Verteilungsmuster. Quantitativ sagt das Machta-Modell uneinheitliche radioaktive Niederschläge mit mengenmäßig größeren Ablagerungen bei annähernd  $40^\circ$  nördlicher und  $40^\circ$  südlicher Breite voraus.

Das Eindringen der Zerfallsprodukte in die Biosphäre und in den menschlichen Organismus ist eine Funktion der Beziehungen zwischen Boden und Pflanzen und eine Funktion der Stoffwechseleigentümlichkeiten der einzelnen radioaktiven Isotope. Alle Isotope können durch direkte Ablagerung auf die Vegetation in die Biosphäre gelangen und durch den ökologischen Zyklus vom Boden über die Pflanzen in die Milch und den menschlichen Organismus. Ökologische Diskriminierung ist zu erwarten, und es ist möglich, die allgemeinen Diskriminierungs-



quotienten in verschiedenen Ländern aus der Totalaufnahme der Hauptnahrung und den Diskriminierungsfaktoren entlang den verschiedenen Stufen des ökologischen Zyklus abzuschätzen. Mit diesen Quotienten kann das Mittel der größten Gleichgewichtsaktivitäten in der Bevölkerung berechnet werden, bei Einstellung der Kernwaffenversuche einerseits und bei Fortsetzung der Versuche im gleichen Rhythmus wie in den letzten 5 Jahren andererseits. Das Mittel der größten durchschnittlichen Gleichgewichtsaktivität von  $\text{Sr}^{90}$  in Knochen kann aus den Diskriminierungsfaktoren von  $\text{Sr}^{90}/\text{Ca}$  und aus den Ergebnissen laufender Knochenanalysen errechnet werden. Auf Grund der ökologischen Diskriminierung mag die gegenwärtig gemessene durchschnittliche Gleichgewichtsaktivität von  $\text{Sr}^{90}$  des menschlichen Knochens auf etwa  $2,8 \mu\mu\text{C}$  pro g Ca geschätzt werden. Schätzungen aus Knochenanalysen lassen eine durchschnittliche Gleichgewichtsaktivität von ungefähr  $1,5 \mu\mu\text{C/g}$  Ca vermuten. Im Falle eines Verzichts auf Kernwaffenversuche dürften sich diese Werte um ca. 10% erhöhen, infolge des weitergehenden Ausfalls aus der Stratosphäre. Wenn die Kernversuche fortgesetzt werden bis zum Gleichgewichtszustand (in ungefähr 100 Jahren), kann das Mittel der durchschnittlich größten Aktivität in Knochen um einen Faktor 10 größer werden.

Plutoniumisotope, die infolge von Kernexplosionen entstehen, scheinen wegen des sehr großen Diskriminierungsfaktors, welcher der Übertragung im ökologischen Zyklus entgegenwirkt, keine potentielle Bedrohung zu bilden. Bei fortgesetzten Versuchen wird Plutonium wegen seiner Halbwertszeit von 24 000 Jahren nie einen Gleichgewichtszustand erreichen. Seine Anreicherung in der Biosphäre wird jedoch nicht groß sein.

Der bei der jetzigen Bevölkerung bestimmte mittlere  $\text{Cs}^{137}$ -Gehalt im Menschen ist etwa  $32 \mu\mu\text{C/g}$  K. Welche Mengen von Caesium auf direktem Wege einerseits und durch den ökologischen Zyklus andererseits in die Biosphäre eindringen, ist noch nicht abgeklärt. Wenn es aus dem radioaktiven Niederschlag direkt eindringt, mag die gegenwärtige Aktivität einen Quasigleichgewichtszustand mit der Rate des stratosphärischen Niederschlages darstellen. In diesem Falle wird die Fortsetzung der Kernversuche bei der Bevölkerung keine Vermehrung des  $\text{Cs}^{137}$ -Gehalts verursachen. Wenn es durch den ökologischen Zyklus eindringt, so integriert es in die Biosphäre und im gleichen Rhythmus wie bisher fortgesetzte Kernversuche werden in etwa 100 Jahren den  $\text{Cs}^{137}$ -Gehalt in der Bevölkerung um das 10fache erhöhen.

$\text{J}^{131}$  kann infolge seiner Halbwertszeit von 8 Tagen nur durch direkte Inhalation in die Biosphäre eindringen und durch Verseuchung der

Nahrung aus dem radioaktiven Niederschlag direkt. Die  $J^{131}$ -Aktivität der Schilddrüse von Haustieren erreicht durchschnittlich 0,01–0,5  $m\mu C$  pro g Schilddrüse. Der Gehalt menschlicher Schilddrüsen ist um einen Faktor von ungefähr 100 niedriger als jener beim Vieh. Obschon der  $J^{131}$ -Gehalt von den meteorologischen Schwankungen in der Troposphäre ebenso wie von der Nähe eines Kernversuchsgeländes stark abhängig ist, kann angenommen werden, daß er sich mit den vergangenen Versuchsraten in einem Quasigleichgewichtszustand befindet; die Fortsetzung der Versuche sollte deshalb keinen weiteren Anstieg des mittleren  $J^{131}$ -Gehaltes der Bevölkerung bringen.

Nur zwei der radioaktiven Kerne in Zerfallsprodukten ( $Sr^{90}$  und  $Cs^{137}$ ) scheinen eine potentielle Bedrohung der menschlichen Gesundheit und des Wohlbefindens zu bilden. Die biologische Bedeutung des gegenwärtigen und zukünftigen Gehaltes dieser Isotope wird diskutiert. Diese Beobachtungen lassen vermuten, daß die bisherigen Versuche, sollten sie während einiger Jahre im gleichen Rhythmus fortgesetzt werden, keine mittleren Strahlendosen erzeugen werden, welche größer sind als die von den nationalen und internationalen Kommissionen für Strahlenschutz für die gesamte Bevölkerung als maximal erlaubt bezeichnete Dosis. Trotzdem dies zum Schluß führt, daß die gegenwärtige Rate der Verseuchung der Biosphäre keine ernsthafte potentielle Bedrohung der somatischen Gesundheit der Bevölkerung der ganzen Welt darstellt, macht es die große, allgemein existierende Ungewißheit aber unbedingt notwendig, daß das Problem, sollten Kernstoffversuche noch weitergeführt werden, ständig unter genauer Kontrolle gehalten wird. Glücklicherweise sind die gegenwärtigen Dosen nicht kritisch, und die langsame Anreicherung in der Biosphäre gibt uns Zeit für fortgesetzte intensive und extensive Untersuchungen.

### *Résumé*

Jusqu'au milieu de 1957, les essais d'armes atomiques ont produit environ 5,5 mC de  $Sr^{90}$  et des quantités variables d'autres isotopes de périodes moyennes ou longues. Parmi ceux-ci, le  $Sr^{90}$  présente la plus grave menace potentielle pour la santé des hommes.  $Cs^{137}$  et  $Pu^{239}$  sont les autres radio-isotopes importants de longue période qui se trouvent dans les débris des explosions atomiques.  $Sr^{90}$  et  $Cs^{137}$  ont été mesurés quantitativement chez l'homme, dans le lait et d'autres aliments; le iode<sup>131</sup>, de période moyenne, a été décelé dans les glandes thyroïdes de l'homme et des animaux domestiques.

Deux théories ont été proposées pour expliquer la distribution des résidus d'explosions atomiques dans le monde. Le modèle de Libby

postule un mélange uniforme et rapide des débris d'explosions atomiques dans toute la stratosphère, avec retombée uniforme à travers la tropopause, puis déposition à la surface du sol conformément aux conditions météorologiques régnant alors dans la troposphère. La théorie de Machta propose, au contraire, un mélange lent et inégal dans la stratosphère, avec retombée inégale au travers de la tropopause, plus importante à proximité des discontinuités de celles-ci. Au point de vue qualitatif, les deux théories aboutissent à peu près au même résultat. En revanche, sur le plan quantitatif, la théorie de Machta prévoit des retombées radioactives non uniformes, avec une concentration maximum aux latitudes de 40° N et de 40° S.

La pénétration des produits de fission dans la biosphère et dans l'organisme humain s'effectue en fonction des relations existant entre le sol et les plantes, et des propriétés métaboliques individuelles des radioisotopes. Tous les radio-éléments peuvent pénétrer dans la biosphère par dépôt direct sur les végétaux, ou par l'intermédiaire du cycle écologique sol-plante-lait-homme. Une discrimination écologique doit se produire et il est possible d'apprécier le rapport total des discriminations pour divers pays, à partir du taux de consommation des aliments principaux et des divers facteurs de discrimination partiels tout au long du cycle écologique. A partir de ces rapports, il est possible d'estimer les niveaux maxima moyens de contamination des populations qui seront atteints, soit dans le cas où les essais nucléaires seraient interrompus, soit dans celui où ils continueraient au même rythme que ces cinq dernières années.

Les taux maxima moyens de  $\text{Sr}^{90}$  dans les os, lorsque l'équilibre est atteint, peuvent être estimés à partir des facteurs de discrimination écologiques  $\text{Sr}^{90}/\text{Ca}$  et des valeurs données par l'analyse chimique courante des os. Sur la base de cette discrimination écologique, le taux de  $\text{Sr}^{90}$  à l'équilibre dans les os, pour la population actuelle, peut être estimé à environ  $2,8 \mu\text{C/g Ca}$ . Les estimations faites à partir des analyses des os laissent prévoir un taux d'environ  $1,5 \mu\text{C/g Ca}$ . Avec la cessation des essais atomiques, il faut s'attendre à une augmentation de ces valeurs d'environ 10 %, due aux retombées stratosphériques. Si les essais atomiques sont continués, jusqu'à l'atteinte d'un état d'équilibre (en 100 ans environ), la valeur maximum moyenne, à l'équilibre, du taux de  $\text{Sr}^{90}$  dans les os sera multipliée par un facteur 10.

Les isotopes du plutonium produits lors des explosions nucléaires ne semblent pas présenter un réel danger, à cause des très grands facteurs de discrimination qui empêchent leur transmission au long du cycle écologique. Si les expériences nucléaires continuent, le plutonium ne pourra jamais atteindre une valeur d'équilibre à cause de sa période de 24 000 ans. Toutefois, son accumulation dans la biosphère ne sera pas grande.

La teneur moyenne de la population mondiale en  $\text{Cs}^{137}$  est actuellement d'environ  $32 \mu\mu\text{C/g K}$ . On ne sait pas encore aujourd'hui dans quelle mesure le caesium pénètre dans la biosphère par retombée directe ou par transmission suivant un cycle écologique. Si le  $\text{Cs}^{137}$  est absorbé directement, le niveau actuel est en quasi-équilibre avec le taux des retombées stratosphériques. Dans ce cas, la continuation des expériences nucléaires n'entraînera pas d'augmentation de la teneur en  $\text{Cs}^{137}$  de l'homme. En revanche, s'il pénètre par un cycle écologique, il s'accumulera dans la biosphère, et la continuation des essais nucléaires pendant environ 100 ans, au rythme actuel, conduira à un taux de  $\text{Cs}^{137}$  environ 10 fois plus fort qu'actuellement.

L'iode<sup>131</sup>, à cause de sa période de 8 jours, ne peut entrer dans la biosphère que par inhalation directe ou par contamination des aliments par les retombées directes. Le niveau de  $\text{I}^{131}$  dans les glandes thyroïdes des animaux domestiques est de 0,01 à 0,5  $\text{m}\mu\text{C/g}$  de thyroïde. Dans la thyroïde de l'homme, ce niveau est environ 100 fois plus faible que chez le bétail. Les teneurs en  $\text{I}^{131}$ , bien que grandement variables avec les conditions météorologiques régnant dans la troposphère d'une part, et la proximité des lieux d'explosions des armes atomiques d'autre part, peuvent être considérées comme sensiblement en équilibre depuis les essais antérieurs; avec la continuation de ceux-ci, la teneur en  $\text{I}^{131}$  de la population ne sera guère augmentée, en moyenne.

Seuls deux radio-éléments produits par la fission nucléaire ( $\text{Sr}^{90}$  et  $\text{Cs}^{137}$ ) semblent présenter un réel danger pour la santé et l'avenir du genre humain. Les conséquences biologiques des niveaux actuels et futurs de ces deux isotopes sont discutées. Il apparaît que même si les essais nucléaires sont poursuivis pendant plusieurs années, au même rythme que jusqu'ici, la dose de radiations reçues, par irradiation interne, ne dépassera pas la dose maximum admissible pour la population, telle qu'elle est fixée par les Commissions Internationale et Nationale pour la Protection contre les Radiations Ionisantes. Quoiqu'il soit possible d'affirmer que le taux actuel de contamination de la biosphère ne présente pas un réel danger pour la santé humaine, les nombreuses inconnues de ce problème obligent à en continuer l'étude, si les expériences nucléaires devaient être continuées. Heureusement, les niveaux actuels ne sont pas critiques et la lenteur de l'accumulation dans la biosphère nous laisse du temps pour poursuivre une étude dense et complète du problème.

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