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MAXIMUM PERMISSIBLE LEVELS OF INTERNALLY-DEPOSITED RADIOISOTOPES IN RELATION TO WORLD-WIDE FALLOUT

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INTRODUCTION

During the past year public attention has been increasingly focused on the potential hazard to the general population of widespread, lowlevely radioactive fallout from nuclear weapons testing (1-50, Although a number of radioisotopes are present in the fission mixture, the principal concern is Sr⁹⁰. Sr⁹⁰ is believed to be the most important isotope because of its similarity to calcium, its long physical and biological half-life, and high relative fission yield. These factors biological materials lead to high incorporation in the blosphere and a long residence time animal in bone. General contamination will result in the bones of the population eventually reaching an equilibrium state with Sr⁹⁰ in the bio-The predominance of Sr⁹⁰ over other long-lived radioelements sphere. as a potential hazard can be deduced in part from data in Table 1, which show that it is the only isotope that combines high fission yield, long half-life, high absorption rate and a low maximum permissible level. These data suggest Cs¹³⁷ as the second most important fission product with report of the long. Firm handly, and its presence in people and foodstuffs has been reported (6, 7). However, for reasons not discussed here, its potential hazard to the population is believed to be less than Sr⁹⁰ by at least an order of magnitude.

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RADIOELEMENTS OF IMPORTANCE TO LONG-TERM FALLOUT PROBLEM

Table 1

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Radio- element	Fission Type Abundance Radiation (per cent)		Radiological Half-Life	Abs. on Ingestion (per cent)	MPL (µc)			
Pu ²³⁹	α		24,000 yr	3 x 10 ⁻³	0.04			
Cs ¹³⁷	β,γ	6.2	28 yr	100	98			
Sr ⁹⁰	β	5.1	28 yr	35	1			
Pm ¹⁴⁷	β	2.6	3.7 yr	3 x 10 ⁻²	25			
Ru-Rh ¹⁰⁶	β,γ	0.5	1.0 yr	5 x 10 ⁻²	4			
Ce-Pr ¹⁴⁴	β,γ	5.3	275 days	3 x 10 ⁻²	1			

Appraisal of the potential hazard from world-wide fallout requires consideration of the present basis for the maximum permissible levels of internally-deposited isotopes, the rate and extent of fallout, and the method of incorporation of radionuclides into the biosphere and the human body. Information on all of these factors is somewhat inadequate at the present time. This paper is an attempt to present a general summary of the present thinking with regard to the above factors.

BASIS FOR MAXIMUM PERMISSIBLE LEVELS OF INTERNALLY-DEPOSITED RADIOISOTOPES

Human experience through the diagnostic and therapeutic use of Xand gamma rays and extensive animal experimentation with all types of radiation have demonstrated that ionizing radiations may produce deleterious biological effects. These effects are manifest in the production of early physiologic aging resulting in life shortening, mutations, irregularities in hematopoietic function (some of which result in increased incidence of leukemia) and specific organ or tissue changes such as cataract of the lens of the eye and tumors of the bone. Damage from ionizing radiations may occur from radioactive isotopes deposited in the tissues and organs, as well as from radiations originating from an external source.

Maximum permissible amounts of radioisotopes deposited in the body are calculated on the premise that no critical organ or tissue will receive an average dose rate greater than 300 mrem per week (the maximum permissible dose rate for external whole body radiation), assuming uniform distribution of the isotope throughout the tissue or organ. It is recognized that calculations based on average dose or uniform distribution in a critical organ or tissue may lead to considerable error since some radioisotopes are unevenly deposited. Although the average dose

rate to a critical organ may be 300 mrem per week, some portions of the organ may receive considerably less than the average and others correspondingly more. This error, however, is believed to be offset at least in part by the fact that 300 mrem is considered the acceptable weekly exposure to the entire body or the blood-forming organs and not a permissible exposure to a small element of tissue.

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The Maximum Permissible Body Burden $q(\mu c)$ of a radioisotope, based on the weekly dose rate of 300 mrem for external radiation, is calculated according to the following expression (8):

$$q = \frac{100 \text{ m W}}{3.7 \times 10^{4} \times 1.6 \times 10^{-6} \times 6.05 \times 10^{5} \times f_{2} \times \Sigma \text{ E(RBE)N}} = \frac{8.4 \times 10^{-4} \text{ m}}{f_{2} \Sigma \text{ E(RBE)N}}$$

in which 100 = ergs/g/rad
$$m = \text{mass of critical organ in grams}$$
$$W = \text{permissible dose of external radiation (0.3 rem/wk)}$$
$$3.7 \times 10^{4} = \text{d/sec/}\mu\text{c}$$
$$1.6 \times 10^{-6} = \text{ergs/mev}$$
$$6.05 \times 10^{5} = \text{sec/wk}$$
$$f_{2} = \text{fraction in critical organ of that in total body}$$
$$\Sigma \text{ E(RBE)N} = \text{weighted energy of absorbed radiation, weighted}$$
for summation of energies of all decays, for their relative biological effectiveness (RBE) and for nonhomogeneous distribution (N).

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For some isotopes that localize in bone the maximum permissible body

burdens are determined by direct comparison with the 0.1 μ c maximum multiply different multiply multiply permissible burden for radium (an <u>internet emitter</u> with which there has been considerable human experience) and not with the 300 mrem per week level for whole body external radiation. In these cases, a factor of safety of 5 is introduced to take into account the uneven distribution of the radioactive material within the bone. Comparison with 0.1 μ c of radium is made in the following manner (8):

 $q = \frac{q_{(Ra)}f_{2(Ra)}\left[\Sigma E(RBE)N\right]_{(Ra)}}{f_{2}\Sigma E(RBE)N} = \frac{16}{f_{2}\Sigma E(RBE)N}$

in which the various symbols have the same definitions given above. Derivations of the above formulas, their parameters, pertinent information on individual nuclides and values for q are given in the Handbooks of the International (8) and the National (9) Commissions on Radiological Protection. These handbooks provide the only official sources of such information for the various radionuclides, from thestendpoint-of-internal-absorbed dose. All of the MPC values presently published in the handbooks refer to continuous occupational exposure.

BASIS FOR MAXIMUM PERMISSIBLE LEVELS APPLIED TO THE GENERAL POPULATION

Maximum permissible levels for non-occupational exposure or exposure of a large segment of the general population were established by taking arbitrarily 1/10th of the value for working personnel (8-10). The

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rationale behind a lower value for the general population is based on the numbers involved in the two groups at risk and the increased heterogeneity of the general population over that of the select working group. The latter group is composed of supposedly healthy workers (over 20 years of age), while the general population group **may** containSchildren, pregnant women, the undernourished, the sick and the old. On the assumption that frequency of response to radiation stress follows a Gaussian distribution (Fig. 1), the probability of injury of a few individuals from a specified dose increases with increase in size and heterogeneity of the group at risk.

The maximum permissible level of Sr^{90} for workers is set at $1 \mu c$ on the basis of comparison with radium, while the recommended level for the general population is set at 0.1 μc^* (11-13). The permissible level of Sr^{90} is predicated on the assumption that chronic and/or delayed effects of radiation are threshold phenomena (Fig. 2). That is to say, there is a threshold dose below which effect rapidly becomes insignificant and above which effect increases exponentially over a limited dose range. If this is indeed the case, 100 mµc $\mathrm{Sr}^{90}/\mathrm{kg}$ Ca must be looked upon as a true maximum permissible level and not an average level.

^{*} There is about 1 kg of calcium in the adult human skeleton; therefore, the MPL of Sr90 in the general population is equivalent to $0.1 \ \mu c/kg$ Ca = 100 $\mu \mu c/kg$ Ca = 100 $\mu \mu c/g$ Ca = 100 Sunshine Units (Libby).



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Fig. 1. Frequency distribution response as a function of dose.

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DOSE (rel.)

As yet, there is not conclusive evidence of a threshold dose for such chronic effects as leukemia, bone tumors, etc. There is, in fact, some indication that genetic response to external radiation is linear with dose and that a given increment produces a corresponding equal increment of effect, regardless of position on the dosage scale (Fig. 2). If chronic effects of radiation are not threshold phenomena, it would seem more reasonable to establish permissible levels for the general population on the basis of probability of risk averaged over the entire group. Present incidence of bone sarcoma and leukemia averaged over the entire population is about 2 and 6 per 100,000, respectively. About 15% of the natural incidence of leukemia (and presumably of bone sarcoma) may be attributable to natural radiation background (14). If this is true, doubling the natural background dose might be expected to increase the incidence of bone tumors and leukemia to 2.3 and 6.9 per 100,000, respectively. Such a small increase may be undetectable in the general Permissible population. On this basis, one might justify an average Sr⁹⁰ level of 100 mic/kg Ca in the general population. Dr. Hardin Jones (14), on the basis of vital statistics data, has stated that an average of 50 rads per life-time might reasonably be assumed to be a doubling dose for chronic radiation effects. On the basis of this value, an average strontium burden of 500 muc/kg Ca in the general population would not be expected to double the incidence of bone sarcoma and leukemia.

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GENERAL WORLD-WIDE FALLOUT FROM BOMB TESTING OPERATIONS

Based on measurements of world-wide fallout, Libby (2, 3) has proposed a mechanism by which atomic debris is disseminated throughout the world. This theory leads to three kinds of fallout, which are illustrated presented graphically in Fig. 3. First is local fallout which 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 residues from the soil and structures which are swept into the cloud in wholly or partially vaporized state. The fraction of the total radioactivity which falls out locally depends very much on those conditions of firing which govern the amount of soil and extraneous debris incorporated in the fireball.

The second type, tropospheric world-wide fallout, is the material, which though not coarse enough to fall out locally, is left suspended in the atmosphere below the tropopause. This material is sufficiently fine that it travels great distances, circling the earth in the general latitude of the explosion, until scrubbed from the atmosphere by rain, fog, contact with vegetation, and other meteorological and/or physical factors. The average tropospheric fallout time is estimated as 20 to 30 days. The fraction of the fallout which is in this category depends

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Fig. 3. Types of radioactive fallout from weapons tests (local, tropospheric and stratospheric).



mainly on the size of the explosion and the conditions of firing. If the weapon is fired close to the ground, a greater percentage of the fission products will deposit as local fallout. If the explosion is large (greater than 1 MT), a large percentage will be carried above the tropopause.

The third type (stratospheric fallout) is composed of fission products that are carried above the tropopause. These are believed to mix throughout the stratosphere and fall out uniformly over the entire surface of the earth with a mean fallout time estimated at from 6 to 10 years.

The above mechanism leads to a general distribution pattern of radioactivity over the surface of the earth as shown in Fig. 4. Dr. Libby's estimates (3) of present levels of Sr^{90} deposition suggest 22 mc/mi² for the upper midwestern section of the United States, 15 to 17 mc/mi² for the area between about 50°N and 10°S latitude, and 3 to 4 mc/mi² for the rest of the world. The higher value for the upper midwestern United States is attributed to greater local and tropospheric fallout because of the proximity of our own continental test site. The 15 to 17 mc/mi² deposited between about 50°N and 10°S latitude is due to tropospheric fallout from all tests of less than 1 MT conducted in the northern hemisphere plus stratospheric fallout from all tests of the rest of the earth is due entirely to stratospheric fallout from all tests of greater than 1 MT. Actually, the general picture is greatly oversimplified.

Fig. 4. General levels of world-wide fallout deposition.

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Once fission products are suspended in the troposphere (either directly by the detonation or from diffusion back below the tropopause from the stratospheric pool), meteorological conditions play a major role in their deposition. Libby (3) has stressed the importance of rainfall, fog and mist. Within any major fallout area one might expect to find fluctuations in the level of surface deposition which correlate with local meteorological conditions. Higher deposition in a local area may not correlate necessarily with total precipitation but rather with the frequency.

PRESENT AND PREDICTED MAXIMUM LEVELS OF SURFACE DEPOSITION

Libby (3) has estimated that the stratospheric reservoir contains the products of about 24 MT of fission and preliminary direct measurements by means of high altitude balloons suggest that this value is approximately correct. One MT of fission results in the formation of enough Sr^{90} to give a surface deposition of 0.5 mc/mi² if uniformly distributed over the entire earth's surface. If all material presently in the stratospheric reservoir were deposited instantaneously and uniformly over the earth, present values would be increased by 12 mc/mi² and the maximum surface deposition of Sr^{90} would result. Maximum deposition, however, will not occur because of the relatively long average stratospheric fallout time (6 to 10 years), which will allow some of the strontium to decay before deposition. Figure 5 shows, however, that the predicted maximum level is not highly dependent on the mean time of fallout.



Fig. 5. Dependence of maximum level of deposition on mean time of fallout.



Although meteorologists appear to favor a fallout half-time of about 4 years, Dr. Libby (2) has chosen to use a value of 7, which corresponds to a mean time of about 10 years. With a mean time of 10 years, the maximum predicted level of Sr^{90} surface contamination should occur in about 1975. Table 2 shows the estimated present levels and the maximum predicted levels that might be expected in about 1975.

Table	2
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PRESENT AND PREDICTED MAXIMUM LEVELS OF Sr⁹⁰ SURFACE DEPOSITION*

Area	Level October 1956 (mc/mi ²)	Maximum Level 1975 (mc/mi ²)
Midwest United States	22	29
Between 50 ⁰ N - 10 ⁰ S Latitude	16	23
Rest of World	3.6	10

* Assuming products of 24 MT fission in the stratosphere January 1, 1957, and a fallout mean time of 10 years.

As stated previously, these figures assume uniform world-wide distribution of the material now in the stratospheric reservoir and no more weapons tests. Under these conditions, the area in the upper midwestern United States would be expected to reach a level of about 29 mc/mi². The area between $50^{\circ}N$ and $10^{\circ}S$ latitude may reach about 23 mc/mi², and the rest of the world may reach a level of about 10 mc/mi².

INCORPORATION OF Sr⁹⁰ INTO THE BIOSPHERE

When Sr^{90} falls upon the earth's surface, it becomes mixed with calcium and is taken into the biosphere by efficient transmission through ecological chains. That which settles diectly on vegetation may remain as surface contamination or it in part may enter the plant through foliate absorption and mix with the plant calcium. The Sr^{90} that falls out on the soil is mixed with the available calcium in the soil, where it is taken in through the root system of the plant, along with calcium. When plants are eaten by animals, Sr^{90} deposited directly on the surface or incorporated in the plant is taken up by the animal and incorporated with the animal calcium. When plant and animal products (i.e., milk) are eaten by man, the Sr^{90} is incorporated into his calcium pool.

It is reasonable to assume that strontium may be discriminated against with respect to calcium in passing up the ecological chain. For example, the $\mathrm{Sr}^{90}/\mathrm{Ca}$ ratio in the bones of people may be expected to be lower than the $\mathrm{Sr}^{90}/\mathrm{Ca}$ ratio in the soil, which is the beginning ultimately of the ecological chain between man and his environment.

One method of obtaining information on the general discrimination between Sr^{90} and calcium in passing up the ecological chain is to consider men's equilibrium level with respect to stable strontium in the

environment (15, 16). Recent comparison of the ratio of stable strontium to available calcium in the soils and rocks of the world suggest an overall discrimination against strontium by a factor of about 10

(i.e.,
$$\frac{(Sr^{s}/Ca)_{b}}{(Sr^{s}/Ca)_{a}} = \sim \frac{1}{10}$$
). Recently Comar and others (17-19) have

attempted to determine the discrimination factors between Sr^{90} and calcium corresponding to the various steps in the ecological cycle. They have attempted to determine $\frac{(\mathrm{Sr}^{90}/\mathrm{Ca})_{\mathrm{sample}}}{(\mathrm{Sr}^{90}/\mathrm{Ca})_{\mathrm{precursor}}}$ going from soils

to plants, from plants to milk, from milk to man, and from plants to man. Assuming 80% of children's dietary calcium comes from milk and 20% comes directly from plants, an overall discrimination ratio in going from soils to human bone via the diet can be estimated. These data are summarized in Fig. 6, which suggest that the $\frac{(Sr^{90}/Ca)_{b}}{(Sr^{90}/Ca)_{diet}}$

 $\frac{1}{6}$ to $\frac{1}{9}$ (10). Considering the uncertainties involved in the various individual discrimination factors, this is in reasonable agreement with the general value derived from consideration of man's equilibrium with stable strontium in his environment. The uncertainties in the individual discrimination factors at the various steps along the ecological chain suggest a very important field of research for the next several years.

The values shown in Fig. 6 are general only and do not take into consideration a wide variety of variables, including effect of soil and



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plant type, soil and plant equilibration times, variability of available strontium and calcium in the soil, nature and source of the diet, age and state of nutrition of the individual, etc.

PREDICTED PRESENT AND FUTURE MAXIMUM Sr⁹⁰ LEVELS IN CHILDREN

From the data in Table 2 and the information in Fig. 6, it is possible to make a general prediction of the present and future maximum Sr^{90} levels in the bones of children, who draw their dietary calcium from the three major areas of fallout.

Assuming an average of 20 g available Ca/ft^2 of soil to a depth of 2-1/2 inches (2), a deposition of 1 mc Sr^{90}/mi^2 is equivalent to 1.8 mµc/kg of available soil calcium (1 kg is approximately the amount of calcium in the adult skeleton). Multiplication of the present and predicted maximum levels of Sr^{90} surface deposition (Table 2) by 1.8 gives the corresponding Sr^{90} content of the soils of the various areas in terms of mµc/kg available Ca. Assuming steady state conditions and $(\frac{Sr^{90}/Ca}{(Sr^{90}/Ca)_{s}} = \frac{1}{10}$, the predicted maximum present and future Sr^{90} levels in the bones of children expressed as mµc/kg bone Ca will be approximately 1/10th of the level in the available soil calcium. Present and future maximum Sr^{90} levels in children calculated in this manner are shown in Table 3. Perhaps the largest uncertainty in these estimations (even greater than the uncertainty in the Sr^{90}/Ca discrimination ratio) is due to their dependence on available soil calcium, which may vary

within the United States from about 1 to 100 g/ft^2 to a depth of 2-1/2 inches. Likewise, the amount of available soil calcium with which the Sr^{90} may be assumed to be in equilibrium is a function of the depth of the feeding zone of various types of plants.

Table 3

Area	Predicted Max.** Spring 1957 (mic/kg Ca)	Predicted Max. 1975 (mic/kg Ca)
Midwest United States	4.0	5.2
Between 50 ⁰ N - 10 ⁰ S Latitude	2.9	4.1
Rest of World	0.6	1.8

PREDICTED PRESENT AND FUTURE MAXIMUM Sr⁹⁰ LEVELS IN CHILDREN*

* Assuming steady state conditions, 20 g available Ca/ft^2 of soil, mean fallout time of 10 years, and $\frac{(Sr^{90}/Ca)_b}{(Sr^{90}/Ca)_a} = \frac{1}{10}$.

** Samples of children's bones showed a level of $\sim 1 \text{ muc/kg}$ Ca January 1956. MPL Sr⁹⁰ for general population = 100 muc/kg Ca.

The data in Table 3 suggest a present maximum Sr^{90} level in bones from the midwestern United States of 4 m_µc/kg Ca and 2.9 m_µc/kg Ca for the general area between about 50°N and 10°S latitude. Bone samples collected at various points in the tropospheric band during the first half of 1956 show levels of about 1 m_µc/kg Ca in children (3). Extrapolation from earlier results would give a slightly higher value for

December 1956. There are a number of reasons why the measured values might be expected to be lower than the predicted present maximum.

(a) It is quite unlikely that a steady state condition between strontium surface deposition and Sr^{90} in bones has been reached.

(b) The bone samples were from children of various ages and much of their calcium may have come from the soils long before the maximum surface levels given in Table 2 were laid down.

(c) It is possible that the calcium in the bone samples was drawn from soils having greater than 20 g available Ca/ft^2 to the depth of the plant feeding zone.

(d) It is possible also that the average discrimination ratio against strontium with respect to calcium in going from soil to bone is greater than a factor of 10.

A wide variety of milk samples collected from the Chicago (3) and New York (4) areas during 1956 showed an average level of about 3 mµc $\frac{Sr^{90}}{\text{kg Ca.}}$ Assuming Dr. Comar's discrimination factor $\frac{(Sr^{90}/\text{Ca})_{\text{b}}}{(Sr^{90}/\text{Ca})_{\text{m}}} = \frac{1}{2}$

in going from bone to milk, samples of newly formed bone derived from milk calcium from these areas might be expected to show a maximum of about 1.5 mic/kg Ca. This value is not seriously out of line with the measured values extrapolated to December of 1956.

On the basis of New York milk-shed data, Eisenbud (4) estimated 8 m_lc/kg skeletal Ca as the upper limit of the foreseeable Sr^{90} burden of the population in the New York area from bomb detonations that have

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already occurred. Admittedly, his estimate was pessimistic since it ignored the possibility that much of the Sr^{90} in milk may be ingested by cows as fresh fallout on the surface of plants and also assumed no discrimination between calcium and strontium during the process of converting milk to human bone. Had he used a discrimination factor of $\frac{1}{2}$, his estimate would have given 4 muc/kg bone Ca, which is in good agreement with Libby's value (2, 3) and the value shown in Table 3.

The most troublesome feature of the above considerations is that they are based on broad averages and make no allowance for individual variations due to local meteorological effects on fallout, soil properties, and dietary habits. Dr. Eisenbud (4) chose arbitrarily a factor of 3 as ample to define the upper limit of hazard that may be anticipated within the United States. Application of this factor to the predicted future maximum Sr^{90} levels given in Table 3 gives 15, 12 and 6 mµc/kg Ca as the maximum local limit for the upper midwestern United States, the area between about 50°N and 10°S latitude, and the rest of the world, respectively. The probability is high that these numbers are on the conservative side.

SIGNIFICANCE OF PRESENT AND PREDICTED Sr⁹⁰ LEVELS IN THE POPULATION

The significance of present and predicted levels of Sr⁹⁰ in bone can be evaluated only in relation to past human experience, which is indeed inadequate. Experience with a limited number of cases of radium

exposure has indicated conclusively that small amounts of radium fixed in the skeleton will produce osteoporosis, necrosis, and sarcoma (21). Bone changes in radium dial painters and persons who received radium therapeutically provide the basis for the value of 0.1 μ c as the maximum permissible level of fixed radium in the skeleton. Bone sarcoma has resulted from a fixed skeletal burden of only 2 to 3 μ c of pure Ra²²⁶. Nondeleterious bone changes have been observed in persons having only 0.4 μ c for a period of 25 years. Necrosis and tumors of the bone have occurred also several years after large doses of X ray (22).

The only other data with which present and predicted levels of Sr^{90} may be compared are natural background radiation levels. Natural background dose to the bone (during a 70-year life-time) may vary from ~ 6 to ~ 35 rads. The major contribution to background variation is differences in the radium levels of soils and minerals. The natural radiation level to bone, estimated by Spiers (23), is shown in Table 4.

Table 4

NATURAL	BACKGROUND	RADIATION	LEVEL	то	BONE	(SPIERS)	

			Dose to Bone				
Conditions	Ra in Skelet	on	External (rem/yr)	Ra (rem/yr)	Total (rem/yr)	Total (rem/70 yrs)	
Average	10-10	g	0.08	0.037	0.12	8.4	
Maximum natural radiation areas	10 ⁻⁹	g	0.1 8	0.37	0.55	38.5	

Table 5 gives a general summary of estimated skeletal radiation doses from accepted maximum permissible levels and from present and predicted Sr⁹⁰ burdens in relation to human experience. The maximum permissible level of Sr⁹⁰ is estimated to deliver about 8 rads 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 \sim 4 below the maximum natural background dose to which 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. According to these data, the present measured levels of Sr⁹⁰ in children will result in a life-time radiation dose of approximately 1% of the accepted maximum permissible level for the general population. The predicted average levels of Sr in about 1975 correspond to a maximum skeletal radiation dose, assuming no further weapons tests, of 2 to 5% of the maximum permissible level. Dependence of Sr^{90} fallout and uptake on meteorological conditions, soil factors, etc., might result in local dose levels of from 1 to 15% of maximum.

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Table 5. General Summary of Estimated Skeletal Radiation Doses in Relation to Human Experience د: د ر



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