BULLETIN OF THE TORREY BOTANICAL CLUB
VOL. 91, NO. 4, PP. 283–323 JULY–AUGUST 1964

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MARTIN, W. E. (Lab. of Nuclear Medicine and Radiation Biology, Dept. of Biophysics, Univ. of Calif., Los Angeles) *Radioecology and the study of environmental radiation*. Bull. Torrey Bot. Club 91: 283–323, 1964.—Among the major scientific problems of the Nuclear Age are those which deal with the collection of data and the development of concepts to be used in making realistic, quantitative evaluations of the biological hazards, if any, resulting from increased environmental radiation due to fallout. This paper presents a brief review of some of the ecological aspects of these problems. The major topics considered are: (a) the kinds and amounts of "natural" and "man-made" sources of ionizing radiation in the biosphere, (b) the formation and dispersal of fallout, including a comparison of local, tropospheric and stratospheric fallout patterns, (c) the redistribution of fallout materials by environmental processes, their accumulation by plants and animals, and their cycling in terrestrial food-chains, and (d) the evaluation of potential biological hazards arising from small increases in external and internal exposure of organisms to ionizing radiation.

Radioecology can be defined as the study of organisms and their external environments in relation to ionizing radiation. As a practical application of ecology to the study of fallout and reactor effluents, radioecology is primarily concerned with: (a) the influence of ionizing radiation on plant and animal populations and communities in their natural environments and (b) the influence of organisms and environmental processes on the distribution of radioactive materials in the biosphere.

Ionizing radiation has always been a part of the natural environments of living organisms. Speculations as to the possible influence of environ-

[1] These studies were supported by Contract AT (04-1) GEN-12 between the U. S. Atomic Energy Commission and the University of California at Los Angeles. The paper was originally presented at the Eighteenth Annual Research Conference of the Bureau of Biological Research at Rutgers University, New Jersey, April 27–28, 1962.
Received for publication February 12, 1964.

mental radiation on organisms began as early as 1896 (Müller 1896). The possibility that ionizing radiation has played an important role in evolution was recognized as early at 1928 (Olson & Lewis 1928).

During the past two decades, the "natural" sources of ionizing radiation in the biosphere have been augmented by the release of considerable quantities of "man-made" radioactive materials. The spread of radionuclides in fallout has been world-wide, and probably there is no place in the biosphere where the level of environmental radiation has not been increased to some extent. Ecological studies of the effects of nuclear detonations were made as early as 1948 (Bellamy 1949). Since then, the subject has been discussed at a variety of Congressional Hearings (Special Subcommittee 1957, 1959a, 1959b) and scientific meetings all over the world (A.I.B.S. 1962, Caldecott and Snyder 1960, Comar 1957, Dunning and Hilkin 1956, Health and Safety Laboratory 1958, Singleton 1958, Sparrow et al. 1958, United Nations 1955, and United Nations 1958).

This paper is a brief review of some of the currently available information concerning: (a) sources of environmental radiation, (b) the formation and dispersal of fallout, (c) the influence of organisms and of environmental processes on the redistribution of radioactive materials in the biosphere, and (d) the possible influence of increased environmental radiation on organisms. Its purpose is to outline a few of the ecological problems involved in evaluating the potential hazard, if any, of increased environmental radiation to populations of plants, animals, and men.

Sources of ionizing radiation in the biosphere. "NATURAL" SOURCES. The natural or "background" radiation to which organisms are exposed in their natural environments is derived either from cosmic radiation or from radionuclides. Radionuclides in environmental media are referred to as "external emitters" while the radionuclides assimilated by plants and animals are called "internal emitters."

1. Cosmic Radiation. Cosmic rays consist chiefly of protons and alpha particles which enter the earth's atmosphere from outer space. Nuclear reactions between cosmic rays and the atmosphere produce a variety of secondary radiations. At the lower altitudes, the ionization associated with cosmic radiation is caused primarily by protons, mesons, high energy electrons and photons.

The average intensity of cosmic radiation at sea level (Burch 1954) is about 0.1 mr/day (mr = milliroentgens). This increases with altitude to a maximum of about 15 mr/day at 70,000 feet. In the thinner air above 70,000 feet, ionization due to cosmic rays is less intense. At sea level the intensity of cosmic radiation increases slightly with increasing latitude.

2. External emitters. In addition to radium, thorium, uranium, and

their daughter products, Lowder and Solon (1956) have listed 17 other radionuclides which are known to occur in air, soil, or water.

Carbon-14 and tritium (H-3) are produced in the upper atmosphere by nuclear reactions between cosmic rays and molecules of nitrogen and water. The air near the ground may also contain small quantities of gaseous decay products which are derived from soil. According to one estimate

Table 1. *Estimated activity of naturally occurring radionuclides in an acre-foot of soil derived from igneous rock**

Radionuclides	Activity in curies/acre-foot	Radionuclides	Activity in curies/acre-foot
K-40	33×10^{-3}	Ra-226	1.5×10^{-3}
Rb-87	6×10^{-3}	Th-232	1.9×10^{-3}
C-14 & H-3	**	U-238	2.0×10^{-3}

* The formula for these calculations and those in Table 2 is:
$C = SA \times AI \times AE \times$ grams/acre-foot
C = Activity of radionuclide in curies per acre-foot
SA = Specific activity of radionuclides in curies per gram
AI = Abundance of radionuclides ($\% \times 10^{-2}$) in natural element
AE = Abundance of element ($\% \times 10^{-2}$) in environmental media
Grams per acre foot =
Soil: 1.5×10^{6}
Water: 1234×10^{6}
Sources of basic data:
"SA": Kinsman (1957)
"AI": Handbook of Physics and Chemistry, 39th ed.
"AE": Lowder & Solon (1956)
** C-14 in organic matter amounts to about 1×10^{-12} c/g carbon
H-3 in soil water amounts to about 3×10^{-10} c/g water

Table 2. *Estimated activity of naturally occurring radionuclides in an acre-foot of sea water**

Radionuclides	Activity in curies/acre-foot	Radionuclides	Activity in curies/acre-foot
K-40	39×10^{-3}	Ra-226	62×10^{-4}
Rb-87	32×10^{-10}	Th-232	***
C-14 & H-3	**	U-238	***

* See footnote following Table 1.
** C-14: ca. 1×10^{-12} c/g of Carbon
H-3 ca. 1×10^{-10} c/g = ca. 1.2×10^{-3} c/acre-foot of Sea Water
*** Not calculated.

(Koval 1945) the gas which emanates from soils containing radium and other long-lived radio-elements has an average activity of ca. 2×10^{-9} c/m^3 (c/m^3 = curies per cubic meter). Most of this is due to the presence of radioisotopes of radon (Rd-219,220,222) produced by the decay of Ra-226. Hess (1943) has reported that the radon content of air near the ground is usually less than 1×10^{-11} c/m^3, but one sample, collected during calm winter weather from a low area having no snow cover, contained 3×10^{-9} c/m^3 of radon.

The principal radionuclides in the earth's igneous crust, and therefore in most soil types, are K-40, Rb-87, Ra-226, Th-232, U-238, and the daughter products of the last three (Alexander et al. 1960). The C-14 content of soils is, of course, quite variable but amounts to approximately 1.0×10^{-12} curies of C-14 per gram of carbon. The theoretically possible concentrations of most of these radionuclides in igneous rock or in soil derived from igneous rock are shown in Table 1.

Estimates for other kinds of materials would vary in several respects. Potassium, for example, is less abundant in sandstone than in granite. Its abundance in shale and limestone is about the same as in granite. Rubidium, on the other hand, is generally absent from limestone, but its abundance in sandstone is lower and, in shale, about the same as in granite.

The natural radionuclides in sea water are derived from the atmosphere and from the land. Using the abundance data given by Lowder and Solon (1956), the approximate activities of certain radionuclides in sea water have been calculated and are tabulated in Table 2.

Because of differences in their geochemistry, radionuclides which are relatively abundant in soil may be relatively rare in sea water. In soil, for example, K-40 is relatively more abundant than Ra-226, but in sea water this relationship is reversed.

According to Love (1951) the average concentration of radium in fresh water is about 10×10^{-12} c/liter. This would be equivalent to 123×10^{-4} c/acre-foot which is about twice the concentration in sea water.

The average ratio of H^3/H^2 in rain water has been estimated as 1.0×10^{-16} and, in surface sea water, about 0.2×10^{-18} (Lowder and Solon 1956). Since the specific activity of H^3 is given as 9.6×10^3 c/g (Kinsman 1957), this would amount to approximately 6.0×10^{-3} curies of tritium per acre-foot of rain water and 1.2×10^{-3} curies of tritium per acre-foot of sea water.

3. Internal emitters. The radionuclides in air, soil, and water are readily incorporated in marine and terrestrial food-chains. Carbon-14 and H-3 are especially ubiquitous. The concentration of naturally produced C-14 in organism is about 1.0×10^{-12} c/gram of carbon while the concentration of H-3 is about 1 to 5×10^{-10} c/gram of water.

According to one estimate (Fry and Kuroda 1959) the above ground parts of green crop plants on an acre of cultivated soil may contain about 110×10^{-6} curies of natural radionuclides (Table 3). Considering only the radionuclides derived from the soil (Table 1), the plant/soil ratio on a unit area basis is approximately 23×10^{-4}. In other words, a single crop of plants could remove about 0.23% of the natural radioactivity in soil.*

Animals may acquire negligible amounts of radionuclides from the atmosphere. They regularly assimilate various fractions of the radionuclides

* This estimate does not include the C-14 derived from air or the H-3 derived from soil water.

ingested with food and water or other nonliving materials. According to Hursh (1955), the most important natural radionuclides assimilated by man are C-14 and K-40 (Table 4).

According to Libby (1955) and others, people who live between sea level and an altitude of 5000 feet above sea level in areas underlain by granite are normally exposed to about 200 mr/year of ionizing radiation from natural sources. Roughly 25% of this total is due to cosmic radiation, 50% to external emitters, and 25% to internal emitters.

b. "Man-made" sources. The major man-made sources of ionizing radiation in the biosphere are radionuclides produced by atomic fission or by neutron induction and released to the atmosphere as reactor effluents or as fallout from nuclear weapons tests.

Table 3. *Estimated activity of naturally occurring radionuclides in the above ground parts of an acre of green crop plants weighing 50 tons*[a].

Radionuclides	Activity in curies × 10^{-9}	Radionuclides	Activity in curies × 10^{-9}
K-40	75,600	Ra-226	400
Rb-87	19,900	U-238	157
C-14	18,500	H-3	156

[a] From: Fried & Heald (1959).

Table 4. *Estimated activity of naturally occurring radionuclides in a man weighing 70 kilograms*[a].

Radionuclides	Activity in curies × 10^{-12}	Radionuclides	Activity in curies × 10^{-12}
K-40	120,000	Ra-226	100 to 200
C-14	85,000	H-3	6.3 to 31.5

[a] After: Hursh (1955).

The direct effects of neutron and gamma radiation associated with the operation of a nuclear reactor are largely confined to the immediate vicinity of the reactor. Reactor effluents containing small quantities of fission products and radionuclides produced by neutron induction may be released to environmental media, but only local areas are affected. The disposal of highly radioactive reactor waste products is usually accomplished in a manner which prevents their release to the environment.

The direct effects of neutron and other "prompt" radiation released by the detonation of nuclear weapons are usually confined to areas in which the effects of blast and thermal radiation are more important. The radionuclides produced by nuclear detonations in the atmosphere are—by virtue of their quantity, world-wide distribution, and potential biological significance—the most important man-made sources of radioactivity in the biosphere.

The slow fission of uranium produces at least 170 isotopes of 35 lighter

elements (Blomeke 1955). Over half of these are radioactive, and most are relatively short-lived. With the exception of I-131 which has a half-life of 8.04 days and a few bone-seekers such as Ba-140 which has a half-life of 12 8 days, short-lived radionuclides are not generally considered to be important as potential biological hazards.

The fission products generally considered to constitute a long term biological hazard are Sr-90 and Cs-137. The biochemical behavior of Sr-90 is similar to that of calcium; and when assimilated by animals, it is deposited in the skeleton. The behavior of Cs-137 is similar to that of potassium and therefore, when assimilated by animals, it contributes to the radioactivity of muscle tissue. There are a variety of other fission products (Caster 1959) which accumulate in different animal organs, but their quantities are rather small in comparison to those of Sr-90 and Cs-137.

Carbon-14 is produced by both "atomic" and "hydrogen" bombs while

Table 5. *Estimated production of major radionuclides by nuclear weapons tests up to the end of 1961.*

Radio-nuclides	Half-life	Estimated Yield in curies per KT "Fission"	"Fusion"	Estimated Weapons Yield "Fission" 127.5 MT*	"Fusion" 162.5 MT*
Sr-90	28 y	143 c*	—	18×10^{6} c	—
Cs-137	33 y	253 c*	—	32×10^{6} c	—
C-14	5500 y	23.4 c**	149 c**	3×10^{6} c	24×10^{6} c
H-3	12.5 y	—	6720 c**	—	1092×10^{6} c

* Estimates based on data given by Dunning (1962).
** Estimates based on data cited by Leipunsky (1959).

H-3 is produced only by thermo-nuclear devices. The assimilation of these radioisotopes by plants and animals has already been mentioned.

Table 5 provides a rough estimate of the amounts of Sr-90, Cs-137, C-14, and H-3 produced by nuclear detonations prior to January 1962. These estimates do not necessarily indicate the amounts of radionuclides potentially available for assimilation by plants and animals. There is considerable uncertainty as to the amounts of these materials currently stored in the stratosphere, and the amounts eventually to be deposited as fallout will not be uniformly distributed. Also, as will be discussed in later sections, biological availability is dependent upon the interaction of a great many biotic and abiotic factors which are quite variable from place to place in the biosphere.

Assuming the area of the earth to be 2×10^{8} square miles, there has been enough Sr-90 and Cs-137 produced by nuclear detonations up to the end of 1961 to result in the world-wide deposition of 90 mc (millicuries) of Sr-90 and 160 mc of Cs-137 per square mile. It has been estimated (Dunning 1962) that the total deposition of these isotopes in the United States may eventually amount to 125 mc of Sr-90 and 230 mc of Cs-137 per

square mile. (Apparently, these estimates are based on the assumption that only two thirds of the Sr-90 and Cs-137 produced are available for world-wide distribution and that the United States, because of its latitudinal position, will receive about twice the world average.)

Rafter & Fergusson (1957) have reported that the C-14 content of wood formed in 1957 was 6.7% higher (in New Zealand) than that of wood formed in 1954. The atmosphere normally contains 2×10^{40} atoms of C-14 produced by nuclear reactions between cosmic rays and nitrogen molecules (Broecker and Walton 1959). Using the data cited by Dunning (1962) and Leipunsky (1959), I have estimated that all nuclear detonations in the atmosphere prior to January 1962 have produced approximately 27×10^{39} atoms of C-14. If 10% of this total were absorbed by sea water and the remainder were uniformly distributed in the atmosphere, this would amount to an increase of about 12% over background. The current (Jan. 1962) concentration of bomb-produced C-14 is probably greater in the northern hemisphere than in the southern, and most of the total in both hemispheres is probably confined to the stratosphere. The time required for thorough mixing is unknown, but it is probably short in comparison to the 5500-year half-life of C-14.

The estimated amount of H-3 produced by nuclear detonations is quite impressive but may be somewhat misleading. While H-3 may make a major contribution to the over-all increase of environmental radiation, its half-life is relatively short; and it is so diluted by rain, by surface and ground waters, and by the water in plant and animal tissues that its concentrations in organisms (Table 4) is generally quite small in comparison to the concentrations of other radionuclides.

While many of the preceding estimates are based on over-simplified assumptions, they should provide an indication of the contrast between natural and man-made sources of ionizing radiation in the biosphere and of the relative concentrations of the former in air, soil, water, plants, and animals. The sections which follow deal primarily with the initial distribution of radionuclides produced by nuclear detonations in the atmosphere and with the subsequent redistribution of these materials by environmental processes and by organisms.

Formation and dispersal of fallout. FALLOUT FORMATION. When a nuclear weapon is detonated, about 50% of the energy released is dissipated as blast and shock, 35% as thermal radiation and light, and 15% as nuclear radiation (Glasstone 1957). About one third (5%) of the nuclear radiation is dissipated during the first 60 seconds after the detonation. The remainder (10%) may be incorporated in or on the fallout particles which are formed during the development of the familiar "mushroom" cloud.

Immediately after the detonation (Fig. 1) an extremely hot, intensely luminous "fireball" is formed. Because of its high internal pressure and

temperature, the fireball expands and rises with great rapidity. The "primary" shock wave travels ahead of the expanding periphery of the fireball and produces a bowl-shaped cloud of dust when it strikes the ground. Some

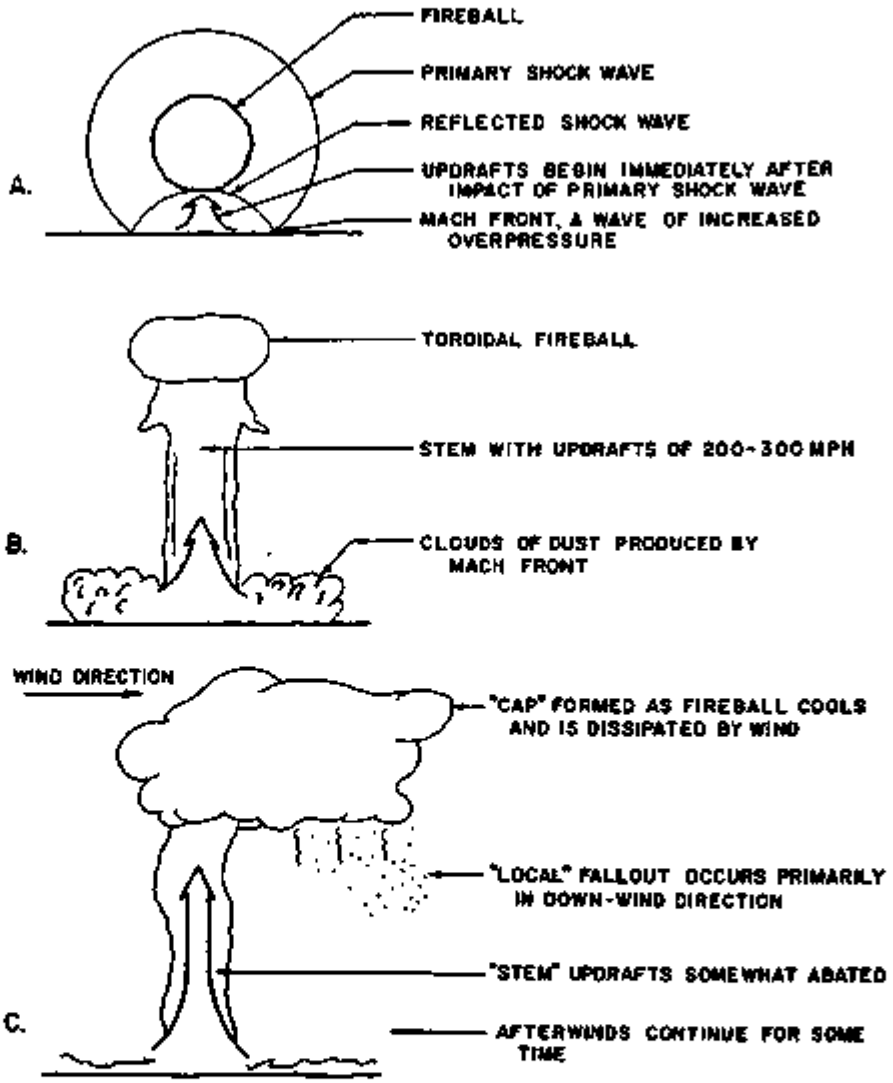


Fig. 1. Stages in the development of a "mushroom cloud" following the detonation of a nuclear weapon a few hundred feet above ground.

of this debris may be carried into the "stem" of the developing cloud by the high velocity updrafts which develop in the wake of the rising fireball.

While the fireball rises, it assumes a toroidal configuration which is similar to a rapidly-revolving, doughnut-shaped ring. Soil materials carried

into the stem by updrafts may be sucked into or circulated through and around the toroidal fireball.

At some distance from ground zero (the point on the ground directly below the detonation), the "primary" shock wave is augmented by a "reflected" shock wave. This combined front, the "Mach" front, travels away from ground zero at about the speed of sound. Moving like a wall of highly compressed air or a wave of increased pressure, the Mach front may produce dense, turbulent clouds of soil and other debris at some distance from ground zero.

After the passage of the Mach front, "afterwinds" of hurricane velocity converge on ground zero and may carry more materials into the stem beneath the rising fireball. The fireball continues to rise until its temperature matches that of the surrounding air. The height to which the fireball and "cap" of the mushroom cloud may rise is dependent primarily upon the amount of thermal energy released by the detonation and on the altitude above the surface at which the detonation occurs.

Initially, the fireball contains only the fission products and other materials vaporized by the explosion. If it intersects the ground, tons of soil may also be vaporized and incorporated directly into the fireball. Considerable amounts of the material carried into the stem by updrafts and afterwinds may also be incorporated into the fireball as vapor or as molten or solid particles. The condensation of particles occurs during the rise and cooling of the fireball. Ordinarily, the whole visible cloud is dispersed by wind in less than an hour.

The condensation of vaporized material in the fireball takes place in reverse order of boiling points (Triffit 1959). Since the amount of radioactive material in the fireball is quite small in comparison to the amount of other materials, most of it condenses in or on particles of metal or soil which are more abundant. The first particles available for this purpose are probably metallic. These may serve as nuclei for the condensation of materials having lower boiling points, or they may collide with and be absorbed by molten soil particles. In the cooler regions of the fireball, solid soil particles may provide surfaces for the condensation or absorption of radioactive materials (Miller 1960).

The precursors of some of the radionuclides in fallout are noble gases, and these do not become associated with solid particles until after they have decayed to form a more active kind of atom. This happens, for example, in the case of Sr-90 which is produced by the decay of Kr-90. As a result of this "isotopic fractionation" process, radionuclides having gaseous precursors are more likely to be associated with relatively small particles which are carried to the upper regions of the mushroom cloud. Particles formed in this manner are likely to be more prevalent in worldwide than in local fallout.

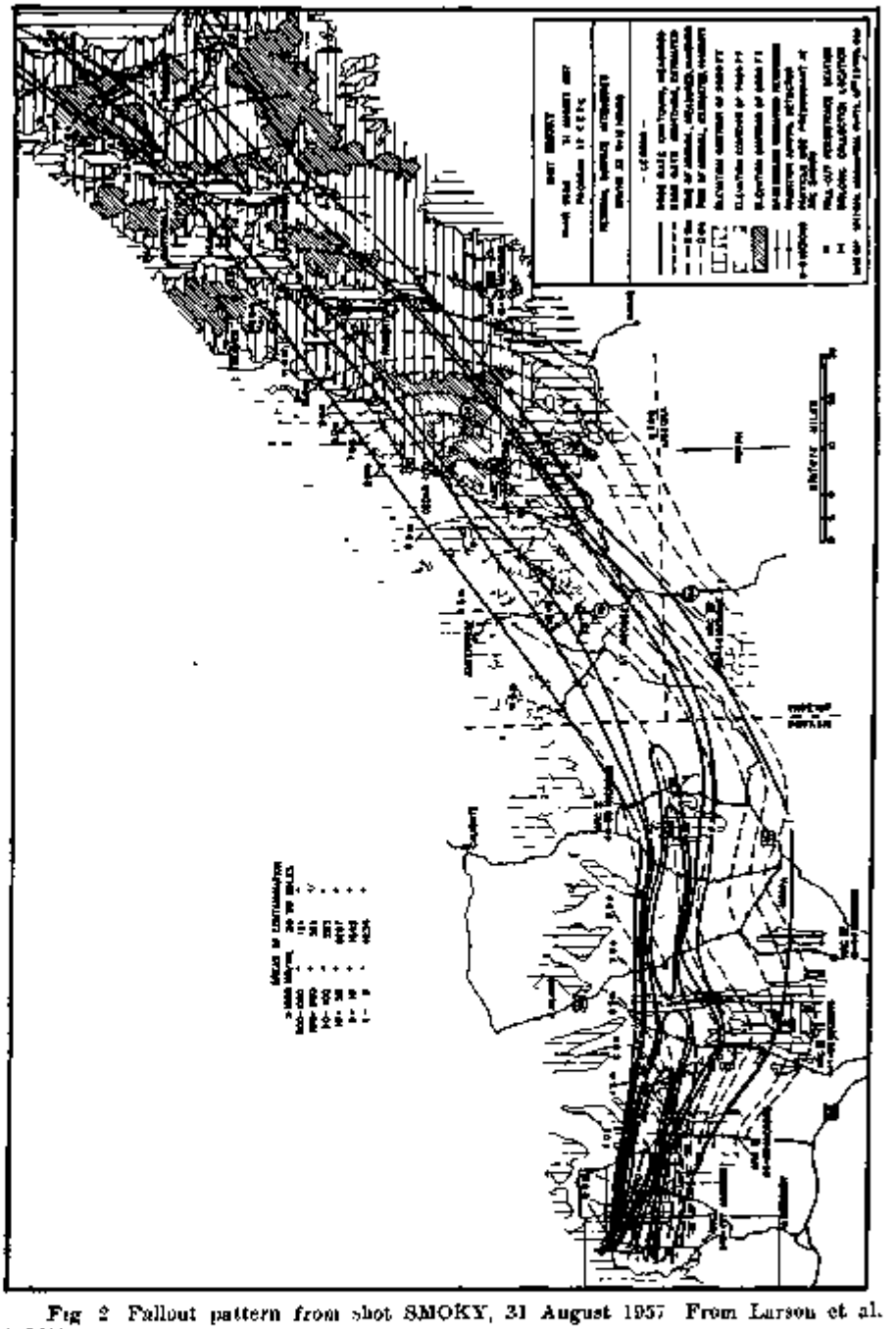


Fig. 2. Fallout pattern from shot SMOKY, 31 August 1957. From Larson et al. (1980).

arson et al.

Needless to say, there are many possible variations on the themes outlined above, and fallout formation is truly a complex process which involves the interaction of a great many variable factors. Factors such as kiloton or megaton yield, kind of support, height of detonation above the surface, and kind of surface are all important in determining the kinds and amounts of material incorporated in the fireball. The kinds and amounts of material which take part in the process are important in determining the physical and chemical properties of the particles formed. And finally, properties such as the size, shape, density, chemical composition, solubility, and radionuclide content of the particles formed are important in determining their initial distribution and subsequent ecological behavior in the biosphere. To determine which of these parameters are biologically significant is one of the major objectives of the study of fallout phenomenology.

FALLOUT PATTERNS. Fallout can be classified as "local," "tropospheric," or "stratospheric." Local fallout is composed primarily of larger particles which fall, usually in a matter of hours, within a few hundred miles of ground zero. The fine debris produced by most detonations in the kiloton range may remain suspended in the troposphere for a period of weeks or months before it is removed by some scavenging mechanism such as rain or snow. The fine debris produced by detonations in the megaton range is carried into the stratosphere where, in the absence of weather, it may remain for a period of months or years before reentering the troposphere. Both tropospheric and stratospheric fallout contribute to "world-wide" fallout.

1. Local fallout. Local fallout can be defined as the fallout occurring in areas corresponding to a fallout time of H + 12 hours (H + 12 hrs. = the 12 hrs. immediately after detonation). Most of the local fallout patterns produced by detonations at the Nevada Test Site (Fig. 2) have been roughly cigar-shaped. In more or less "typical" patterns, the isodose contour for 1.0 mr/hr (milliroentgens per hour), as determined by ground and aerial surveys, has extended downwind from ground zeros for distances of 200–300 miles. The lateral distances from the midline of fallout (the line of maximum radiation in the fallout path) to the 1.0 mr/hr contour have been on the order of 15–30 miles.

In most cases, the isodose contours have been approximately concentric. Occasional hot-spots have been found (Larson et al. 1960), but most of these were small in area. Isodose contours of more than 1000 mr/hr at H + 12 hours have been measured near ground zeros, but the areas so contaminated have been small, and these levels of contamination persist for only a few days or a few hours.

As indicated by isodose contours and by analyses of fallout collections (Baurmash et al. 1958, Larson et al. 196?, and Rainey et al. 1954), the unit area activity of local fallout decreases (Table 6) with increasing distance

from ground zeros and/or from midlines. Concurrently, the percentage of activity associated with particles $< 44\mu$ (microns) and with particles $< 88\mu$ in diameter show a marked increase.

For a given detonation, the percentage of theoretical fallout activity* associated with particles of $< 44\mu$ is inversely related to the amount of material incorporated in the fireball. Devices detonated as air drops at altitudes > 1500 feet have produced no detectable fallout within 200 miles of the Nevada Test Site. The local fallout from a balloon-supported device whose fireball did not intersect the ground, accounted for only 0.2% of the theoretical fallout activity produced by the detonation. For a similar balloon-supported device, whose fireball did intersect the ground, local fallout accounted for 2.12% of the theoretical total. Tower-supported detonations, with and without fireball-ground intersection, resulted in local fallout patterns containing 6.7% and 24.5% respectively of the total activity produced.

Of all the radioactive material produced by nuclear detonations at the Nevada Test Site, $< 25\%$ of the total (and $< 10\%$ of the radiostrontium)

Table 6. *Beta activity of fallout in relation to its particle size composition and distance from ground zero.*

Miles from G.Z.	12	20	46	60	79	96
Particle Size	(% of activity in particle size range)					
$< 44\mu$	3.3	12.7	16.0	18.8	22.9	24.9
44–88	1.9	52.7	49.8	77.2	66.5	72.4
88–125	1.8	24.3	27.0	1.3	7.4	1.9
> 125	94.0	10.3	7.2	2.7	3.2	0.8
Total Fallout	(Activity in $\mu c/ft^2$ at H + 12 hrs)					
on Soil	4817	2593	533	202	184	160

After: Baurmash et al. (1958).

has been deposited as local fallout (Larson et al. 1960). Presumably, the remainder has been available for long distance transport and deposition chiefly in areas to the east and northeast of Nevada.

2. Tropospheric fallout. The fine radioactive debris released to the troposphere in southern Nevada may be carried by high-altitude winds across the United States and around the globe in a girdle ranging from 30° to 60° north latitude. Some tropospheric debris has been observed to circumnavigate the globe in a period of from 4 to 7 weeks (Glasstone 1957). Some tropospheric debris may be deposited by gravity fall, and some may be deposited by impaction on vertical surfaces, but the removal of the major part is probably accomplished by rainfall and other forms of moisture precipitation (Greenfield 1957).

During the 1957 test series in Nevada, rainfall east of the Rocky Mountains contained considerable quantities of 1–12 day-old fission products. In

* Based on an estimated gamma activity at H + 1 hr of 3×10^6 curies per kiloton of yield.

many cases, single rainfalls deposited 1–3 curies of beta activity per square mile. On July 9 & 10, 1957, the rain falling on Jefferson City, Missouri contained 7.6 c/mi² of fresh fission product activity (U. S. Atomic Energy Commission 1959b). In the same year, increased levels of Sr-90 were observed in North Dakota milk (Pfeiffer 1958) and in Minnesota wheat (Cas-

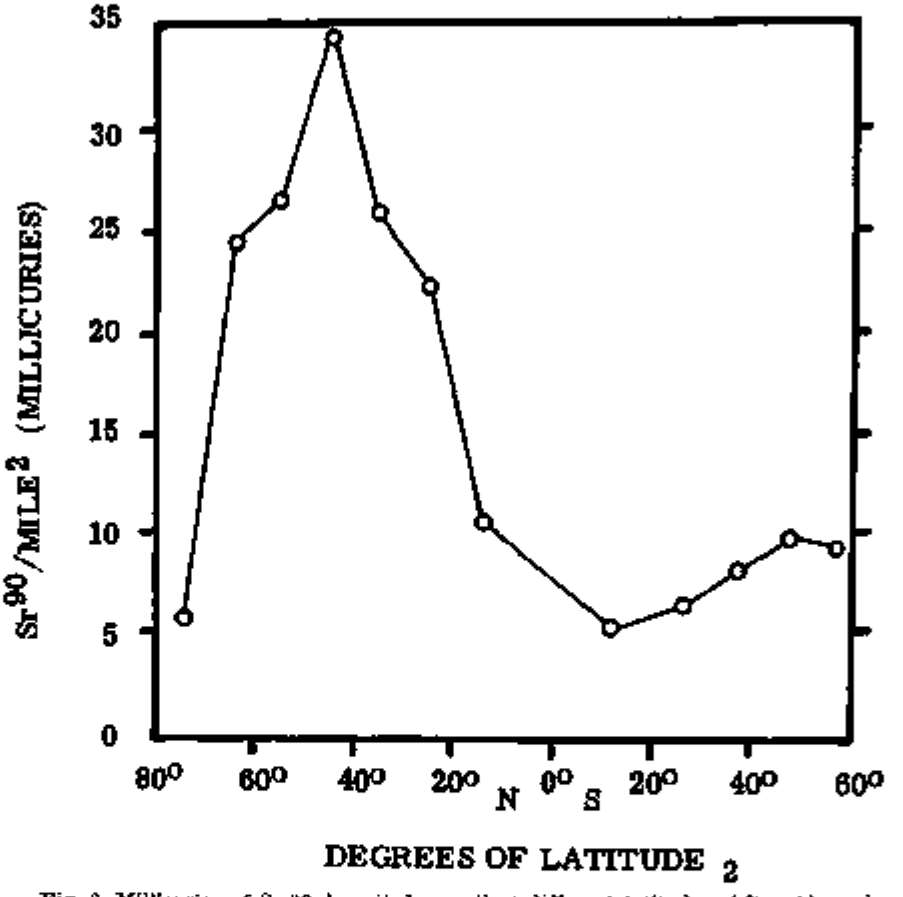


Fig. 3. Millicuries of Sr-90 deposited on soil at different latitudes. After Alexander (1960).

ter 1959), and small quantities of fallout were detected on vegetation in New England (Bormann et al. 1958). Since these areas are also in the band of maximum world-wide fallout, there is some doubt as to what fraction of these increases was due to tropospheric and what fraction to stratospheric fallout.

In general, these temporary, usually localized increases of environmental radiation and of the radionuclide content of foods have not been regarded as particularly hazardous (U. S. Atomic Energy Commission 1959a). They

are ecologically interesting because they illustrate the operation of environmental processes which may result in the concentration of radioactivity in a particular area of the biosphere. A more recent case of long distance, apparently tropospheric fallout may illustrate this point even better.

On September 10, 1961 two nuclear devices were detonated at Novya Zemlya in northern Russia. The air masses containing debris from these detonations traveled southward over Labrador and along the Atlantic Coast of the United States, then westward, and finally northward into the Mississippi drainage basin where it was deposited from cold, descending air. The radioiodine content of milk collected in St. Louis, Missouri on September

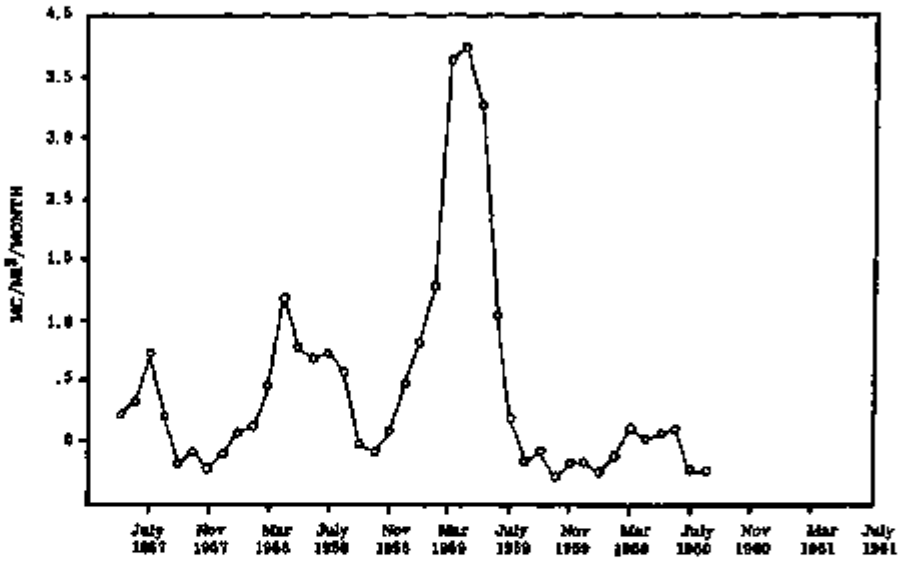


Fig. 4. Rate of Sr-90 deposition on the United States based on average monthly deposition at 10 HASL collection stations. After Kuroda (1960).

20 was 80 $\mu\mu c/l$ (micro-microcuries per liter); on September 27 it was 500 $\mu\mu c/l$; on October 6, 250 $\mu\mu c/l$; and October 31, 80 $\mu\mu c/l$ (Dunning 1962).

3. Stratospheric fallout. Except at extremely high latitudes, surface or near surface detonations of less than 200 kilotons do not produce sufficient thermal energy to carry significant quantities of radioactive debris into the stratosphere. Detonations of larger yields usually produce fireballs which penetrate the tropopause, and most of the fine debris produced by a nuclear detonation in the megaton range is readily carried into the stratosphere.

The latitudinal distribution of Sr-90 (Fig. 3) indicates that stratospheric debris may be deposited in either hemisphere, but the greatest deposition has been in the northern hemisphere between 30° and 60° latitude (Alexander et al. 1960). Also there is evidence (Fry and Kuroda 1959, Health and Safety Laboratory 1958, and Kuroda et al. 1960) that the rate of fallout

in the mid-latitudes of the northern hemisphere is higher in late winter and early spring than at any other time of year (Fig. 4).

Several hypotheses have been offered to explain these latitudinal and seasonal variations in the amounts and rate of stratospheric fallout but several important issues have not yet been resolved (Arnold and Martell 1959, Libby 1956a, 1956b, 1957, 1958, 1959, Machta 1959, Machta and List 1956a, 1956b, 1958, Martell and Drevinsky 1960, and Stewart et al. 1957).

Estimates of the residence time or effective half-life of radioactive debris in the stratosphere have ranged from "a matter of months" (Martell and Drevinsky 1960) to as much as 5 to 10 years (Libby 1956a). Apparently, the residence time and rate of stratospheric fallout are not uniform but tend to decrease with decreasing altitude and with increasing latitude of introduction. The rate of stratospheric fallout is highest for materials introduced in the lowest part of the stratosphere at high latitudes and lowest for materials introduced at higher altitudes near the equator.

In both the troposphere and the stratosphere the transport of materials is toward the northeast at mid-latitudes and toward the southwest at higher latitudes. In mid-latitudes, the transfer of the stratospheric materials into the troposphere probably occurs in the vicinity of the subtropical jet stream. At high latitudes, this transfer occurs in high pressure (anti-cyclone) areas where there is a pronounced subsidence of stratospheric air during late winter and early spring. In both hemispheres the rate at which stratospheric materials are removed from the troposphere is closely related to precipitation patterns. The inter-hemispheric exchange of atmospheric debris probably occurs in conjunction with the seasonal, latitudinal migration of global high and low pressure belts and their associated wind systems.

The actual processes involved in these exchanges between the stratosphere and troposphere and between the northern and southern hemispheres are imperfectly understood. Studies now in progress may help to resolve a variety of questions concerning the mechanisms of stratospheric and tropospheric fallout, but the solution of these problems will probably be a greater boon to meteorology than to radioecology.

The critical problems in radioecology are related to the measurable aspects of fallout rate and the distribution of particular radionuclides. The radionuclides generally considered to be most important in the evaluation of the potential biological hazards of world-wide fallout are C-14, Sr-90 and Cs-137. The C-14 produced by nuclear detonations and suspended in the atmosphere as $C^{14}O_2$ may eventually be distributed uniformly in the atmosphere of both hemispheres. Most of the Sr-90 and Cs-137 will probably be deposited in the northern hemisphere between 30° and 60° latitude.

A better understanding of the mechanisms which determine the rate and geographical distribution of world-wide fallout would be of particular value

in making long-term predictions of biospheric contamination. The problems of major concern in radioecology are those dealing with the fate and biological consequences of fallout materials after they are deposited in the biosphere.

Movement of fallout materials in terrestrial ecosystems*. The fallout in a given land area may be deposited directly on soil, plants, or animals (Figure 5). These materials may be redistributed by environmental processes and/or by organisms. They become parts of the ecosystems in which they are deposited and may be incorporated in a variety of biogeochemical cycles.

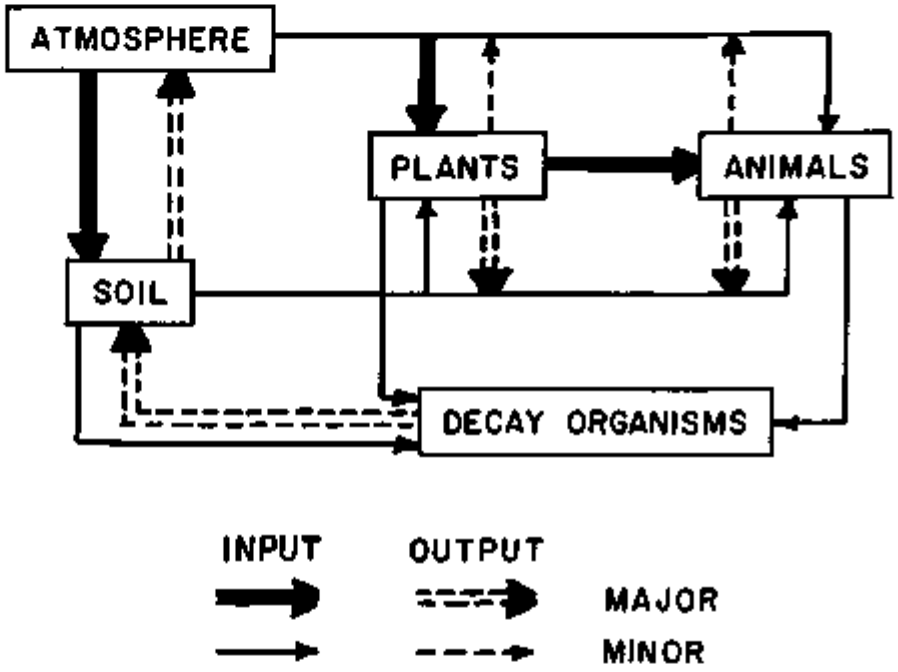


Fig. 5. Principal pathways of fallout materials transfer in a terrestrial ecosystem.

Most of the current research effort in radioecology (AIBS 1962) is devoted to studies of the occurrence, redistribution, and cycling of radionuclides in various kinds of ecosystems. In connection with fallout, the general objectives of these studies are: (a) to determine the properties and relative distribution of fallout particles as they are initially deposited on soils, plants, and animals, (b) to evaluate the biotic and abiotic factors which influence rates of radionuclide redistribution and cycling by different routes of transfer, and (c) to develop and test mathematical models for predicting, on the basis of data available immediately after contamination, the probable

* "Any area of nature that includes living organisms and nonliving substances interacting to produce an exchange of materials between the living and the nonliving parts is an ecological system or ecosystem." Odum (1959).

future concentrations of radionuclides in or on soils, in or on plants, and in various animal tissues and animal products. The general purpose of such studies is to provide data and to develop concepts which can be used in making realistic quantitative evaluations of the potential biological hazards of fallout under a wide variety of environmental circumstances.

The problem of evaluating biological hazards due to environmental contamination has been arbitrarily but conveniently divided into two parts (Dunning and Hilkin 1956, Lindberg and Larson 1956). The "acute" or immediate and short-term hazards arising from environmental contamination by fallout are associated primarily with external emitters and secondarily with internal emitters. The "chronic" or long-term hazards are associated primarily with internal and secondarily with external emitters.

The exposure of plants and animals to ionizing radiation from external emitters in a fallout contaminated environment may be relatively high at the time of fallout, but it usually decreases in proportion to the radioactive decay of fission products. The early decay rate of mixed fission products in fallout debris (*ca.* $t^{-1.2}$) approximates a 10-fold decrease in activity for every 7 days after contamination. If the level of environmental contamination at D + 7 days is 100%, it will be approximately 10% at D – 14 days, 1.0% at D + 21 days, and 0.1% at D + 28 days.

This rapid, early decay rate of fresh fallout is due to the predominance of short-lived fission products. Hence, the acute or short-term biological hazards of fallout are associated with: (a) the exposure of plants and animals to relatively high levels of ionizing radiation from external emitters most of which have short radioactive half-lives, and (b) the assimilation of these short-lived isotopes by plants and animals.

The chronic or long-term biological hazards of fallout are associated with: (a) the continuing exposure of plants and animals to low levels of ionizing radiation from long-lived fission products which are trapped in the environment, (b) the persistence of medium and long-lived radionuclides in plants and animals and especially in or on soils, and (c) the continuing availability of radionuclides in or on the soil for the recontamination of plants and for biogeochemical cycling through consumer and decomposer food chains.

INITIAL DISTRIBUTION OF FALLOUT PARTICLES. The most important factors which influence the formation, dispersal, and geographical distribution of fallout particles were described in preceding sections. In studies of the occurrence, redistribution, and cycling of fallout materials, it is often necessary to begin by determining the physical and chemical properties of fallout particles and their distribution to soil, plants, and animals in given areas.

1. Soil. The total amount and particle size composition of fallout deposited in different parts of a local fallout field can be estimated by assays of fallout collected on trays of polyethylene pellets (Romney et al. 1959) placed, in open areas, on the soil surface. Unit area soil samples can also be

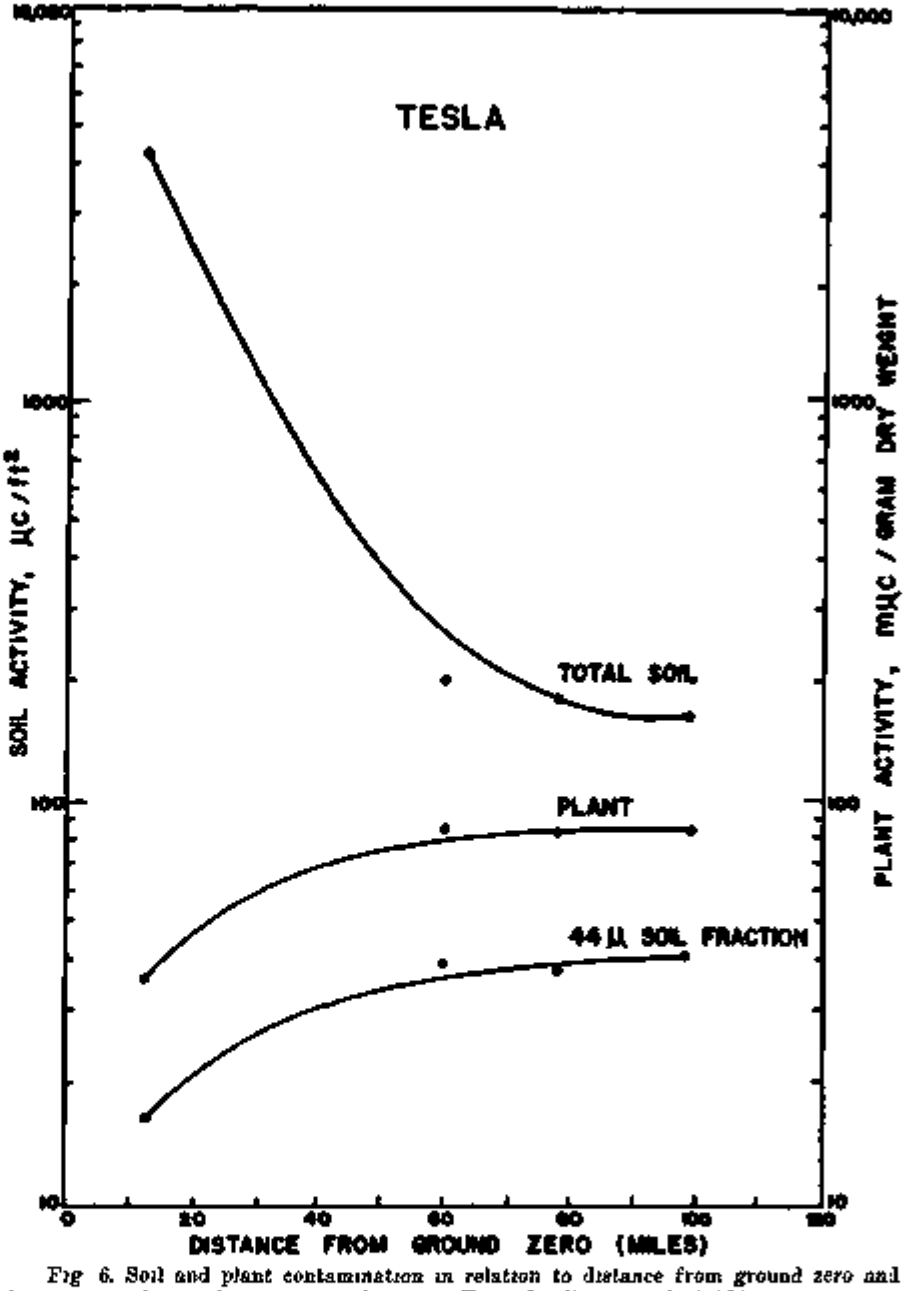
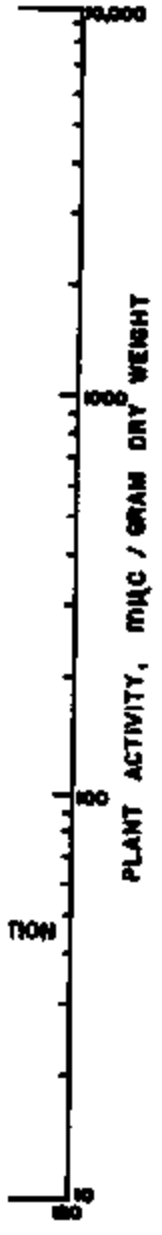


Fig 6. Soil and plant contamination in relation to distance from ground zero and the activity of particles < 44μ in diameter From Lindberg et al (1959)

used for this purpose, but the analysis of data may be confounded by the presence of natural radionuclides and of fission products previously deposited on the soil



In most local fallout patterns, the unit area activity and the mean particle size of fallout deposited on fallout collectors and/or on soil surfaces have been found to decrease with increasing distance from ground zero and with increasing time of fallout deposition. Concurrently, there is an increase in the percentage of total activity associated with particles $< 44\mu$ in diameter (Table 6). In general, the radioactive materials associated with smaller fallout particles are more readily soluble in distilled water and in dilute acid than are those associated with larger fallout particles. As much as 90% of the activity associated with fallout particles $< 44\mu$ in diameter may be soluble on 0.1 N HCL. The local fallout from balloon-supported detonation

Table 7. Soil and plant contamination in relation to distance from ground zero and the particle size composition of fallout.

Detonation	Apple II, 1953			Smoky, 1957		
Miles from Ground Zero	7	48	106	132	205	259
Miles from Midline	0.6	0.8	24	0.5	0	3.5
Particle Size:			(% of beta activity in size range)			
$< 44\mu$	0.1	15.3	71.7	21.1	41.8	NS
$44–88\mu$	0.1	2.2	4.0	71.1	55.1	NS
$> 88\mu$	99.8	82.5	24.3	7.7	3.0	NS
Fallout on			(μ c beta activity per square foot)			
Soil	2070	636	20	75	133	85
Fallout on Plants:			(μ c beta activity on plants per ft² of soil)			
Red Clover*	2.35	4.22	0.25	NS	NS	NS
Wheat*	4.14	15.09	1.16	NS	NS	NS
Alfalfa**	NS	NS	NS	3.7	NS	9.9
Mixed grasses**	NS	NS	NS	NS	5.0	4.5
% Interception by Plants:			(soil activity/plant activity × 100)			
Red Clover	0.11	0.66	1.25	NS	NS	NS
Wheat	0.20	2.37	5.80	NS	NS	NS
Alfalfa	NS	NS	NS	4.93	NS	11.74
Mixed grasses	NS	NS	NS	NS	3.75	5.32

* Grown on soil flats and relocated on D-1 day.
** Collected from farms in fallout area.
NS = No sample.
After: Romney et al. (1963).

has been found (Larson et al. 1960) to be composed primarily of $< 44\mu$ particles, and this size fraction was found to contain relatively higher concentrations of Sr-89, Sr-90, Y-91, Ru-106, and Ba-140 but less Zr-95 and Ce-144 than larger size fractions.

2. Plants. Studies of soil, plant, and animal contamination were made during Operation Teapot in 1955 (Lindberg et al. 1959) and during Operation Plumbbob in 1957 (Larson et al. 196?). The results of the plant contamination studies have been summarized recently by Romney *et al.* (1962). Their summary is briefly reviewed below.

It has been established by autoradiographic and microscopic examinations of foliage exposed to fallout that leaves in general are selective collectors of fallout particles $< 100\mu$ in diameter. Tallies of the size and number of

radioactive particles per unit area of leaf surface have indicated that most of the particles retained by foliage are $< 44\mu$ in diameter and have a mean diameter of about 20 μ. Leaves having sticky or hairy surfaces retain more particles per unit area than do leaves having smooth or waxy surfaces.

As indicated by Figure 6 and Table 7, the best correlations between fallout and plant activity have been obtained by comparing the activity of fallout $< 44\mu$ with the gross activity of plants. Both the total and the percentage of fallout intercepted by vegetation tend to vary in relation to the mechanical distribution of fallout particles $< 44\mu$ in diameter.

The percentage of fallout intercepted by a unit area of vegetation, e.g., by the clover or wheat growing on a square foot of soil surface tends to increase with increasing distance from ground zero and increasing time of deposition. In a given fallout pattern, the maximum contamination of vegatation usually occurs in areas where the total fallout activity per unit area is moderately high, and a relatively large percentage of the total activity is associated with particles $< 44\mu$ or $< 88\mu$ in diameter. Depending on the conditions of detonation and the particle size composition of fallout, maximum plant contamination may occur in areas close to ground zero or at distances of from 50 to several hundred miles from ground zero.

3. *Animals.* The external contamination on the skin and pelts of small mammals collected from 24 to 48 hours after fallout has been observed to account for 22% to 62% of the total beta activity associated with those animals (Lindberg et al. 1959). Experimental studies (Taplin et al. 1957) have indicated that some animal contamination may result from the inhalation of radioactive particles and aerosols. Field studies at the Nevada Test Site (Lindberg et al. 1954, 1959) have indicated that the average activity of lung tissues following fallout is rarely more than two or three times normal.

4. *Acute vs. chronic exposure.* For both the plants and the animals living in a fallout-contaminated environment, maximum exposure to external emitters occurs at or about the time of fallout deposition and then diminishes in accordance with the decay rate of mixed fission products. In a general way, the acute phase of plant contamination coincides with the period that fallout materials are retained on foliage and/or assimilated by foliar absorption. In desert ecosystems, this period is probably less than a year and may be said to end with the loss of contaminated foliage. In the case of animals, the acute phase of exposure to increased ionizing radiation may be said to last as long as the exposure to external emitters and/or the total body content of mixed fission products is more than twice the pre-contamination levels of exposure.

Admittedly, these criteria are arbitrary, but they should be useful in distinguishing between: (a) the short-term biological hazards associated with relatively high levels of exposure to external emitters or to internal

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emitters which have short half-lives and (b) the long-term hazards associated

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SHORT-TERM ASPECTS OF REDISTRIBUTION AND CYCLING. *1. Soil.* Fallout particles deposited on the surface of bare sandy soil or desert pavement may be redistributed by wind or water. In some situations (Bellamy et al. 1949) erosional processes may result in the concentration of radioactive materials on surfaces of deposition. Detailed studies made during and following Operation Plumbbob (Larson et al. 196?) provided no evidence to support a general hypothesis of fallout dilution by erosion or concentration by deposition. The results of these studies indicate that the surface materials in small study areas (100 ft. × 100 ft.) were shifted about in a random fashion so that the variability of surface contamination was increased, but mean values showed no significant change. For periods of 1 to 5 years, the average fission product activity of bare soil surfaces remained higher than that of surfaces such as dunelets under shrubs and slight depressions in the desert pavement on which eroded materials were deposited.

Of the airborne materials which were redistributed during the first three weeks after fallout 9.7% to 21% of the total was composed of particles from 44μ to 88μ in diameter, and 68.3% to 85.4% of the total was composed of particles $< 44\mu$ in diameter. From D + 2 to D + 16 days there was a steady decrease in the amount of redistributed fallout. A severe rain storm on D + 17 resulted in the redistribution of particles $< 100\mu$ "in quantities almost as high as the original levels of contamination" (Larson et al. 196?).

Since the levels of soil contamination 6 months after fallout were consistent with predictions based on the decay rate of mixed fission products,

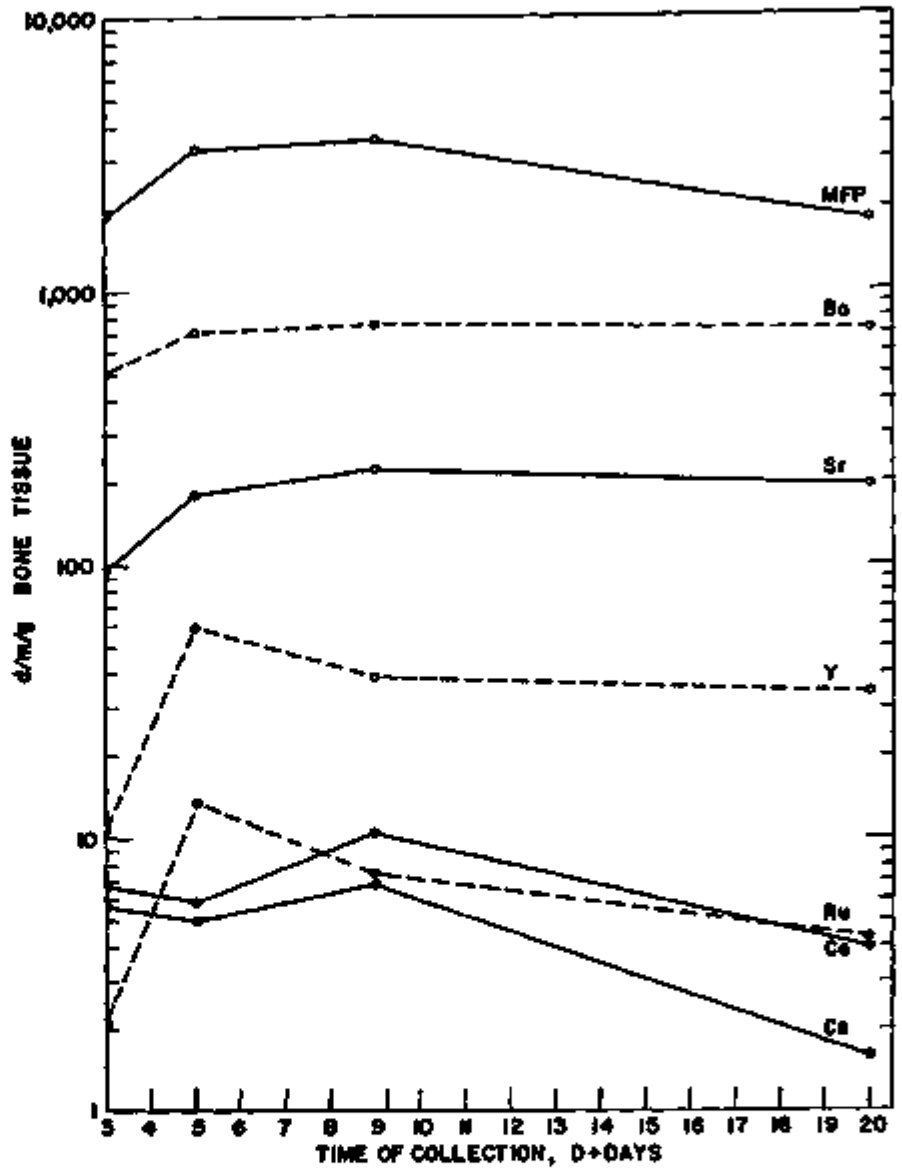
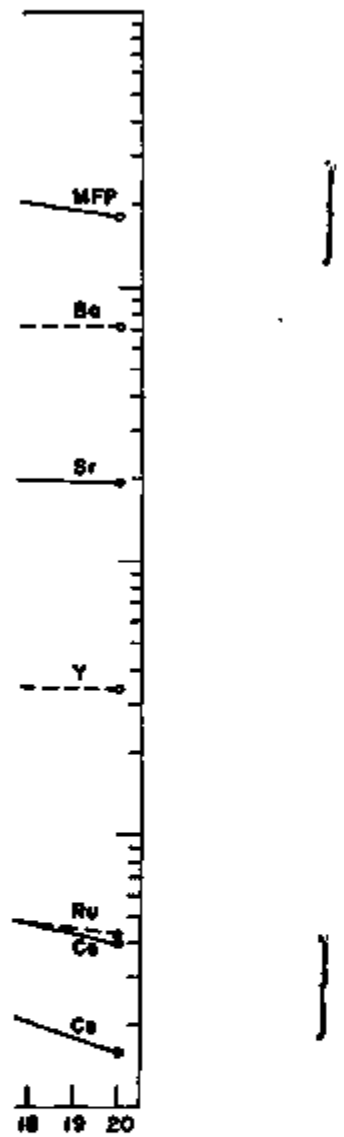


Fig. 7. Mixed fission product (MFP) and radioisotope activity of jack rabbits collected from an area contaminated by fallout from the Nevada Test Site. From Larson et al. (1960).

eral applicability of such observations has not been determined. Field studies based on the serial measurement of radioactivity on fallout contaminated plants (Larson et al. 1962), have indicated that the decline of activity



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on plants during a period of three weeks following contamination was parallel to the beta decay of $< 44\mu$ fallout particles.*

Under favorable conditions, some of the water soluble material deposited on leaves may be assimilated by foliar absorption. Russell et al. (1955, 1959) and Russell and Possingham (1959) reporting on fallout studies made in Australia and in the United Kingdom, have indicated that considerable quantities of water soluble fission products may be retained by foliar absorption. Since there have been no direct observations of foliar absorption of fallout originating from detonations at the Nevada Test Site (Romney et al. 1962) we may suppose that foliar absorption is probably more significant in humid grassland or pasture regions than in arid grassland or desert-shrub regions.

3. Animals. The possible modes of fission product ingestion by herbivores are: (a) ingestion of fallout contaminated plants, (b) ingestion of fallout

Table 8. *Average beta activity in tissues of jack rabbits serially collected from an area contaminated by fallout from the Nevada Test Site* (80 miles from Ground Zero).*

Time of Collection, Days after Contamination	Average beta activity extrapolated to time of sampling and expressed as $\mu\mu c/g$ fresh tissue					
	Lung	Caecum	Liver	Kidney	Muscle	Bone
2	116	16,500	817	67	385	639
3	61	4,350	990	55	376	103
10	21	606	21	18	10	60
220	1	6	3	2	3	3
Background**	1	7	1	1	1	2

* After: Lindberg et al. (1959).
** Based on animals collected before contamination.

from the pelt during preening, (c) accidental ingestion of contaminated soil while digging or feeding, and (d) swallowing of material cleared from the lungs and nasopharynx. During the first few weeks after fallout, the correlation between the activity of plants and that of the gut contents of herbivores is usually quite close, and the other three possible modes of ingestion are probably of minor significance.

The maximum concentration of mixed fission products in the tissues of herbivores living in fallout contaminated environments occurs usually within a few days after fallout. The total body burden of internal beta emitters then decreases (Figure 7) in proportion to the decreased activity of forage plants, and this may approximate the rate of mixed fission product decay.

* Studies completed after this paper was written (Martin 1963, 1964) have shown that the effective half-lives of radionuclides on fallout-contaminated plants are significantly shorter than their respective radioactive half-lives, and losses in excess of radioactive decay have been attributed primarily to wind action.

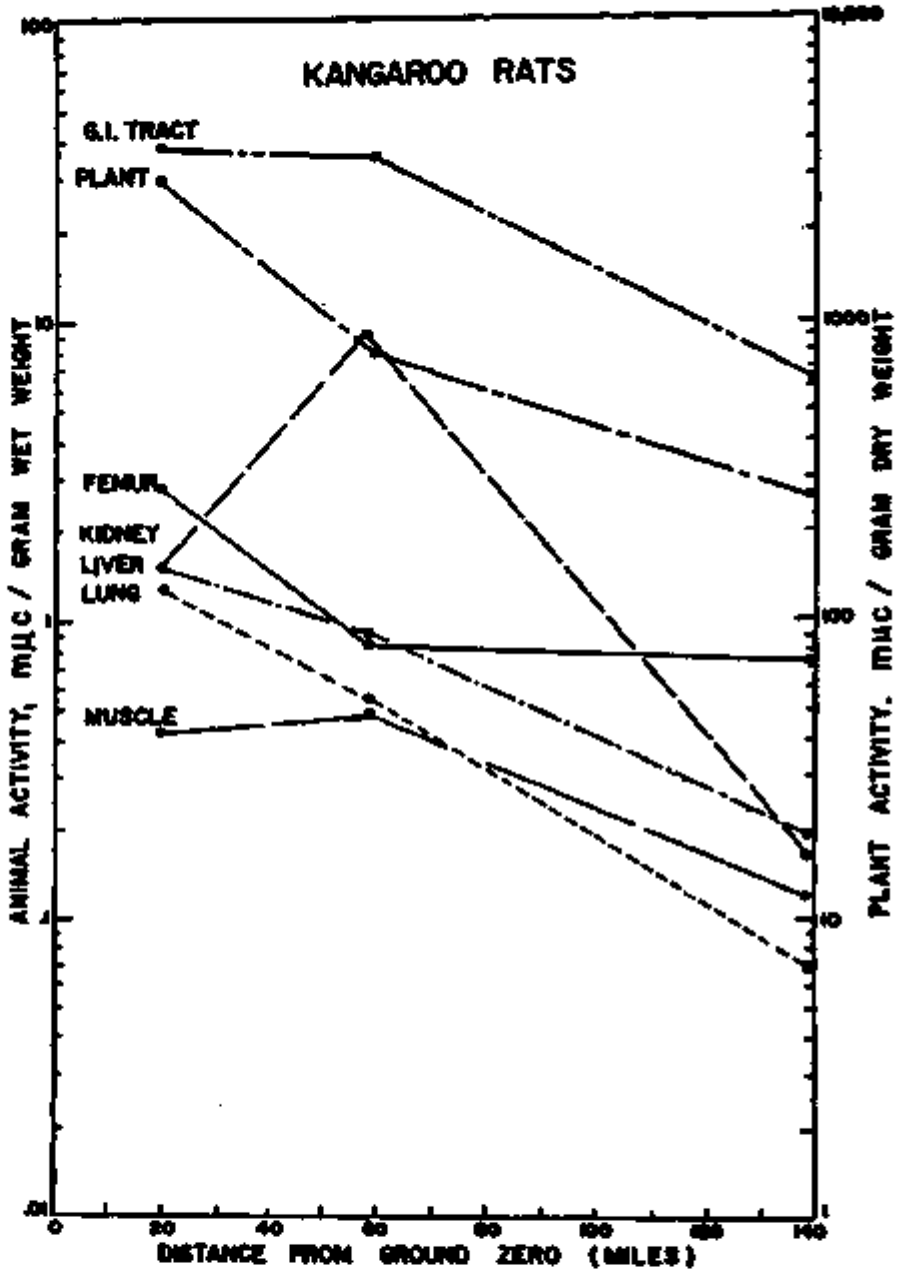

Fig. 8. Fission product activity of kangaroo rat tissues collected two days after fallout in areas located 20, 60, and 120 miles from ground zero. From Lindberg et al. (1959).

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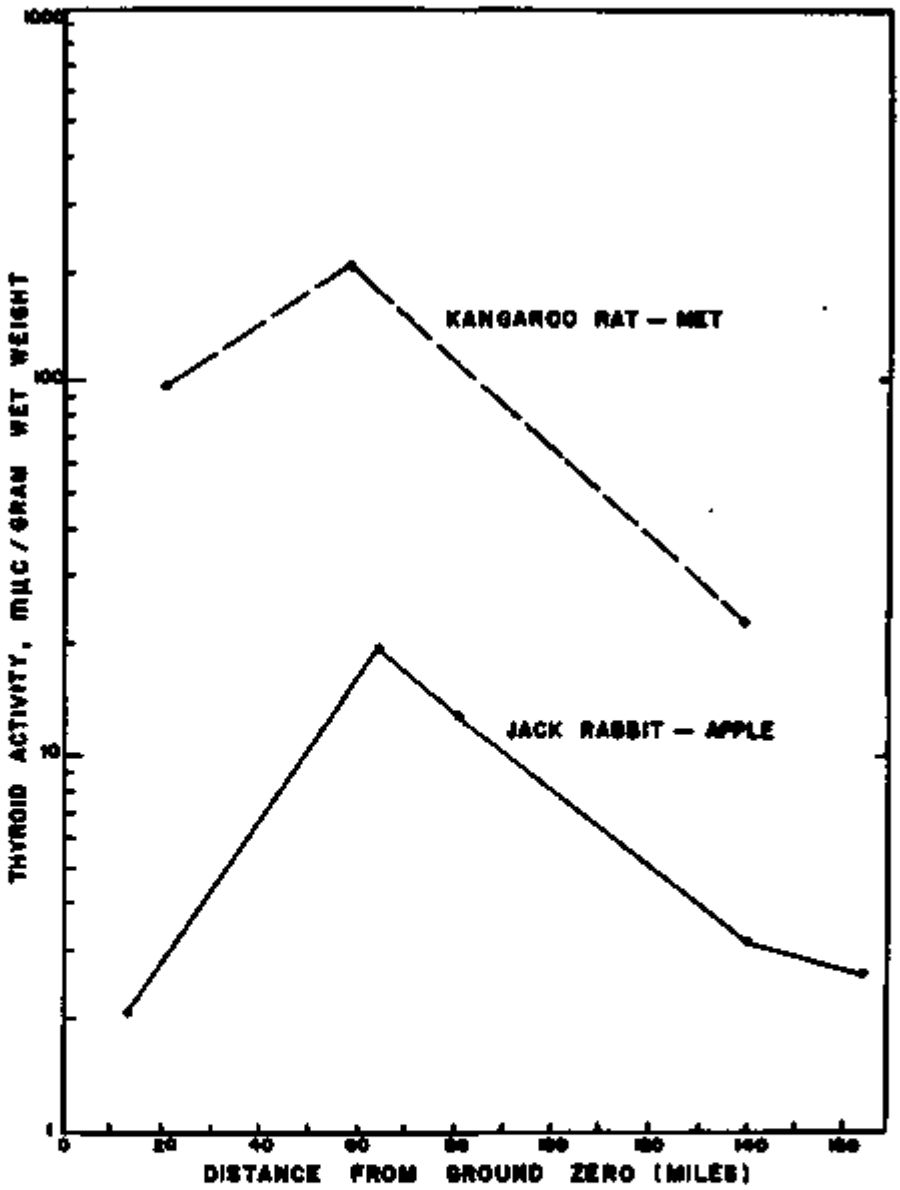

Fig. 9. Concentration of radioiodine in the thyroids of kangaroo rats and jack rabbits living in fallout contaminated areas 12 to 160 miles from ground zero. From Lindberg et al. (1959).

Table 8 illustrates the persistence of mixed fission product activities in the tissues of jack rabbits living in a fallout contaminated environment.

In this and in other contaminated areas which were serially sampled, the average tissue burdens 7 months after fallout were slightly higher than pre-contamination background levels. During the same period of time, the concentration of radiostrontium in the bones of animals living in contaminated environments may have increased from two to five times the background concentration.

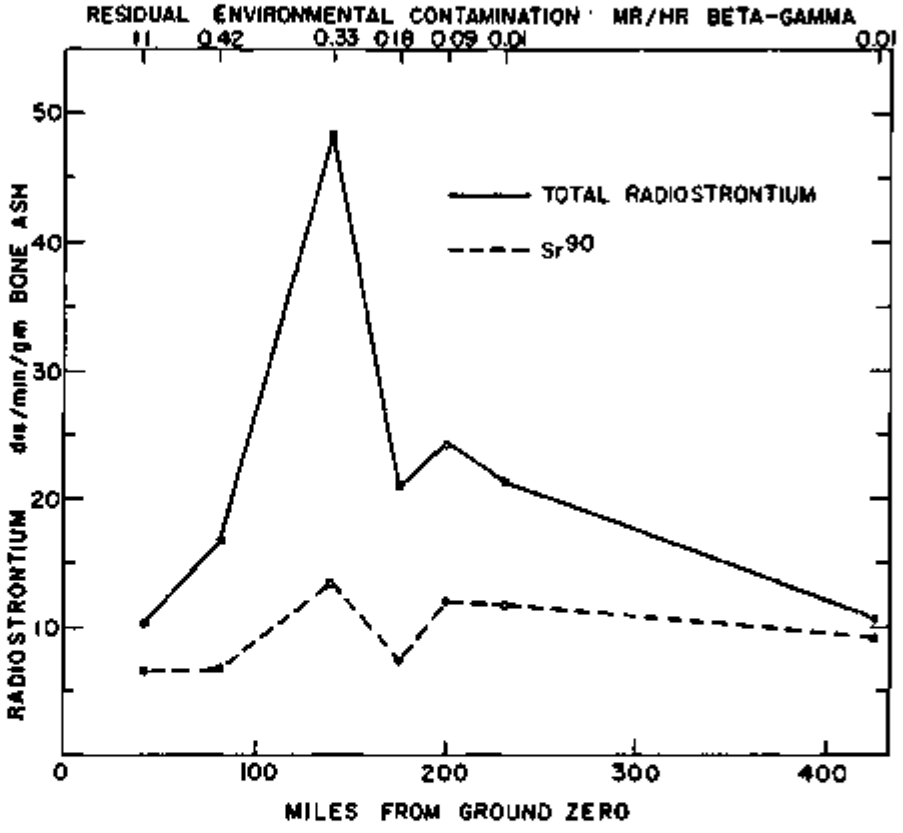

Fig. 10. Radiostrontium in bones of jack rabbits collected 6 months after fallout in midline areas 20 to 425 miles from ground zero. From Lindberg et al. (1959).

In general, the radioactivity of animal tissues collected a few days after fallout (Figure 8) decreases with increasing distance from ground zero. In two cases (Figure 9) the thyroid activity of kangaroo rats and jack rabbits was found to be higher at a distance of 60 miles from ground zero than at distances of either 12 or 160 miles. Six months after fallout (Figure 10), the maximum concentration of radiostrontium in the bones of jack rabbits was found to occur at a distance of 130 miles from ground zero. The concentration of Sr-90 in the bones of animals collected at a distance of more

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than 400 miles from ground zero was not significantly lower than that of animals collected at a distance of 200 miles, but it was somewhat higher than that of animals collected at a distance of only 50 miles from ground zero.

4. Discussion. These results provide evidence that the amount of fission product activity deposited on plants in different parts of a local fallout field is independent of total fallout deposited on soil but is closely related to the distribution of particles $< 44\mu$ in diameter. The assimilation of mixed fission products by herbivorous mammals living in fallout contaminated environments is closely related to the ingestion of fallout contaminated plants. Consequently, the intake of radioactive debris by herbivores is also independent (at least during the first few months after fallout) of total fallout activity and soil contamination; but it is closely related to plant contamination, and therefore dependent on the distribution of $< 44\mu$ fallout particles.

The distribution of a specific isotope is apparently dependent on the physical and chemical properties of its precursors and their behavior during particle formation and dispersal. The distribution patterns of radioiodine and radiostrontium, as indicated by their accumulation in the thyroids and skeletons of herbivores, appear to be similar and to correspond approximately to the distribution of the relatively small particles retained by foliage. The differences between radioiodine and radiostrontium distribution patterns may be related to differences in the half-lives of their precursors and in the height at which they become associated with particles in the rising fireball or stem of the mushroom cloud.

Long-term aspects of redistribution and cycling. *1. Soil.* Following the loss and replacement of contaminated foliage and the decay of short-lived fission products to an insignificant level of activity, most of the medium- and long-lived fission products in a fallout contaminated ecosystem will be found on or in the soil. The soil acts as a fission product reservoir and provides a continuing, usually low level, source of ionizing radiation to which plants and animals may be exposed either externally or internally or both. As mentioned earlier, the redistribution of external emitters by environmental processes tends to diminish with time.

In undisturbed soils, the downward movement of fallout materials deposited on the surface may be accomplished by the leaching and percolating action of rain water. This is a relatively slow process, and most of the activity deposited on undisturbed desert soils may remain, for a matter of years, in the upper one or two inches of the soil profile. Most of the radioactivity of soils contaminated by fallout from the first atomic bomb explosion was found, after 9 years, to be confined to the upper 2 inches. In leaching experiments using water equivalent to 84 inches of rainfall only a fraction of the activity from these materials was displaced downward, and the maximum displacement was only 0.5 of an inch (Olafson et al. 1957). Similarly,

most of the Sr-90 deposited by tropospheric and stratospheric fallout tends to remain in the upper 2 inches of undisturbed forest soils (Alexander et al. 1960).

2. *Plants.* Most of the fission product in the upper few inches of fallout contaminated soil is probably not available to deep-rooted plants, but the preliminary results of experiments now in progress (Romney et al. 1962) seem to indicate that such materials may be readily available to plants such as wheat which have "feeder" roots near the surface. This possibility is especially interesting because many desert plants and a variety of cultivated plants have root systems of this sort.

The availability of radionuclides in fallout contaminated soils for uptake by the root systems of plants is dependent upon an impressive array of biological and environmental factors (Hope 1959, Hansen et al. 1959, Menzel 1959, 1960, Nishita et al. 1960). Aside from the physical and chemical properties of the fallout and its distribution in and on the soil, most of these factors are directly or indirectly related to the composition of the soil solution (Hansen et al. 1959), and/or to various aspects of plant metabolism (Hope 1959). While it is not within the scope of this paper to discuss all of these factors, it is well worth noting that the best criterion of availability is actual uptake by plants.

Experimental studies have shown that the amount of radioactive material absorbed by the root systems of crop plants may vary considerably in relation to: the kinds and concentrations of radionuclides in the soil, the physical and chemical properties of different soil types, and the age and species of the plants considered (Nishita et al. 1960). After a series of studies concerning the uptake of fission products by various crop plants from a variety of soil types contaminated by fallout materials, Nishita et al. (1960) reported that, "The total uptake of fission products expressed as a percent of total residual activity in the soil at harvest time ranged from 0 to 0.06%."

When solutions of radionuclides are added to soil, a larger percentage of the total activity may be removed by plants. Menzel (1959) has reported that from 0.5% to 5% of the Sr-90 added to soil as a solution may be removed by a single crop of plants. Comparable figures for Ba-140 are 0.025% to 0.25%; but generally less than 0.1% of the Cs-137, Ru-106, and Ce-144 added to soil as a solution can be removed by a single crop of plants. Subsequent crops remove smaller fractions of the residual activity.

In spite of variations related to soil type, species, and stage of development, the relative order in which major fission products are absorbed by root systems from fallout contaminated soils is consistently as follows: Sr-89 Sr-90 >> I-131 > Ba-140 > Cs-137 > Ru-106 > Ce-144 (Nishita et al. 1960). The respective half-lives of these fission products are: 53 days, 28 years, 8.04 days, 12.8 days, 33 years, 1 year, and 282 days.

The distribution of mixed fission product activity in the above ground parts of plants grown on fallout contaminated soils is also relatively consistent. The concentration of activity is generally greater in leaves than in stems and much greater in stems in either fruits or seeds (Nishita et al. 1960).

The uptake of fission products from cultivated soils can be modified by the addition of fertilizers, organic matter, etc.; but according to Menzel (1959), "The effects of amendments on availability of Sr-90 in the soil are rather small. Additions of lime, gypsum, fertilizers, or organic matter in practical doses usually reduces uptake by less than half. . . . The optimum use of soil amendments for maximum crop production may often coincide with their optimum use for the reduction of Sr-90 uptake. Amendments may have larger effects on the availability of other fission products such as Ru-106, Cs-137, and Ce-144, but these fission products are normally less of a hazard than Sr-90."

Table 9. *Sr-90 in the bones of jack rabbits living in an area contaminated* by fallout (ca. 100 mi. from Ground Zero).*

Date of Collections	Sr Units** in Bone	Date of Collection	Sr Units in Bone
Sept. 3, 1957	20.7***	July 1958	19.4
Sept. 5, 1957	22.7	Aug. 1959	20.0
Sept. 9, 1957	26.8	May 1960	19.3
Sept. 13, 1957	25.0	May 1961	< 10.0

* Fallout occurred on Aug. 31, 1957.
** = 2.2 d/m Sr90/g Ca.
*** Precontamination level was ca. 20 Sr units.
After: Neel & Larson (1961).

3. Animals. Studies made during the Upshot/Knothole, Teapot, and Plumbbob Test Series (Lindberg et al. 1954, 1959, Larson et al. 196?) have provided a considerable amount of data concerning the biological availability and accumulation of Sr-90 by small mammals living in areas contaminated by fallout from the Nevada Test Site. The significant results of these studies have been summarized by Neel and Larson (1961) and by Larson et al. (1960, 1962).

Table 9 provides data which illustrate the accumulation and persistence of Sr-90 in the bones of jack rabbits (*Lepus californicus*) living in a fallout contaminated environment. The bones of animals collected in this area prior to its contamination by fallout on August 31, 1957 contained Sr-90 equivalent to an average of 20.6 Sr Units (Sr Unit = 2.2 d/m Sr-90/g Ca), and the Sr-90 level in soil was about 65 mc/mi^2.

The maximum level of Sr-90 in the bones of jack rabbits (26.8 Sr Units) occurred about 10 days after fallout, and a comparable level (19.3 Sr Units) was maintained for about 3 years. Soil samples collected in 1958 and 1959

had essentially the same average Sr-90 content (*ca.* 100 mc/ml²) as samples collected in 1957 immediately after contamination.

Of the 43 animals collected one year after fallout (1958), the bone ash of 41 contained Sr-90 equivalent to > 10 Sr Units. Of the 53 animals collected 3.7 years after fallout (1961), the bone ash of 39 contained Sr-90 equivalent to < 10 Sr Units, and only 14 contained Sr-90 equivalent to > 10 Sr Units. All the 1961 animals found to have higher levels of Sr-90 in their bones were judged, on the basis of weight, to be members of the older age groups. Since the average life-span of jack rabbits is about 3.2 years, the older animals collected in 1961 could have been present during the first year after fallout. Most of the animals collected in 1958 probably were present at the time of fallout.

Table 10. *Sr-90 in jack rabbits and soils of areas contaminated by fallout from the Nevada Test Site*.*

Miles from NTS	Sr Units in Jack Rabbit Bone	Sr 90 mc/mi² in Soils		
		Total**	Acid Soluble***	Percent Soluble
< 1	50.4	9014	980	10.9
2	23.2	933	58	6.2
80	21.5	142	18	13.0
140	13.8	41	27	66.7
205	12.9	32	16	50.3
350	20.9	67	48	71.8

* Samples collected in 1958.
** Based on sodium carbonate fusion method of analysis
*** Based on extraction with 6 N HCl
After: Larson et al. (1962).

Table 10 illustrates the relationship between Sr-90 in the soils of areas contaminated by fallout from the Nevada Test Site and the concentration of Sr-90 in the bones of jack rabbits living in those areas. As indicated by these data, the highest levels of Sr-90 in rabbit populations are associated with higher concentrations of Sr-90 in soils, but the relationship is not linear. Large increases in the total or acid soluble Sr-90 content of soils may be accompanied by relatively small increases in the Sr-90 content of jack rabbit bone ash.

4. Discussion. While the results of these and similar studies in other areas close to the Nevada Test Site are not conclusive, they may be explained in relation to the following observations.

Both solubility and particle size have been suggested (Bryant et al. 1960, Lindberg et al. 1959) as indices of the "biological availability" of Sr-90 fallout. The "availability" of Sr-90 to the animals living in a fallout contaminated area depends on "availability" for ingestion and on "availability for assimilation" during its passage through the gut. It has been demonstrated that Sr-90 is associated primarily with fallout particles < 44μ

in diameter (Larson et al. 1960), that plants are selective collectors of $< 44\mu$ particles (Romney et al. 1962a), and that the solubility of fallout particles in 0.1 N HCl tends to increase with decreasing particle size (Larson et al. 1961, Lindberg et al. 1954, 1959).

During the first few weeks after fallout, the jack rabbits living in a contaminated environment feed on vegetation containing relatively large amounts of Sr-90 which can be assimilated and deposited in bones. The period of maximum assimilation of Sr-90 by jack rabbits probably coincides with the period that fallout particles are retained by plant foliage. After the loss of externally contaminated foliage, the Sr-90 content of desert plants is probably quite low. External contamination by redistributed fallout debris is negligible, and the uptake of Sr-90 by desert shrubs from contaminated soil has never been demonstrated (Larson et al. 1962). Probably, the availability of Sr-90 for ingestion by jack rabbits is greatly reduced during the first year after fallout.

If the average life-span of jack rabbits is 3.2 years, the population 3.7 years after fallout should be composed largely of individuals born more than 6 months after fallout. Most of the Sr-90 assimilated by these animals (assuming no further deposition of fallout on vegetation) is probably derived from soil materials accidentally ingested while feeding or while preening after dust baths, or from plants contaminated by world-wide fallout or other means.

The maintenance of relatively high Sr-90 levels in a jack rabbit population for a period of 2 to 3 years after fallout seems to indicate either a steady-state condition in which input (assimilation) is approximately equal to output (excretion) or else a condition in which both are negligible. In the case described above, a moderately high input value could have been maintained by stratospheric fallout from previous detonations of high yield.

According to this rather speculative hypothesis, the Sr-90 level in a jack rabbit population sampled 4 to 5 years after a single contaminating event should be relatively close to equilibrium. Also, the level at which this equilibrium is established should be related to the amount of biologically available Sr-90 in the soil. But, a non-linear relationship between the Sr-90 content of jack rabbit bones and the Sr-90 content of soil (total and acid soluble) might indicate that the rate of stratospheric fallout is more important in determining the rate of Sr-90 assimilation by jack rabbits than either the total or the potentially available Sr-90 in or on the soil.

Ecological models. *1. Discrimination Factors.* Strontium-90 and calcium are chemically similar and may follow the same routes of transfer in an ecosystem. According to Langham and Anderson (1958, 1959), there is little or no discrimination between Sr-90 and Ca in the transfer from soil to plants. So, the Sr90/Ca ratio in plants divided by the Sr90/Ca ratio in

soils (= DF_1) may vary from 0.8 to 1.0. In the food-chain transfer from plant to cow to milk, the discrimination factor (DF_2) has been estimated as 0.13. The discrimination factor for the transfer from plants to bone (DF_3) and that for transfer from milk to bone (DF_4) have both been estimated as 0.25.

If 80% of the calcium in the diet of a human population (e.g., the population of the United States) is derived from milk and 20% is derived from other sources, the overall discrimination factor in going from soil to bone ($OR_{bone/soil}$) = $(0.8 \times DF_1 \times DF_2 \times DF_3 + (0.2 \times DF_1 \times DF_4) = (0.8 \times 1.0 \times 0.13 \times 0.25) - (0.2 \times 1.0 \times 0.25) = 0.058$ and indicates an equilibrium Sr-90/Ca ratio equal to 5.8% of the Sr-90/Ca ratio in soil or plants.

If the average concentrations of available Sr-90 and Ca in soils are known, this model can be used to predict the specific Sr-90 activity of Ca deposited in the skeleton during a given period of environmental contamination. Allowances for dietary differences can be made by using a general expression, $OR_{bone/soil} = (M_f \times 0.25 + (R_f \times 0.25)$, in which M_f and R_f are the fractions of dietary Ca derived from dairy products and from other sources, respectively.

The admitted defects of this model are that it makes no allowance for direct foliar contamination and assumes that both Sr-90 and Ca are uniformly mixed in the plow zone of cultivated soils. Models of this sort are useful for predicting equilibrium or steady-state concentrations of radioisotopes in ecosystems which have been contaminated by fallout but are receiving no new additions from sources outside the system.

2. Proportionality Constants for Prediction of Sr-90 in Human Diet. The Health and Safety Laboratory (HASL) of the U. S. Atomic Energy Commission, the U. S. Public Health Service and many other agencies in the United States and throughout the world have established monitoring programs to measure the levels of Sr-90 in soils, in milk and other human foods, and in fallout—especially the fallout deposited in rain and snow. It is generally agreed that the amount of Sr-90 in or on plants, in the diet and bones of domestic animals and in the diet and bones of human populations may vary from place to place in relation to: (a) the total (or available) concentration of Sr-90 in soil and (b) the rate of Sr-90 deposition in world-wide fallout.

Knapp (1961) in the United States and Kulp et al. (1960) have studied the data derived from monitoring programs, and they have proposed empirical formulas to account for the observed levels of Sr-90 in human diets and human bones and to predict the future levels of dietary and assimilated Sr-90. The formula proposed by Knapp to account for the Sr-90 in U. S. milk supplies is based on empirically derived proportionality constants and seems to fit the available data.

$$\begin{bmatrix}\text{Average Sr-90 in U. S. milk, expressed as } \mu\mu\text{c/l}\end{bmatrix} = \frac{1}{9}\begin{bmatrix}\text{Average Sr-90 in U. S. soil, expressed as mc/mi}^2\end{bmatrix} + 2.6\begin{bmatrix}\text{Average Sr-90 deposition for preceding month, expressed as mc/mi}^2\end{bmatrix}$$

According to estimates based on this formula, 51% to 83% of the Sr-90 in U. S. milk (an average of 69%) during 1958 was derived from soil, and the remaining 17% to 49% (an average of 31%) was derived from world-wide fallout. Burton et al. (1960) have reported that 20% of the Sr-90 in milk produced in the United Kingdom during 1958 was derived from soil deposits and 80% from fresh fallout. The difference between Knapp's interpretation of Sr-90 levels in U. S. milk and Burton et al.'s interpretation of data collected in the United Kingdom have not been explained, but it seems quite likely that a reasonable explanation of these differences could be found by making detailed studies and comparisons of the Sr-90 fallout and its cycling in representative pasture ecosystems in the two countries.

The formula outlined below is similar to those proposed by Knapp (1961) and by Langham and Anderson (1959) and has been used by Dunning (1962) to predict the maximum possible levels of Sr-90 in U. S. milk and in the total diet of people living in the U. S. The soil and fallout levels used for this purpose are based on the prediction that fallout from the 1961 test series in Russia will raise the average concentration of Sr-90 in U. S. soils from about 80 to about 125 mc/mi^2.

According to Dunning (1962):

$C = aT + bR$, where:
C = concentration (Sr Units) in diet
T = total deposition (mc/mi^2 = 125)
R = rate of Sr-90 fallout (mc/mi^2/yr = 45)
a & b = proportionality constants:
For U. S. milk : a = 0.1 & b = 0.4
For total diet : a = 0.15 & b = 0.2

By calculation, $C = (0.1 \times 125) + (0.2 \times 45) = 21.5$ Sr Units in milk; and $C = (0.15 \times 125) + (0.2 \times 45) = 36.75$ Sr Units in the total diet of people living in the U. S. during 1962. Because the formula assumes that 100% of the fallout on the U. S. will occur in one year, these estimates are probably too high.

Ecological models of the kind described above are particularly useful in helping to evaluate, on a nation-wide or world-wide scale, the potential short- and long-term hazards of fallout to human populations. They provide a basis for predicting future levels of particular fission products in human populations and in parts of the food-chains leading to man.

The problem of developing mathematical models to describe and predict the concentration of a given radionuclide in a particular ecosystem is perhaps too complex to be solved by the application of simple, empirical, or deterministic proportionality constants. A major source of the complexity of this problem is the wide variability of radionuclide concentrations in the biotic and abiotic components of a given ecosystem. Geographic variability may be even greater.

3. Other Deterministic Models. In the case of short-lived radionuclides such as I-131, a deterministic model may be adequate for quantitative studies during the acute phase of environmental contamination. French (1959) has used such a model to account for the acitvity of jack rabbit thyroids following a single release of radioiodine. The formula used by French is as follows:

$$A = I \times F \times \left(\frac{e^{-\lambda_p t} - e^{-\lambda_E t}}{\lambda_E - \lambda_p} \right)$$

A = activity in thyroids
I = total I-131 ingested on first day
F = fraction of I-131 reaching thyroid
λ_E = effective I-131 decay constant in jack rabbits
λ_p = physical decay constant for I-131
e = 2.7183
t = time

If "A" is known, "I" can be estimated by transposition; and this value can be interpreted as an estimate of plant contamination, but there are several possible sources of error. French found that F varied from 15% in the summer to 29% in the winter while λ_E varied from .693/1.5 in winter to .693/2.5 in summer. This model assumes that the loss of I-131 activity from plants is equal to the physical decay constant (λ_p), but Chamberlain (1958) has reported that the loss of radioiodine from plants may occur at a greater rate. It is possible, therefore, that some expression should be included in the formula to account for this source of variability.*

4. The Need for Stochastic Models. Mathematical models for longer-lived isotopes require expressions to account for an even greater variety of variables. In considering the circulation of radiostrontium, for example, in a simple desert ecosystem, it would be necessary to consider several input and several output factors for each of the major abiotic and biotic components of the system (Fig. 5). To account for the radiostrontium in or on plants (= animal diets) the input factors would be related to fallout (external de-

* Studies completed since this paper was written (Turner and Martin 1963, 1964) have used field data to test this model; and the model has been rewritten, in stochastic form, to estimate the frequency distribution of "A" based on limited, random variations in the other parameters.

position) and to soil contamination (root absorption). The output expressions would have to account for physical decay, loss of particles due to the mechanical action of wind and rain, loss of contaminated foliage by leaf fall, and loss of foliage to herbivorous animals. Similar input and output expressions would have to be applied to all the integral parts of the system (e.g. atmosphere, soil, plants, animals, and decomposers). Even then a deterministic model would be no more than an approximation of its ecosystem analog; but successively closer approximations could be obtained by the inclusion of expressions to account for seasonal changes in ecosystem dynamics, for species differences, and for other significant rate-determining parameters.

To account for the variability normally encountered in nature and to facilitate the comparison of radionuclides cycling in similar and dissimilar ecosystems under different conditions of contamination, deterministic models might profitably be rewritten in a stochastic (= probabilistic) form and programmed for analysis by analog or digital computers. This approach as taken by Olson (1961), and perhaps by others unknown to the writer, seems to be representative of the current frontiers in radioecological research concerning the occurrence, redistribution, and cycling of radionuclides in the biosphere.

Evaluation of potential biological hazards related to fallout. The scientific literature in which various estimates of the biological cost of radioactive fallout have been presented is quite formidable both in size and diversity. It is not within the scope of this paper to review the literature dealing with the problem of fallout hazards to man, but it does seem appropriate to conclude with a few general remarks concerning the nature of the problem and the kinds of evidence which are available to those who are attempting to solve it.

Plants, animals and people have always been exposed to ionizing radiation in their natural environments. Because of variations in the distribution of cosmic radiation and the radionuclides which occur naturally in the atmosphere, lithosphere, and hydrosphere, organisms in certain habitats, especially at higher elevations, have normally been exposed to radiation doses at least 3 times as high as the average for the biosphere. The average worldwide increases in background radiation as a result of fallout has been considerably less than this normal variation (Larson et al. 1962). The increased exposure of organisms to radiation from fission products may also be rather small in relation to natural sources of internal radiation. Milk, for example, which contained 3 to 12 $\mu\mu$ c/l of Sr-90 in 1961 also contained 800 to 1400 $\mu\mu$ c/l of K-40. (Consumers Union 1962).

"The net result of fallout," according to Langham and Anderson (1958), "has been a small increase in the radiation background to which life is exposed. The problem of evaluating the potential hazard of world-wide fallout then becomes one of trying to ascertain the magnitude and significance of

this increase in the background dose with regard to its potential effect on man's health and well-being."

The most pessimistic estimates of the potential effects of fallout on human populations have been based on the statistical probability that small increases in exposure to radiation may result in a shortening of the average life-span or in the increased incidence of detrimental gene mutations, leukemia, and bone tumors. Even a small increase in the incidence of detrimental genetic or somatic effects, may involve a large number of individuals. In the present world population, an overall increase of one leukemia case per million people would involve approximately $(1 \times 10^{-6}) \times (3 \times 10^{9}) = 3000$ people; but, for obvious reasons, increases of this magnitude are extremely difficult to detect.

Most of our knowledge of the specific biological effects of ionizing radiation have been derived from experiments in which animals (or plants) were exposed to relatively high levels of irradiation. Most of the detrimental effects of radiation are known to occur spontaneously, and the experimenter must rely on statistical tests to measure differences between the spontaneous and the experimentally induced incidence of such effects. In order to obtain statistically valid results at low levels of radiation exposure, it would be necessary to use very large numbers of organisms. In practice it is usually expedient to use higher levels of radiation exposure and smaller numbers of organisms.

Most of our knowledge of the effects of ionizing radiation on people has been derived from clinical studies in which groups of individuals who had been exposed, occupationally, to higher than average doses of radiation were compared to similar groups of people who had not been exposed to higher than average doses of radiation. It has been demonstrated statistically, for example, that radiologists have a shorter average life-span than other physicians (Advisory Committee on Biology and Medicine 1958).

Our direct evidence of the biological effects of fallout on human populations is meager and has been derived from studies made after accidents such as the Windscale incident in Great Britain (Chamberlain 1958), the Lucky Dragon incident and the contamination of the Rondelapp Islands in 1954 (Cohn et al. 1960). During the several test series at the Nevada Test Site, there have been a few reports of superficial radiation burns on cattle, but there have been no observations of detrimental radiation effects on native plants (Special Subcommittee on Radiation 85th Congress 1957) or animals (Libby 1956a).

Because of the paucity of direct evidence, most attempts to evaluate the potential hazards of fallout to man have been based on extrapolations from experimental and clinical evidence. In most cases this involves extrapolation from animals to man or from high to low levels of exposure to radiation or both.

According to the Advisory Committee on Biology and Medicine (1958),

estimates of the potential genetic effects of fallout are probably more reliable than estimates of potential somatic effects. If there is no threshold for the induction of mutations by ionizing radiation, any increase in the exposure of gametes to radiation will result in an increase in the number of mutant genes; and the incidence of mutations will be proportional to the amount of radiation received by gametes. In this situation, it is a relatively simple matter to extrapolate from higher to lower levels of gametic exposure to ionizing radiation.

Extrapolations from clinical and experimental evidence of somatic effects are often confounded and made unreliable by a variety of uncertainties. For example, the mechanisms by which ionizing radiation may cause leukemia or bone tumors or result in the shortening of human life are not clearly understood. In most cases, the possibility that radiation below some critical threshold may produce no increase in the incidence of a given somatic effect can neither be affirmed nor denied.

According to Dunham (1958), "Atomic energy like the other great technological advances is bound to exact some price of the society which makes use of it whether in peaceful pursuits or in its national defense effort. Our present knowledge of the hazards of radiation though incomplete is greater than for any other environmental hazard. It is for the radiobiologist to continue to define the cost in more and more precise terms while it is up to society to decide whether the price is acceptable, and if the answer is in the affirmative, it must make certain that the cost is kept at a minimum."

Radioecology can contribute to the solution of these problems by continuing to study the fate of fission products in the biosphere, and by the establishment of long-term ecological studies to determine the consequences, if any, of the chronic exposure of plant and animal populations and communities in their natural environments to low levels of ionizing radiation from external and internal sources. Studies of the former kind have been described in this paper. Some long-term ecological studies have already been inaugurated (Wolfe 1961) but significant results from these studies may not be forth-coming for 10, 20 or 50 years.

In order to gain a thorough understanding of the long-term effects of fallout and other sources of ionizing radiation on the biosphere, it would be desirable, as soon as possible, to begin continuing studies of fundamental ecology and radiation effects in every major part of the biosphere. According to Wolfe (1961), the ecological effects of increased environmental radiation are not known. "Nor can they be determined by experiment alone, nor by considering only a single source of increased radiation, such as fallout. Determination of the total impact of this factor on man's biotic environment, and the evolution of living organisms therein, is a continuing problem." It is this problem, and its many ramifications, that justifies the description of radioecology as an application of fundamental biology to the needs of man.

Acknowledgments. The writer gratefully acknowledges the assistance given by Mr. Kermit H. Larson, Dr. William A. Rhoads, and Dr. Thomas G. Hennessy who read the manuscript in rough form and offered many helpful suggestions for its improvement. Thanks are due also to Mr. Richard Rowland and Mrs. Ruth Pratt who did the art work, and to Mrs. Ann Kehrer and Miss Lois Eby who helped in preparing the manuscript. Special thanks are due to the biological faculty of Rutgers University and to the National Science Foundation who made it possible to present this paper at the 18th Annual Meeting of the Bureau of Biological Research on April 27, 1962.

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Radionuclide Concentrations in Surface Air: Direct Relationship to Global Fallout

Abstract. *By relating the average monthly hemispheric concentrations of radionuclides in the surface air to the monthly hemispheric fallout of these nuclides, we have derived proportionality constants. For every disintegration per minute of a radioactive nuclide per 1000 cubic meters of the surface air in the Northern Hemisphere, 3.3 kilocuries of that nuclide will be deposited on the surface; in the Southern Hemisphere, 5.0 kilocuries will deposit for every disintegration per minute per 1000 cubic meters. On the basis of these factors, the global deposition of radionuclides can be estimated from a relatively small number of measurements of the radioactivity in the surface air.*

Using data obtained in two U.S. AEC Health and Safety Laboratory (HASL) sampling programs, we report here on an attempt to relate the concentration of strontium-90 in samples of surface air to the deposition of Sr^{90} on a worldwide scale. Since the beginning of 1963, HASL has been conducting measurements of the monthly concentrations of selected radionuclides in surface air (I) at about 25 stations, mostly along the 80th meridian ranging in latitude from Thule, Greenland, at about 76°N to the South Pole. Likewise, the monthly deposition of Sr^{90} has been measured since 1959 at more than 100 sites (*2*, *3*).

If we assume that the concentration of a nuclide in surface air is directly proportional to the amount of that nuclide deposited on the surface of the earth, the following equation may be written:

$$D_n = X C_n \qquad (1)$$

where D_n is the deposition (in kilocuries) of a nuclide on the earth's surface during month n, X is a constant which relates the concentration of that nuclide in the surface air to its deposition, and C_n is the average concentration of that nuclide in surface air in disintegrations per minute per 1000 standard cubic meters (disintegrations per minute per 1000 m^3) during month n (calculated by weighting the concentrations measured in each 10° band of latitude according to the surface area of that latitude band).

This equation also assumes that, within each hemisphere, the troposphere is rapidly mixed longitudinally, and, therefore, that the sampling stations for the surface air are representative of their respective bands of latitude. Because the rate of interhemispheric exchange of tropospheric air is relatively slow (*4*), we consider the hemispheres to be completely insulated from each other, and separate equations are written for each.

If we substitute the measured monthly hemispheric depositions of Sr^{90} and the average monthly hemispheric concentrations of Sr^{90} in surface air into Eq. 1, the value of the constants in both the Northern and Southern hemispheres can be computed for each month in the period 1963 through 1967. The constants average 3.3 ± 0.7 kc per disintegration per minute per 1000 m^3 in the Northern Hemisphere and 5.0 ± 0.7 kc per disintegration per minute per 1000 m^3 in the Southern Hemisphere. Thus, for every disintegration per minute of Sr^{90} per 1000 m^3 of surface air in the Northern Hemisphere, 3.3 kc will be deposited on the surface of that hemisphere; similarly, for every disintegration per minute of Sr^{90} per 1000 m^3 of surface air in the Southern Hemisphere, 5.0 kc will be deposited.

The monthly values of X in each hemisphere exhibit seasonal variability. In general, it appears that the ratio of the deposition of a radionuclide to its concentration in surface air is lower in both hemispheres during the seasons of maximum fallout, the spring and winter. Because of this seasonal variability, deposition estimates for individual months may be in error by about

Table 1. Measured (M) and estimated (E) deposition of Sr^{90} (in kilocuries).

Year	Northern Hemisphere		Southern Hemisphere	
	M	E	M	E
1963	2620	3714	325	444
1964	1660	2083	436	426
1965	780	820	351	334
1966	330	376	208	282
1967	165	146	114	115
1968	190	181	90	120

Table 2. Worldwide deposition of Pu^{238} from a SNAP-9A power source (in kilocuries).

Year	Source of data		
	Measured deposition	Stratospheric depletion (*3*, *4*)	Surface air
1967	4.2	5.1	5.7
1968	3.0	2.7	3.3

30 percent however, estimates of annual deposition are not affected

The two constants, X_1 and X_2, are probably not the same value, because their averages, each based upon 60 individual monthly determinations, are not within one standard deviation The significance of the difference is not immediately attributable to any particular meteorological phenomenon, although it is intriguing to speculate on the possible contributions from the relative areas of sea in the two hemispheres and from interhemispheric transfer in the troposphere

Using the derived hemispheric constants and the measured concentrations of Sr^{90} in surface air for the period from 1963 through 1968, we calculated the annual Sr^{90} deposition These estimated deposition values are compared with the measured values in Table 1 The agreement is reasonably good especially in the most recent years Although the relatively large deviation for 1963 and 1964 might suggest that X varies with the age of debris, there is no evidence of such a variation in 1967 and 1968 when the French and Chinese atmospheric tests occurred and most of the deposited radionuclides were "fresh"

This method of computing fallout from measured concentrations of Sr^{90} in surface air should be applicable to all materials dispersed like Sr^{90} in the atmosphere The only other radionuclide whose fallout we could verify with deposition measurements was Pu^{238} which was disseminated during the reentry and burnup of a SNAP-9A (Systems for Nuclear Auxiliary Power) power source on a weather satellite (*5*) The estimates of plutonium-238 deposition for 1967 and 1968 are shown in Table 2, along with the measured values (*6*) and values derived from the annual depletion of the stratospheric reservoir extrapolated to the end of 1968 (*7*) Unfortunately, the data on Pu^{238} in surface air prior to 1967 are not reliable, and comparisons with early deposition measurements could not be made The estimated values are in good agreement with the measured values from 1967 and 1968

It appears therefore, that the annual deposition of a nuclide can be accurately calculated from average monthly hemispheric concentrations of that nuclide in surface air The significance of this relationship between the concentration of a radionuclide in surface air and its corresponding fallout lies in the relative simplicity of surface air sampling and the sensitivity of the existing system If we can make worldwide estimates of fallout with acceptable accuracy, based simply upon a limited number of measurements of radionuclides in surface air, we can effect substantial savings in both time and funds Further refinement of this relationship to smaller specific geographic areas should make possible estimates of local deposition and may lead to a greater understanding of hemispheric differences In addition, the possibility of estimating the worldwide contamination from pollutants dispersed in a manner similar to that of global fallout is of great value

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16 June 1969

Reprinted from SCIENCE,
17 October 1969 volume 166, pages 376 377

FILE - ENVIRONMENTAL
ABVE.S - AIR

RHL-69-5

STRONTIUM-90, STRONTIUM-89, PLUTONIUM-239, AND PLUTONIUM-238 CONCENTRATION IN GROUND LEVEL AIR FROM 1964 TO 1969

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STRONTIUM-90, STRONTIUM-89, PLUTONIUM-239, AND PLUTONIUM-238 CONCENTRATION IN GROUND LEVEL AIR FROM 1964 TO 1969

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STRONTIUM-90, STRONTIUM-89, PLUTONIUM-239, AND PLUTONIUM-238 CONCENTRATIONS IN GROUND LEVEL AIR FROM 1964 to 1969

INTRODUCTION

The radionuclides strontium-90, strontium-89, plutonium-239*, and plutonium-238 are present in the atmosphere as the result of nuclear weapons tests. In the case of plutonium-238, in addition to the above source, an injection of this radionuclide into the atmosphere took place in April 1964 when 17 kilocuries of plutonium-238, contained in a SNAP-9A power source, burned up south of the equator off the coast of Africa.(1) Information on airborne strontium and plutonium isotopes is of interest in evaluating the potential health hazards of these materials and in investigating the transport processes involved. Analysis of ground level airborne particulate samples for the radionuclides of strontium has been performed at the Northeastern Radiological Health Laboratory (NERHL) since October 1963. For the plutonium isotopes, radiochemical and alpha spectroscopic analysis has been done since May 1965. Results of these analyses to April 1966 (for strontium-90) and to February 1967 (for the plutonium isotopes) have been previously reported.(2,3) Extensive measurements of these radionuclides in air particulates and precipitation have been performed by the Health and Safety Laboratory (HASL) of the Atomic Energy Commission for several years.(4,5) In addition, the Joint Nuclear Research Center, ISPRA Establishment, Italy, has published data on SNAP-9A plutonium-238.(6)

This paper presents results on ground level airborne concentrations of the above radionuclides from the inception of the sampling and analysis program at NERHL to March 1969. A recent assessment of doses from these radionuclides, based on the concentrations presented in this paper, indicates that only an infinitesimal portion of the total strontium-90 dose to bone is due to inhalation of airborne material. On the other hand, the dose from plutonium-239 to the tracheobronchial lymph nodes was reported to be 160 millirem in 50 years, second in magnitude only to the dose from strontium-90 (from ingestion) to the bone. The dose from plutonium-238 to this lymphatic tissue was about one-quarter of that from plutonium-239.(7) In this publication attention will be given to the origin of these radionuclides and to the contribution of various atmospheric nuclear tests and the burnup of the SNAP-9A satellite to the total quantity of the radionuclides present in ground level air. A summary of reported atmospheric detonations or vents which occurred during the period of this study is given in Table 1.(8)

METHODOLOGY

Monthly composite particulate samples, representing 12,000 m^3 of air, are collected on 8' X 10' membrane filters (pore size 0.8 micron at a height of one meter above the ground. The samples are wet ashed with a mixture of nitric and perchloric acids. The sample is split into two parts for radiochemical analysis.

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* The energies of the alpha particles from plutonium-239 and plutonium-240 are not sufficiently different to be separated by alpha spectroscopy. Therefore, when plutonium-239 is referred to in this report, it is meant to represent the sum of the activities of plutonium-239 and plutonium-240.

One-half the sample is analyzed for strontium-90 and strontium-89 as follows: Strontium carrier is added and the strontium is precipitated from the samples first as the carbonate then as the nitrate. Further purification is made by barium chromate precipitation and hydroxide scavenging. After an ingrowth period, yttrium carrier is added and the yttrium is extracted into TTA (2-thenolytrifluoroacetate) at pH = 5.0 The yttrium is stripped from the TTA with dilute nitric acid, precipitated as the oxalate and beta counted for yttrium-90. The strontium is precipitated as the carbonate and beta counted for total radio-strontium. The strontium-89 activity is calculated from the total radio-strontium measurement after correction for the strontium-90 content.

Analysis of the other half of the sample for the plutonium isotopes is briefly described below: Plutonium-236 tracer is added, the plutonium reduced to the +3 state and co-precipitated with lanthanum fluoride. The lanthanum fluoride is converted to lanthanum hydroxide, dissolved in 7.2 M nitric acid and the plutonium oxidized to the +4 state. The solution is passed over an anion exchange resin in the nitrate form. The resin is washed with additional nitric acid and then with 9 M hydrochloric acid. The plutonium is then eluted from the resin with a mixture of 0.36 M hydrochloric acid and 0.01 M hydrofluoric acid, electroplated onto a stainless steel planchet from a sulfuric acid-ammonium sulfate electrolyte, and counted with a silicon surface barrier detector linked to a multichannel analyzer. With the sample electroplated onto a 3.1 cm^2 area, the counting efficiency is ~31% and the resolution 75 KeV.

The minimum detectable quantities are 0.10 fCi/m^3, 0.40 fCi/m^3, and 0.003 fCi/m^3 for strontium-90, strontium-89 and the plutonium isotopes respectively. The maximum 2σ counting error for the strontium-90 is 25 percent of the reported value, although in the majority of cases the 2σ counting error is approximately 10 percent of the reported value. The corresponding values for strontium-89 are about twice as high. The maximum 2σ counting errors are 50 percent and 67 percent of the reported values for plutonium-239 and plutonium-238 respectively, but are appreciably lower as the results increase above the minimum detectable levels.

RESULTS AND DISCUSSIONS

The results of strontium-90 analyses of ground level airborne particulates from monthly composite samples are presented in Figure 1. Difficulties in sampling and analysis caused the loss of two samples (October 1963 and August 1967), and during a change over in personnel responsible for collection and analysis of the data, several months of ambiguous data (September, October, November, and December 1967) occurred. Figure 1 illustrates the expected spring maximum which occurs each year and which appears to be extremely reproducible in terms of time of appearance. A line (solid) indicating the expected spring maximum highs, based on the strontium-90 concentrations in the spring of 1964 and a stratospheric residence of half-time of ten months,[9] is shown. The expected strontium-90 curve (dashed), having the shape of the 1964 curve because no fresh intrusion of material occurred until the end of that year, is also shown. The difference in the expected and actual curves indicates strontium-90 of post 1963 origin was present.

A double peak in strontium-90 levels appears in 1968. Although not apparent from this study other investigators have noted the presence of fresh fission products during August 1968 and attribute the rise in radioactivity to the rapid movement of debris from the French tests in July 1968.[10] Strontium-90 levels in the latter part of 1968 would be elevated if such an intrusion occurred.

Strontium-89 concentrations are shown in Table 2. Strontium-89 values are indicated only in months where the levels of this radionuclide were above the minimal detectable level. All the appearances of strontium-89 followed Chinese atmospheric tests. This fact, together with information that, except in the case noted above, French tests were not measurable 10°N latitude[11] and that the reported yields of releases at NTS were low and of a limited nature[3], appears to make the Chinese detonations the principal source of increases above the expected strontium-90 levels.

The contribution of strontium-90 from a particular test may be estimated from the strontium-89 to strontium-90 ratio. These estimates are also presented in Table 2. The theoretical ratios are for uranium-235 fission.[12] In most cases, the contribution of strontium-90 ground level air from a recent nuclear test based on the strontium-89 ratios, appears to be of trospheric origin. However, the strontium-89 to strontium-90 ratio following the fifth Chinese test appears to be influenced by the presence of strontium-89 of stratospheric origin beginning in March of 1967.

Early low yield tests contributed only small amounts to ground level strontium-90 concentrations. The fifth Chinese test (December 27, 1966) appears to have contributed a substantial quantity of strontium-90. There was no evidence of a fresh strontium-90 intrusion following the sixth Chinese test (June 17, 1967), although complete data are not available. For the two months following the test, for which results were available, there was no indication of elevated strontium-89 levels. Data from HASL also indicate the absence of strontium-89 from this test.*[11]

From the differences in projected and actual levels of strontium-90, an estimate of the strontium-90 from atmospheric testing after 1963 can be made. Table 3 presents the mean predicted (based on a 10-month residence half-time and the mean 1964 results) and measured strontium-90 concentrations in ground level air. Because of the magnitude of errors in analyses, it is believed that only the values for 1968 are truly significant, although a gradual increasing trend in newly injected strontium-90 since 1965 is indicated. In 1968, 62 percent of the total ground level strontium-90 was due to post-1963 tests. HASL estimates that 87 percent of the total reservoir of strontium-90 in 1968 could be attributed to post-1963 debris.[14]

Concentrations of plutonium-239 in ground level air particulates are presented in Figure 2. The pattern of minimums and maximums is similar to those for strontium-90. The same limitations (sample loss and ambiguity) hold as for the

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* It has been suggested that this device exploded at a comparative low altitude (14 km), but that the bulk of the debris was carried to a high altitude by the rising fire-ball.[13]

strontium-90 data. The average ratio of plutonium-239 to strontium-90 for the period May 1965 through February 1968 was 0.017 with a 2σrange of ± 0.009. There is a gradual rise in this ratio following this period, the average value being 0.028 from March 1968 to March 1969. This increase in the plutonium-239 to strontium-90 ratio is attributed to the sixth, seventh and eighth Chinese tests, which were reported to be high yield events.

The ratio of plutonium-238 to plutonium-239 remained relatively constant at 0.04 ± 0.02 from May 1965 to July 1966. A change in the ratio was fairly evident at NERHL in mid-1966[3] This change in ratio indicated a new source of plutonium-238 other than that which was previously present. Other investigators announced this change in plutonium-238 to plutonium-239 ratio somewhat earlier in June and July of 1966[6], the conclusion being that SNAP-9A plutonium-238 had reached ground level in the Northern Hemisphere.

Figure 3 is a plot of the SNAP-9A plutonium-238 concentrations in ground level air based on the above ratio of plutonium isotopes prior to influx of the satellite debris. The dip in levels in May of 1968 appears to be an anomaly. The levels of SNAP-9A plutonium-238 are slightly elevated over the 1967 levels. This elevation is expected, based on prior predictions of increasing plutonium-238 levels in 1968.[15] A comparison of the predicted results, the reported results at ISPRA, and NERHL results is presented in Table 4. The NERHL results (3-month averages) are somewhat lower than the predictions.

SUMMARY

Data have been presented on the concentrations of strontium-90, plutonium-239, and plutonium-238 in ground level air particulates. From these data, the following observations may be made:

1. The Chinese atmospheric nuclear tests appear to have contributed the majority of the influx of fresh strontium-90 in ground level air particulates.

2. It is estimated that in 1968 approximately 60 percent of the strontium-90 present in ground level air is of post-1963 origin.

3. The ratio of plutonium-239 to strontium-90 is somewhat elevated since early 1968 due to recent Chinese atmospheric tests.

4. The levels of SNAP-9A plutonium-238 debris in ground level air are somewhat lower than had been predicted.

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TABLE 1

REPORTED ATMOSPHERIC DETONATIONS[8]

Date	Source	Comments
October 16, 1964	Chinese (1st)	Low**
May 14, 1965	Chinese (2nd)	Low
May 9, 1966	Chinese (3rd)	Intermediate 200 KT
July 2, 1966	French	70-80 KT
July 19, 1966	French	120 KT
September 24, 1966	French	150 KT
October 4, 1966	French	Intermediate 200-300 KT
October 27, 1966	Chinese (4th)	Low-Intermediate 20-200 KT
December 28, 1966	Chinese (5th)	Few hundred kilotons
June 5, 1967	French	Low
June 17, 1967	Chinese (6th)	2 MT
June 27, 1967	French	Low
July 3, 1967	French	Low
December 24, 1967	Chinese (7th)	Megaton Range
July 7, 1968	French	Low
July 15, 1968	French	Low
August 3, 1968	French	Low
August 24, 1968	French	2 MT
September 9, 1968	French	2 MT
December 28, 1968	Chinese (8th)	2 MT

** Less than 20 KT

TABLE 2

STRONTIUM-89 CONCENTRATIONS AND CONTRIBUTION OF FRESH STRONTIUM-90 FROM ATMOSPHERIC TESTS

st and te	Sample Collected Month and Year	^{89}Sr Concentration (fCi/m^3)	Time Elapsed Since Test (days)	Ratio ^{89}Sr to ^{90}Sr		^{90}Sr From Test(?
				Theoretical(12)	Measured	
nese #1 t.16,1964)	Nov. 1964	10 ± 3*	30	111	1.6	1.5
	Dec.	ND	60	74	ND	-
	Jan. 1965	16 ± 3	91	49	0.8	1.6
nese #2 y 14,1965)	June 1965	57.9 ± 2.7	30	111	2.9	2.6
	July	28.2 ± 2.6	60	74	1.7	2.3
	August	2.4 ± 1.1	90	39	0.3	0.8
	Sept.	1.5 ± 1.2	120	33	0.4	1.2
nese #3 y 9, 1966)	June 1966	13.1 ± 2.8	37	100	1.8	1.8
	July	10.4 ± 1.7	67	67	2.2	3.3
	August	4.3 ± 0.8	108	38	1.7	4.5
	Sept.	1.8 ± 0.7	139	25	0.9	3.6
ese #4 .27, 1966)	Nov. 1966	18.7 ± 1.7	19	129	8.5	6.6
	Dec.	6.5 ± 0.8	49	86	5.3	6.2
ese #5 . 27,1966)	Jan. 1967	11.9 ± 1.2	19	129	7.0	5.4
	Feb.	15.5 ± 1.1	49	86	5.3	6.2
	March	27.4 ± 1.6	81	55	9.9	18
	April	34.7 ± 1.6	112	36	8.5	23
	May	14.0 ± 1.8	142	24	4.5	19
	June	5.3 ± 0.9	173	16	2.0	13
se #6 17,1967)	No evidence of ^{89}Sr in August or September 1967. July sample lost and September through December sampling or analytical ambiguities occurred.					
se #7 24,1967)	Jan. 1968	6.5 ± 0.6	22	124	5.8	4.7
	Feb.	18.3 ± 1.1	53	81	7.2	11
	March	14.8 ± 1.1	81	55	5.1	9.3
	April	13.1 ± 1.1	111	36	3.4	9.5
	May	3.1 ± 0.7	141	24	1.3	5.5
	June	2.9 ± 0.7	172	16	0.9	5.6
e #8 28, 1968)	Feb. 1969	2.3 ± 0.5	48	87	2.2	2.5

counting error.
ot determined.

TABLE 3

ESTIMATED CONTRIBUTION OF POST-1963 TESTS TO ^{90}Sr

Year	Mean ^{90}Sr (fCi/m^3) Predicted	Measured	Estimated Contribution Post-1963 Tests (%)
1964	24.1*	24.1	--
1965	10.5	11.3	7
1966	4.6	4.6	--
1967	2.0	2.3**	15
1968	0.9	2.4	62

*
**Predicted value based on measured results for 1964.
No data for July, October, November, and December.

TABLE 4

SNAP-9A PLUTONIUM IN GROUND LEVEL AIR (fCi/m^3)

Date	Predicted	ISPRA	NERHL
January 1966	0.011	0.001	---
July 1966	0.013	0.003	---
January 1967	0.015	0.005	0.004
July 1967	0.016	0.015	0.011
January 1968	0.017	0.012	0.014
July 1968	0.018	---	0.010

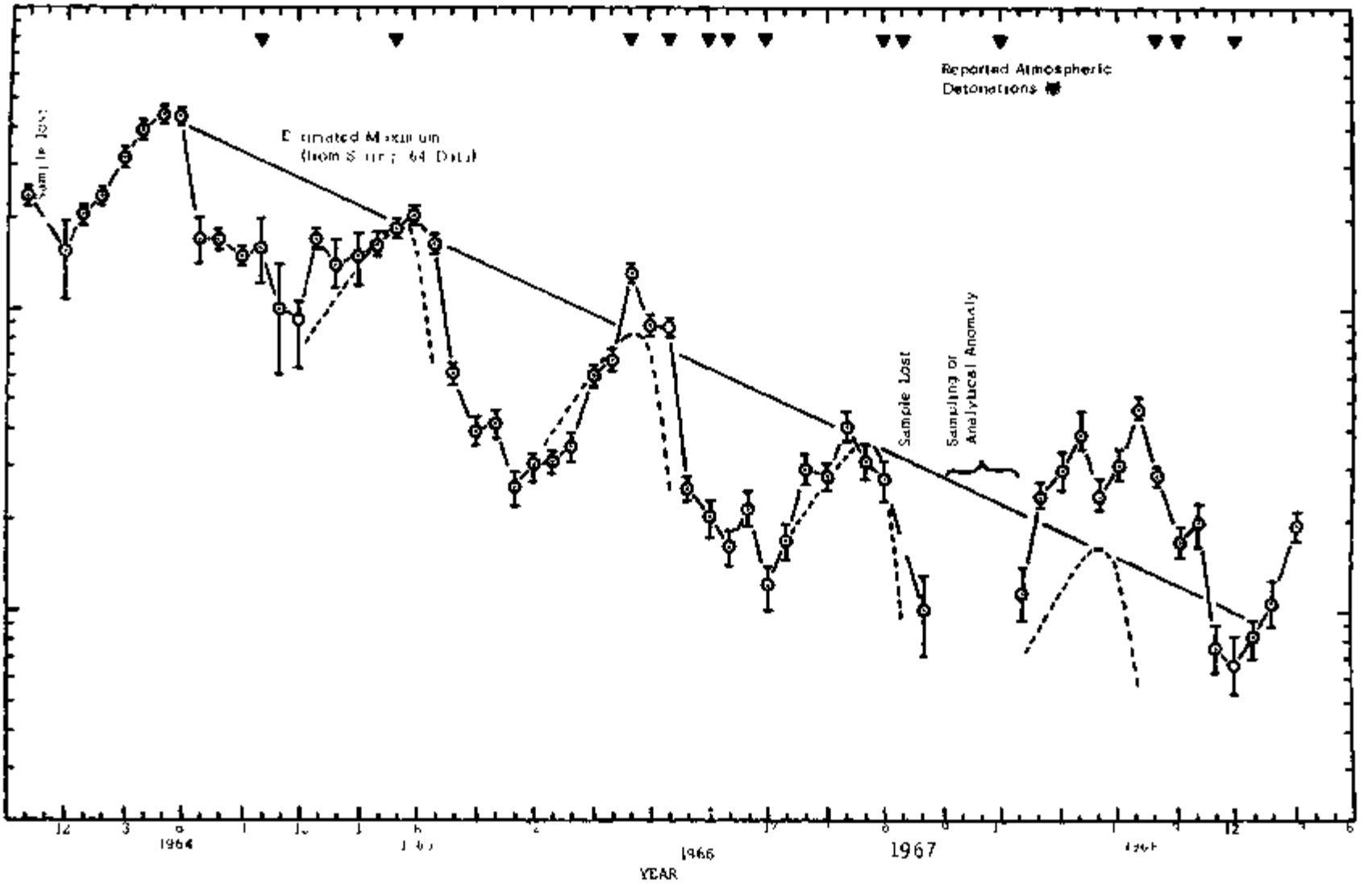

Figure 1 - Strontium-90 in Ground Level Air

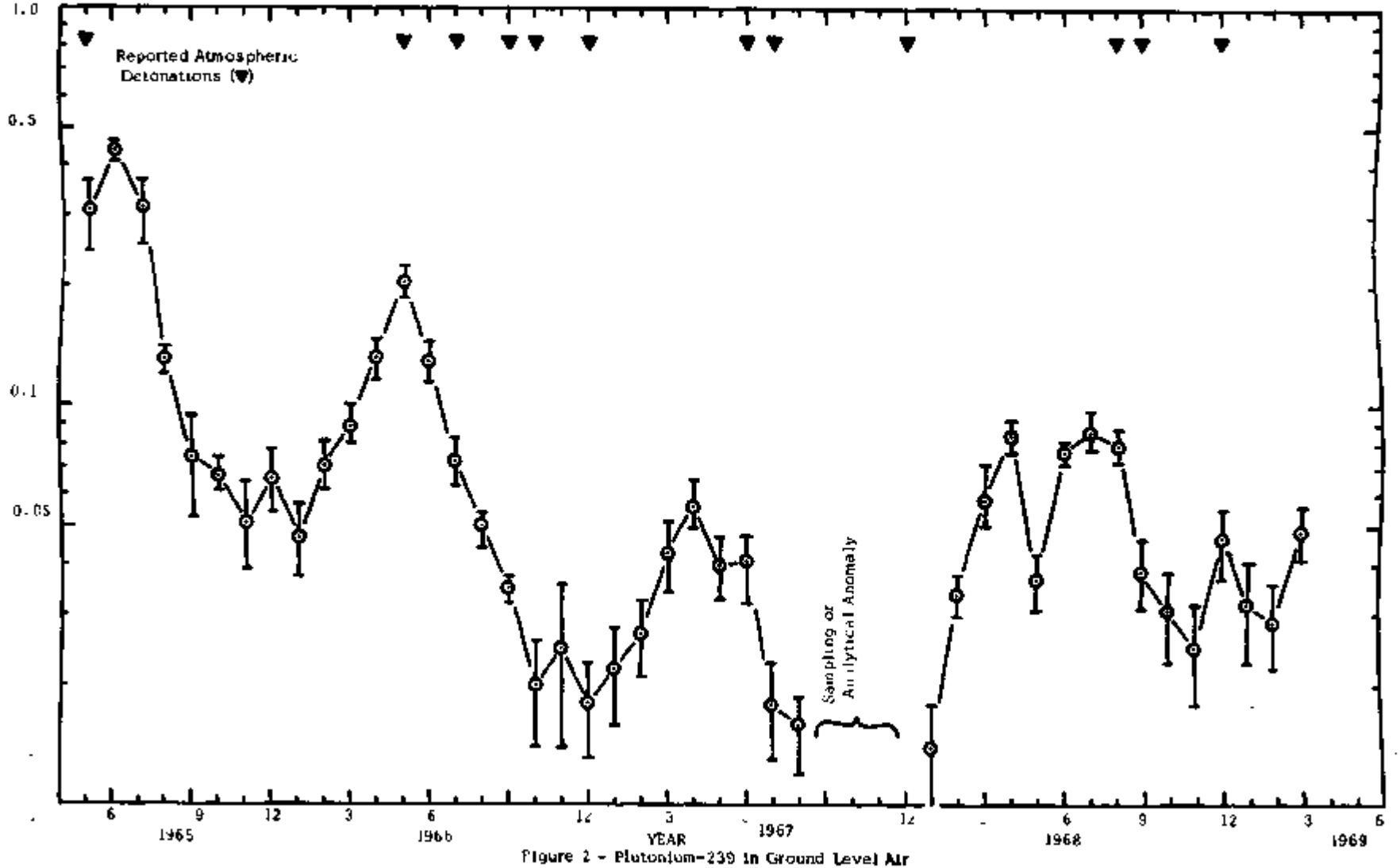

Figure 2 - Plutonium-239 in Ground Level Air

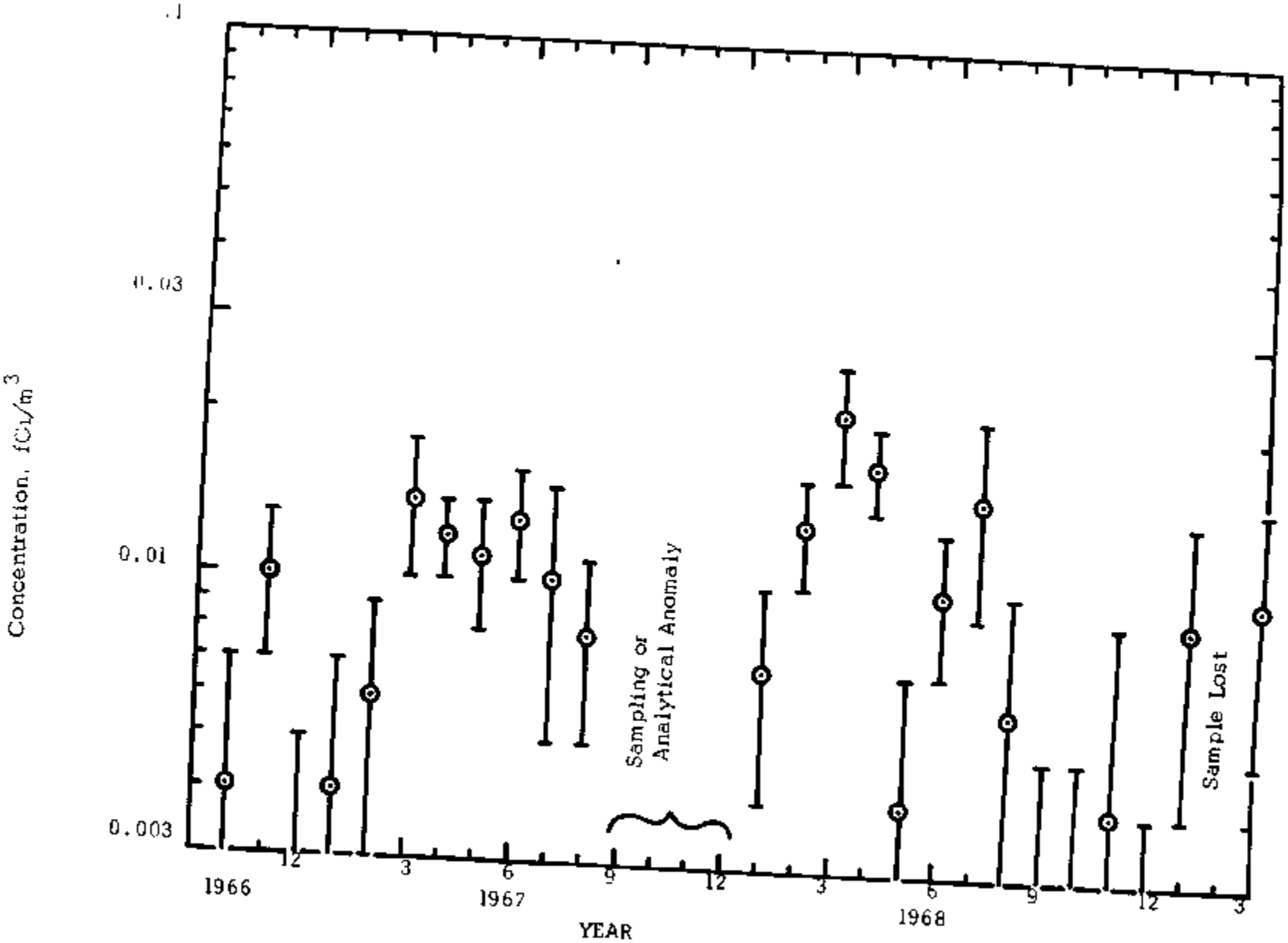

Figure 3 - Estimated SNAP-9A Plutonium-238
In Ground Level Air