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THE BEHAVIOR OF RADIOACTIVE FALLOUT IN SOILS AND PLANTS

A Review Prepared for the

Committee on Effects of Atomic Radiation on Agriculture
and Food Supplies
National Academy of Sciences—National Research Council

by

M. H. Frere and R. G. Menzel
Soil Scientists
U. S. Soils Laboratory
Agricultural Research Service
U. S. Department of Agriculture
Beltsville, Maryland

K. H. Larson
Chief
Environmental Radiation Division
Laboratory of Nuclear Medicine and Radiation Biology
University of California
Los Angeles, California

Roy Overstreet
Professor of Soil Chemistry
Department of Soils and Plant Nutrition
University of California
Berkeley, California

R. F. Reitemeier
Soil Scientist
Division of Biology and Medicine
U. S. Atomic Energy Commission
Washington, D. C.

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FOREWORD

Although man has always been exposed to some radiation from naturally-occurring radionuclides in his environment, and although the food he consumes has always carried some small burden of radioactivity, the coming of the "Atomic Age" has already brought with it a rise in the level of radiation to which man is exposed and the appearance of some new sources of radiation that did not in the past constitute a part of the natural background. The latter has been tacitly accepted as being of no great concern, even though the level of exposure may vary widely from place to place. The testing of nuclear weapons has resulted in the appearance in man's environment the world over of radionuclides not formerly present.

Man's exposure to radiation is in part external—from the materials around him—but it is also in part internal, by reason of the ingestion of food and water having some radioactive components and the inhalation of radioactive particulates or gases in the atmosphere. The effects, if any, on the well-being of the individual "depend upon the radiation dose (and dose rate) delivered to various tissues and upon the radiosensitivity of the tissues."

This report deals with some of the early steps in the sequence of events that transfers radionuclides in the environment to the tissues of man in what has come to be referred to as the "food chain." The food chain of man is not inherently more complicated than those of other organisms that are herbivorous or carnivorous. There is, however, the difference that man has considerable freedom of selection and, at least in industrialized countries, subsists on foods, fresh or processed, derived from diverse and often remote locations. The dietary exposure of man to radionuclides is therefore a most complex question that can be approached realistically only by examining the principles involved in the various steps of the food chain.

Recognizing the fact that the human diet is derived from the soil directly or indirectly through animals, the Committee sought to have prepared a comprehensive review of the fate of fallout radionuclides in cultivated soils and their transfer to or incorporation in crop plants growing thereon. This report is essentially a discussion of the principles involved and makes no attempt at evaluation of hazards to man, which have been discussed elsewhere in reports prepared by the related Committees on Pathological Effects and Genetic Effects of Atomic Radiation.

A. G. Norman, Chairman
Committee on Effects of Atomic Radiation
on Agriculture and Food Supplies

II. GENERAL

The nature and magnitude of the fallout hazard to agriculture depends upon the chemical and physical properties of fallout, characteristics of the soil and land surface, and the type and density of vegetation, as well as upon the amount of fallout. Thus, the choice of reclamation and decontamination measures would also be influenced by these factors.

The radioactivity in fallout is derived principally from fission products, and therefore depends on the fission yield of a nuclear explosion. If the fission yield gives energy equivalent to the explosion of one million tons of TNT, the gamma radiation activity of the fission products would be as shown in Table 1 (31). The beta radiation activity would be 2-20 times as great as the gamma activity (51), but unless it is in direct contact with the body, it is of less physiological significance. Alpha activity from unfissioned materials and the radioactivity from neutron-activated products is usually negligible by comparison.

TABLE 1

Total Gamma Radiation Activity of Fission Products from a 1-Megaton Explosion (31)

Time After Explosion	Activities (Megacuries)
1 hour	300,000
1 day	6,600
1 week	640
1 month	110
1 year	5.5

The rate of decay of fission products is rapid at first and becomes progressively slower with increasing time after the explosion (Table 1). This change in rate of decay is caused by the presence of a mixture of short- and long-lived nuclides in fresh fission products. The known fission products include 170 isotopes of 35 elements, ranging from zinc-72 to terbium-161 (9). Some short-lived nuclides of importance in agricultural products are iodine-131, barium-140, and strontium-89.

Many fission products of interest have radioactive daughters by decay. The chemical and biological properties of these daughter nuclides are different from those of their parents. If the half-life of the daughter is sufficiently great, its distribution in the soil or plant depends upon its characteristics, not those of its parents. Some possible effects of this phenomenon are discussed in detail elsewhere (47). The half-lives of 13 parent nuclides and 8 daughters are listed in Table 2.

TABLE 2

Half-Lives of Fission Products of Possible Significance in Food Chains and of Some Radioactive Daughter Nuclides

Atomic Mass	Parent Nuclide		Daughter Nuclide	
Number	Element	Half-Life	Element	Half-Life
91	Strontium	9.7 hours	Yttrium	58 days
131	Iodine	8.0 days		
140	Barium	12.8 days	Lanthanum	40 hours
86	Rubidium	18.6 days		
141	Cerium	28 days		
103	Ruthenium	41 days	Rhodium	5.4 minutes
89	Strontium	54 days		
95	Zirconium	65 days	Niobium	35 days
144	Cerium	275 days	Praseodymium	17 minutes
106	Ruthenium	1.0 year	Rhodium	2 hours
147	Promethium	2.3 years		
137	Cesium	26.6 years	Barium	2.6 minutes
90	Strontium	27.7 years	Yttrium	64 hours

The discussion of possible fallout patterns is beyond the scope of this report, but it should be stated that fallout distribution depends on many parameters. These include meteorological conditions, yield of the explosion, elevation of the burst, and the nature of the terrain.

The fallout from a particular surface nuclear explosion may be classified in four categories—dropout, close-in, tropospheric, and stratospheric. These categories differ in distance and time from the point of detonation. Dropout occurs at or very near ground zero, where the prompt effects of the burst are greatest. Close-in fallout consists of solid particles settling to earth under gravity within a few hours after the explosion. It may extend several hundred miles downwind from the site of a large nuclear explosion. Tropospheric and stratospheric fallout consists of very small particles which may remain suspended in air for a long time. The scavenging action of precipitation is important in bringing these particles to earth.

High concentrations of radioactive materials are found in areas receiving close-in fallout, and their subsequent distribution in soils and crops is therefore of special significance. Yet, it may be possible to take remedial actions in these areas, whereas such actions might be precluded in areas affected by dropout because of the vast physical destruction.

Less than one fourth to more than one half of the fission products formed in a nuclear explosion at or near the ground surface may return to earth as close-in fallout (51; 135, pp. 105-106). If early rain is associated with the fallout cloud, the amount of close-in fallout increases. Explosions that are so high that the fireball does not touch the ground may produce little close-in fallout.

The fate of the radioactive isotopes in deposited fallout depends on the physical properties of the fallout and the chemical behavior of the nuclides. Surface bursts in the kiloton range, over continental soils, yield predominantly siliceous radioactive particles (3, 83, 92). Particles from tower bursts in the same energy range reflect the incorporation of tower materials (83). Megaton bursts over coral islands have produced primarily calcareous particles (92). It has been reported that the dust from the Castle Bravo burst of 1954 was mainly calcite. Presumably, aragonite was evaporated, recrystallized as calcite, and precipitated as aggregates (44, 126).

This wide range in gross chemical composition, considered along with the observed range of particle sizes, leads to the conclusion that the biological availability of the constituent radioactive isotopes cannot be predicted for a particular material without some knowledge of its characteristics. The solubility in distilled water of selected particles from a continental detonation ranged from 0.28 to 1.2 per cent of the total radioactivity. One to 74 per cent was dissolved in 0.1 N HCl (83, 51). In another study (8), it was found that the solubility in 0.1 N HCl of deposited particles from four tower shots ranged from 20 to 30 per cent and that of airborne particles from 65 to 85 per cent.

Some of the nuclides of agricultural importance, notably strontium-90 and cesium-137, may be partially depleted in the local and close-in fallout. This fractionation results from the fact that precursors of these nuclides are noble gases early in the condensation of fallout particles (136, p. 72).

A major part of the biological experimentation with fallout constituents has been conducted with soluble sources of the respective isotopes. Consequently, the observed effects exceed those that would be obtained from the same amount of the isotope in the less soluble fallout. It is presumed that the use of soluble sources generally provides maximal effects.

In addition to the variability in the composition and solubility of fallout, the soil and plant aspects of the food chain contamination are complicated by variations in soil properties and differences in the structure and physiology of plant species. This will be the subject of discussion in the following sections.

III. RELATIVE AVAILABILITY OF FALLOUT CONSTITUENTS

Single crops of plants may absorb about two per cent of the total radioactivity in a soil contaminated by a nuclear explosion, but usually they absorb less than 0.1 per cent (85, 120). Strontium-89 and strontium-90 are the major nuclides absorbed (52) and may account for as much as 70 per cent of the absorbed activity from one-year-old, mixed-fission products (99). It is generally accepted that about one per cent of the applied strontium and less than 0.1 per cent of the other elements are taken up by single crops of plants (47, 78, 94, 96, 104, 105). Higher amounts of strontium uptake, 4 to 8 per cent, have been observed in pot experiments (86, 105).

The uptake of an element depends on its concentration in the external medium (17, 30, 67, 76). The ratio of plant-tissue concentration to the external-medium concentration, called a concentration factor, is used to indicate the relative uptake of the different elements. Results from solution culture studies have been based on the fresh tissue weight; oven-dry weight has been used for soil culture studies.

There is relative agreement in the order of concentration factors for different isotopes in pot experiments using the Neubauer technique or other techniques and in field experiments (95). Some reported concentration factors for fallout constituents in soil culture are 0.05 for the alkaline earth group, 0.009 for the rare earths, 0.05 for total beta activity in barley, and 0.02 for total beta activity in beans (117, 120).

Using soluble forms of isotopes in nutrient solutions (94), concentration factors from 0.05 to 1.0 have been found for strontium, cesium, iodine, and barium. The range was 0.0001 to 0.001 for ruthenium, yttrium, and cerium.

IV. SOIL REACTIONS

A. Adsorption

The adsorption of cations by soil particle surfaces from solution can occur by several processes; ion exchange is one of the most important. It was found that ion exchange increases the sorption of calcium and strontium by a volume of soil 10 times greater than that held in solution in the pore space (88, pp. 191-211). The adsorption of plutonium, cesium, strontium, yttrium, and cerium ions from solution was found to be nearly complete up to amounts equal to 0.01 times the saturation capacity of the soil (61, pp. 170-190; 62).

Strontium has a slightly higher adsorption energy than calcium (38, 48, 59). Leaching and uptake experiments indicate sites of differential adsorption (15, 38). The rate of exchange from solution to surface is rapid.[1] For soils of high "cation exchange capacity" (CEC), the reaction is essentially complete in one minute, whereas for soils of lower CEC there is a significant rise in adsorption over a longer period of time. The equilibration of strontium-89 and calcium-45 with labile soil calcium is complex, and the differential behavior of strontium and calcium increases up to 70 days (59).

Leaching soil columns with mixed-fission product solutions resulted in 80- to 85-per cent adsorption of the total radioactivity in the first few centimeters of the soil (47). This accumulation in the top few centimeters agrees with analyses of soils from test sites (108, 117, 120). Much work has been done on the adsorption of fission products from solution in relation to the disposal of waste products. In such experiments, the concentrations of radioisotopes and salts are usually in excess of those expected in agricultural soils, but some of the results at lower concentrations may be applicable.

B. Desorption

Rare earth isotopes contribute one half to three fourths of the activity in some soils contaminated by fallout (117, 120). In one soil, 50 volumes of water, corresponding to 320 inches of rain, were required to leach 10 per cent of the beta activity from one soil volume. The rate of leaching was nearly constant after the first 20 volumes. About four per cent of the radioactivity in fallout from Operation Hurricane was leached through 20 cm of soil in a 12-week field experiment (108). The leached radioactivity was mainly ruthenium-106 and rhodium-106. The activity of an equilibrium mixture of the soil, 405 days after the blast, was due mainly to ruthenium-106, rhodium-106, cerium-144, and praseodymium-144.

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[1] Unpublished results, Soil and Water Conservation Research Division, Agricultural Research Service, U. S. Department of Agriculture, Beltsville, Maryland.

Strontium is leached slowly through the soil at a rate related inversely to the CEC. Under cropping and fertilizer treatments in soil columns, calcium-45 moved about four inches downward (11), but no detectable movement three inches laterally or four inches downward was observed after 14.5 inches of rain in 89 days of field experiments (12). Strontium-90 from worldwide fallout was located primarily in the upper two inches of uncultivated soil during 1954 and 1955. In 1957, as much as one half of the strontium-90 was found in the two- to six-inch layer of some soils (1, 2).

The desorption of cesium is less than that of strontium, possibly because of fixation by micaceous minerals (113). The rate and depth of leaching increases with increments in salt concentration, acidity, and complexing agents, and with a decrease in base saturation and buffer capacity of the soil. Lime and organic matter also reduce the desorption of strontium and cesium (47).

C. Effects of Other Ions

The complementary ion exerts a strong effect on the adsorption of a cation. All cations tend to reduce strontium and cesium adsorption if used in large amounts. The order of replacement on soil materials is usually lithium < sodium < potassium < ammonium < rubidium < cesium < hydrogen < magnesium < calcium < strontium < barium < iron < aluminum < lanthanum (48; 60; 62; 124; 128, pp. 158-181; 134). Various exchange equations have been suggested, but none appears universally applicable. The effect of anions cannot be neglected, for it was found that nitrate, chloride, and sulfate reduce the sorption of strontium in that order, whereas oxalate and phosphate tend to increase it (61, pp. 170-190).

D. pH Effects

In most studies of pH effects, the pH of the leaching solution has been varied. In agriculture, the pH of the soil rather than the contaminating solution is variable. The results of some studies (61, pp. 170-190; 91; 100; 101) indicate that the maximum adsorption of strontium occurs between pH 7 and 9, cesium at 6 and higher, yttrium and cerium above 6, and plutonium from 2.5 to 9.0. Since highly acid and alkaline conditions result in the decomposition of the soil minerals, it is expected that under such conditions there would be less fission-product adsorption as a result of competition by the products of decomposition, particularly aluminum.

E. Clays

Clays differ in their exchange capacity per unit weight and in the energy with which adsorbed ions are held. Exchange capacities in terms of milliequivalents of strontium per g of some representative clays (38) are vermiculite, 1.36; Utah bentonite, 1.28; illite, 0.23; and kaolinite, 0.05. The percentage of water-soluble strontium in a bentonite suspension is 4 compared to 30 for kaolinite (82). Less strontium is adsorbed on illite than on bentonite and a greater uptake of strontium is observed in plants grown in illite (63).

F. Organic Matter

Additions of decomposable organic matter can reduce the uptake of strontium (40, 80, 81). A major factor is probably the increased microbial population, although adsorption to the organic matter itself is also important (56, 93, 108, 117).

G. Fixation

Fixation is a general name for processes occurring in soils that convert ions from forms available to plants into those not available. Soil culture experiments, such as those conducted with the Neubauer technique, have been used to evaluate the amount of applied fertilizer that is available for plant uptake. Neutral normal salt solutions are often used to extract the exchangeable cations, which are considered to constitute the major source of the available quantities of many nutrients. The difference between the applied amount and the available or extractable amount is usually considered fixed. Thus, fixation is an arbitrary term which depends upon the experimental conditions.

Proposed mechanisms for fixation include precipitation as slightly soluble materials, physical trapping between clay platelets and in other insoluble precipitates, and diffusion into existing crystals (89, 97, 113, 114).

Nonexchangeable amounts of strontium (82, 98, 103, 114) and cesium (82, 83, 113, 129) have been found in some soils. In other soils, no fixed strontium was found (43, 47). The magnitude of strontium fixation ranged as high as 20 per cent of fallout strontium-90 in North Carolina soil samples taken in 1958 (103). Laboratory studies showed that increasing the temperature from room temperature to 60° C tripled the amount fixed in these soils. Increasing the equilibration time from one to two weeks doubled the amount fixed at room temperature but had no effect at 60° C (98). In other laboratory studies with these soils, fixed strontium appeared to be extractable at 80° to 90° C.[1]

H. Erosion

Because most fission products are strongly adsorbed to clays, it is expected that any redistribution of surface soil will cause a similar redistribution of fallout. The strontium-90 concentration in runoff from field plots was 10-30 times the concentration in the soil and was almost entirely associated with the sediment (69). The strontium-90 concentration on cultivated watersheds, where the amount of soil erosion was known, was one third to two thirds of the concentration on watersheds where there had been no erosion (25). Slope and cropping history appeared to be related to the amount of loss.

V. PLANT RELATIONS

A. Uptake

The uptake by plants of ions from solution has been the subject of many investigations. There have been short-term experiments with adsorption periods of minutes or hours and long-term experiments with adsorption periods of days, weeks, and months. Short-term experiments are useful in studying mechanisms of the initial steps, whereas longer-term experiments elucidate over-all effects and the general distribution within the plant.

Numerous mechanisms have been proposed for the uptake of ions by plant roots. Most hypotheses state that the process of concentrating the ions within the plant root is a metabolic function. Essential in most of these mechanisms is a biological compound that serves as a carrier (20). Evidence has been obtained that calcium, strontium, and barium compete for an identical carrier, whereas potassium, rubidium, and cesium compete for a different carrier (20, 21, 28). Hydrogen appears to compete with all ions (27).

In addition to the accumulation of ions within the root, there is adsorption of ions on the root. The CEC of roots can be increased by nitrogen fertilization (121). A linear correlation was observed between the CEC of different species and the uptake of strontium-90 (75). The exchange adsorption does not appear to be controlled directly by metabolism (19, 45) and has been considered by some investigators (50) to be independent of active transport.

The point of maximum uptake of strontium and iodine appears to be within a few mm of the root apex (45), and no enhanced uptake is observed with root hairs. Other work (140) indicates that the tips of barley roots absorb various ions readily but that the greatest translocation occurs from a region 30 mm above the tip.

It has been suggested that strontium can partially substitute for calcium (133, 137) and even that strontium is an essential element (141). Sixty to 70 per cent of the strontium in the plant has been found to be water-soluble, whereas 97 per cent of the cesium and only 16 per cent of the cerium-144 was water-soluble (83).

A possible error in short-term experiments is the exchange of the radioactive isotope for the stable isotope already in the plant. This is particularly true for ions of slow turnover rate, such as calcium. Some workers (73, 133) consider the first 24 hours of calcium uptake to be largely nonmetabolic exchange.

B. Translocation

Most investigators believe that translocation of ions is governed less by metabolism than by the uptake process (5). The translocation of rubidium from the root to the top has been related to the transpiration stream (29). However, no such relationship was found for calcium (7).

The upward translocation of strontium and calcium relative to that of phosphorus, sulfur, iodine, and rubidium is limited. The main path appears to be the central zone of the vascular tissue (58). The redistribution of strontium, calcium, yttrium, and other multivalent cations is much less than that observed for cesium, rubidium, and potassium (35, 131).

C. Aerial Contamination

A principal pathway of intake of fallout nuclides, immediately following deposition, is through contamination of aerial plant parts. A conclusion from studies of plant material near the Operation Hurricane test site (108) is that the radioactive contaminants were carried by airborne soil, which lodged upon the leaves, and were then partially dissolved by nocturnal dew. Most of the fission-product radioactivity associated with vegetation near the Nevada tests was in external dust (83). The retention of the particles by foliage was enhanced by mechanical trapping by hairs, glands, and stomata. Particles of less than 44 μ diameter were preferentially retained on foliage, whereas particles having diameters over 88 μ were rarely retained. Greater absorption is generally expected from contaminants in solution than from dry contaminants.[1]

It also appears that aerial contamination of plants from stratospheric fallout is important in the entry of nuclides into the food chain. The cesium-137 to strontium-90 ratio in milk in 1959 and 1960 was rather constant, although cesium uptake through roots is known to be much less than that of strontium. The conclusion was reached that cesium and strontium in the forage are largely derived from foliar deposition (49). By determining the specific activities (strontium-90/strontium) of different parts of wheat plants, other workers concluded that over 90 per cent of the strontium-90 in the grain came from current fallout in 1959 (70). About two per cent of the strontium-90 fallout during the time the heads were exposed was retained in the grain. The whole crop retained about three per cent of the deposited strontium-90 and removed only about 0.2 per cent of the strontium-90 in the soil. Ryegrass grown in flats absorbed directly 23 per cent of the current strontium-90 fallout (74). This accounted for 55 to 80 per cent of the total plant strontium-90.

Autoradiograms show that strontium enters directly through the intact epidermis of the tomato fruit (58). About four per cent of the applied strontium and two per cent of applied ruthenium is absorbed by tomatoes (46). The stage of maturity of the fruit had some effect on the amount absorbed.

The species of plant is important in the absorption of foliar-applied elements (71), partly because of the degree of waxiness of the leaves and partly because of the death of some leaves before maturity. Wheat plants absorbed 85 per cent of the applied strontium and 93 per cent of the cesium, but cabbage absorbed only five or six per cent of either of these elements.

The time of contamination in relation to the maturity of the plant is important also, especially for the relatively nonmobile elements. Less than 0.1 per cent of the applied strontium is found in wheat grain if the surface deposition occurs before head development; up to one per cent is found when the head is contaminated (71).

Iodine-131 can occur in the gaseous state, and in this form it is taken up by both the mesophyll (35 to 40 per cent) and the epidermal tissue (118). The rate depends upon the concentration. Stable iodine does not reduce iodine-131 absorption, but it does reduce translocation.

The washing effect of rain can reduce the foliar intake of strontium by a factor as large as six (71). Intake of cesium is reduced to a lesser extent. Contaminated dust is satisfactorily removed from leaves by washing with 0.1 N HCl, but spray contamination is much more difficult to remove (4). Over 50 per cent of the iodine in foliar contamination is removed by washing, 70 to 85 per cent by different adhesives, and up to 97 per cent by stripping off the upper and lower epidermis (42). Not only can washing remove surface contamination, but it can also remove ions from within the plant (55). Sodium, potassium, and manganese are readily leached; calcium, magnesium, sulfur, potassium, and strontium are moderately leached; and iron, zinc, phosphorus, and chlorine are leached with difficulty (130).

D. Plant-Base Absorption

Some British workers have given attention to the absorption of fission products from the root mats of long-established grass pastures (107). The root mat is composed of roots, basal portions of stems, and organic matter. The strontium-90 in rainfall and that washed off the plants may be held in the mat long enough for considerable absorption to occur. This pathway bypasses soil reactions and may be very important where the root mat is present.

The proposed plant-base mechanism provides a reasonable explanation for the strontium-90 concentration in pasture vegetation, which appears far too high to be accounted for from expected soil uptake and foliar absorption.

E. Distribution in Plants

Strontium tends to accumulate in the aboveground portions of plants (16, 47, 67, 78, 106, 123), particularly in the vascular tissues (77), although the root concentration increases with time (58). The greatest concentration of strontium is usually found in the older leaves (85, 94), with only about one tenth as much in the seeds (78, 94, 120). However, the seeds of a few plants, e.g., Euphorbia, accumulate strontium to a greater extent (13).

Cesium and rubidium are similar to potassium, and therefore it is expected that they are distributed more uniformly throughout the plant than strontium. About 10 per cent of the total plant cesium is found in the grain of wheat and oats (47), and other work (67) indicates that there is a slight tendency for both cesium and rubidium to accumulate in the young leaves and flowers.

In four plant species, the highest concentration of iodine was found in the roots, followed by older leaves and, finally, the younger leaves (119). Other radioactive constituents of fallout, which have not been specifically mentioned, concentrated in the roots, with little translocation to the top (47, 85, 94, 106).

F. Species Differences

The order of fission-product uptake by the different plant families is Leguminosae > Solanaceae > Compositae > Gramineae for the tops and Leguminosae > Gramineae > Compositae > Solanaceae for the roots (142). Other workers report no consistent differences between the lower and higher orders of the plant kingdom (79). The calcium and strontium content of eight legumes was about three times that found in eight grasses (138). The absorptive power of a given species for strontium is considered to be proportional to its absorptive power for calcium (17, 66).

Characteristics of the root system may be very important in determining the uptake of radioisotopes from soil. Russian thistle can absorb strontium from a soil depth greater than 3-1/2 feet (116). Since the plants of the grass family have relatively shallow root systems, they will preferentially absorb nuclides occurring near the surface rather than those placed at a greater depth. With a grass-clover mixture, it was found that both the strontium content and the strontium-to-calcium ratio were reduced 70 per cent by plowing under the surface contamination (72). More deeply rooted crops showed only small effects from this deeper placement.

Bicarbonate has differential effects on plant species, with beans taking up lesser amounts of cations than barley in the presence of bicarbonate (32). Additional interactions of plant species with rate of uptake, distribution, temperature, and other factors are probably of minor importance when considering broad differences.

VI. RADIATION EFFECTS

The severity of associated heat and blast effects from nuclear test detonations have tended to obscure radiation effects on plants. However, radiation effects may be a significant force in modifying the ecological systems after a nuclear attack.

At present, the effects of ionizing radiation have been observed for only a few hundred of the more than a million and a half different kinds of organisms. Most of these data were obtained under experimental conditions of minimum environmental stress.

A. External Radiation

The median lethal dose for flowering plants ranges from about 1,000 to 150,000 roentgen units, and the sensitivity of a particular plant may vary widely according to the particular stage in its life cycle (90). The variation in sensitivity between plants has been correlated with characteristics of the cell nucleus. Plants with low chromosome number and high nuclear volumes are the most sensitive (122).

Pine trees appear to be relatively more sensitive than other trees. At an unshielded reactor site, pines died after receiving 2,000 or more rads in an initial burst, but pines at greater distances died after accumulating about 8,000 rads; hardwood trees in the area showed little effect (90). With gamma radiation from cobalt-60, pines showed detectable effects from two roentgens per day for an average of 240 days per year over a period of nine years (122).

Several other observations have been made on irradiated trees (90). The winter dormancy is prolonged by an amount proportional to the dose received during the preceding summer—one to two weeks' delay for several hundred rads. The terminal buds are more sensitive than the lateral buds and, of the lateral buds, those farthest from the trunk are most sensitive.

Two years after a nuclear explosion at the Marshall Islands, the number of different plant species showing pathological effects and abnormalities increased with an increase in fallout (23). However, differences in edaphic factors such as soil fertility may confound these observations (39).

B. Internal Radiation

The radiation emitted by the absorbed radionuclides may also cause damage.

In greenhouse experiments, at concentrations of 5 μc of strontium-90 or 13 μc of cesium-137 per g of wheat leaves, the protein levels decreased and the carbohydrate levels increased (34). A 30- to 50-per cent decrease in yield of grain was observed at those concentrations of radioactivity. Resistance to radiation damage increased with age of the plant.

In young barley plants, phosphorus-32 radiation damage was confined to cells in zones of active division (10). The lowest specific activity level at which damage was produced corresponded to 3.2 mc of phosphorus-32 per g of phosphorus, or about 170 μc of phosphorus-32 per g of dry plant tissue.

A more complete treatment of this subject is found in the Proceedings of the First National Symposium on Radioecology (115).

VII. SOIL-PLANT RELATIONS

A. Basic Aspects

The soil and plant components of the soil-plant system are individually complex, as is evident from the preceding sections. The combination of these two components increases the difficulties of understanding and generalization. Both systems, independently and together, are dynamic. Plant growth requires the continuous net removal of ions from the soil into the plant. On the other hand, changes in moisture and the removal of ions by the plants continually change the quantity of the ions available to the plant.

B. Competing and Carrier Cations

The kinds and amounts of the complementary ions affect the availability of a given ion (48, 87). Two types of processes can be distinguished: the exchange reactions governing the distribution of ions between clay and solution (37, 48) and the competitive effects during the course of ion absorption by plants (17, 20, 21, 27, 28). Since several cations compete for the same carrier site, increasing the concentration of one should decrease the uptake of others in the same group. Examination of this hypothesis in greenhouse and field experiments has shown this to be true within certain ranges.

Increasing the calcium concentration in nutrient solution from zero to two milliequivalents per liter reduces the uptake of strontium (43). Further increases in calcium reduce strontium uptake only slightly. A fourfold reduction of strontium uptake in field experiments appears to be the maximum that can be achieved by the addition of calcium to acid, low-calcium soils. Even smaller reduction occurs in soils richer in calcium.

The addition of stable strontium has little effect on radioactive strontium uptake because of the similarity of strontium to calcium and the thousandfold greater abundance of calcium in soils (68, 127). In one experiment, no effect of stable strontium was observed (43) and a slight increase in strontium-90 uptake was found in another experiment (104). It was postulated that small increments of strontium displaced some of the strontium-90 from the exchange complex into solution. It is estimated that five tons of strontium amendments per acre would be needed to reduce the strontium-90 uptake appreciably (104).

A depressing effect of potassium on plant uptake of calcium, magnesium, and strontium has been observed (47, 54, 65). Potassium treatments decreased strontium uptake 20 per cent in wheat plants (47) and 40 per cent in radish plants (54).

In a field comparison of plant concentrations of different elements with the corresponding soil concentrations (57), it was found that varying levels of calcium and magnesium brought about only slight changes in the strontium content of four pasture species. However, potassium and sodium reduced the strontium content of bluegrass as much as 34 per cent and that of redtop 51 per cent, whereas sodium additions increased strontium in Korean lespedeza.

Similar observations have been made on the uptake of cesium-137. When soil potassium is low, additions of potassium reduce the cesium uptake, but additions of stable cesium often increase cesium-137 uptake, presumably by displacement of exchangeable cesium-137 into solution (84). Rubidium, ammonium, and calcium increased cesium uptake 8, 3, and 1-1/2 times, respectively, but when carrier cesium-137 was used, practically no effects of these ions were observed (128).

C. Distribution Factors

Because of the similarity in chemical behavior between certain fission products and certain essential elements, fission-product uptake is often reported relative to the uptake of the chemically similar essential element. The "Observed Ratio" (OR) (18), or the "Distribution Factor" (DF) (66), for strontium is the ratio of strontium to calcium in plant or plant part divided by the ratio of strontium to calcium in the nutrient medium. The term "discrimination factor" is expressed in the same manner but usually applies to a single step in the various successive processes that determine the over-all relative distribution of the two elements between substrate and tissue.

In nutrient solution experiments, when only the plant discrimination processes are measured, the strontium/calcium DF is close to 1.0 (67, 105). This indicates little discrimination between strontium and calcium and is true for most of the plant parts except the roots, where DF values as high as 6.0 were observed for low-solution concentrations of strontium. The average DF values of rubidium/potassium and cesium/potassium for millet, oat, buckwheat, sweetclover, and sunflower plants were 0.85 and 0.20, respectively (67). This indicates some discrimination by the plant against rubidium and more against cesium.

Alfalfa and wheat grown on eight soils (98), wild plants and corn grown on soil in a radioactive waste disposal area (6), and beans grown on a Sassafras sandy loam with added calcium (105) had strontium/calcium DF values close to 1.0, indicating little discrimination in soil reactions. However, the DF values can vary within a given plant, ranging from 2.6 for corn flowers to 0.5 for corn grain (6).

The calculated DF will vary to some extent, depending upon the method of extracting the cations from the soil. Based on the amounts of strontium-89 and calcium-45 added to Cinebar soil, the DF for beans ranged from 0.64 to 1.2 (43). Another experiment with strontium-89 and calcium-45, using a dilute calcium chloride extract of the soil, gave an average DF of about 0.7 (111). Discrimination factors from 0.8 to 1.6 were found for strontium/calcium in eight grasses and eight legumes grown in three soils, using ammonium acetate for extraction (138). In a study of soils and vegetation in a disposal area (33), the best soil index of strontium-90 uptake by plants appeared to be concentrations of strontium-90 in the saturation extract. Others (112) have also suggested that a water extract may provide a better measure of the availability of strontium and calcium in the soil than the exchangeable fraction.

Barley, buckwheat, and cowpeas grown on an Evesboro sand gave a cesium/potassium DF of 0.02, based on the amount of radioactive cesium added to the soil and the acid-soluble potassium (66). The range was from 0.06 to 0.77 for wild plants and corn grown in a radioactive disposal area (6). Upland rice, wheat, and beans grown on a Japanese soil gave much lower DF values—0.002 to 0.003 (41). Discrimination factors of 0.02 for barium/calcium and 0.4 for rubidium/potassium were also found for the barley, buckwheat, and cowpeas grown on the Evesboro sand (66).

Discrimination in plant uptake of strontium and calcium is usually slight in pot experiments, except for roots, but apparent discrimination against either strontium or calcium can occur in the field. The strontium-90 is normally concentrated near the soil surface, or in the plow layer, the distribution of exchangeable calcium in the profile usually is nonuniform, and the root zone varies with the plant species and with soil conditions.

D. Effects of Clays and Anions

Twice as much strontium is taken up from illite clay suspension as from bentonite clay (63), which indicates that bentonite holds strontium more strongly than illite. Clays have more of an effect than just as an anion, for the aluminum concentration affected calcium uptake from calcium sulfate but not from calcium clay (64).

Anions have differential effects on various species. In tobacco, calcium uptake is nearly the same from the carbonate, sulfate, or phosphate salt, but alfalfa seems to prefer carbonate to sulfate or phosphate as a calcium source (102). Strontium uptake by bean plants in one soil was reduced 40 per cent by calcium carbonate but only 15 per cent by calcium sulfate (105). Bicarbonate in nutrient solutions (32) reduced strontium uptake by 70 per cent, rubidium by 43 per cent, ruthenium by 24 per cent, and cerium by 19 per cent. Cesium was the only ion studied in which the uptake was not adversely affected. Thus, the reduced strontium uptake from alkaline calcareous soils may be due to a bicarbonate ion effect as well as to a calcium effect.

Strontium added to soils as the sulfate, oxalate, hydroxide, fluoride, carbonate, or phosphate was one tenth as available to plants as strontium added as the chloride or nitrate (132). Also, calcium sulfate and calcium carbonate were more effective than calcium chloride in reducing strontium uptake. This indicates an effect of solubility, since all unavailable strontium salts are of low solubility. Massive doses of phosphate have reduced strontium uptake 50 per cent on alkaline soils but have given no reduction on acid soils (132). Other studies (14, 53) report conflicting results from the addition of phosphate. Hydroxyapatite and fluoroapatite are insoluble calcium phosphates that exist under alkaline conditions. The strontium analogs of these compounds are expected to be similarly insoluble (26).

VIII. MANAGEMENT

A. The Effect of Liming

The effect of calcium on strontium uptake has been discussed to some extent in two previous sections. A practical application of this is in the liming of acid soils. The uptake of strontium from different soils increased in proportion to the reciprocal of the exchangeable calcium (24, 66). No relation has been found between the total calcium or calcium carbonate content and strontium uptake (22, 30). Since soils have finite exchange capacities, overliming has little additional effect on strontium uptake. This is demonstrated by the application of lime to neutral or alkaline calcareous soils, which reduces the strontium uptake only slightly (30, 98, 105, 139). A fourfold reduction of strontium uptake by liming acid soils is generally the most to be expected (109, pp. 18-49).

The ratio of radioactive strontium to calcium in the plant is as important as the radioactive strontium concentration because deposition of radioactive strontium in animal skeletons depends on the calcium content of the diet (18). Liming tends to decrease this ratio until the exchange capacity of the soil becomes saturated with bases. Then, when the soil is no longer acid, additional lime remains undissolved and thus unavailable to the plant. Therefore, although liming serves a twofold purpose of reducing both the strontium concentration and the strontium-to-calcium ratio, the lowest values for both are usually achieved at a point that coincides with the amount of lime needed for maximum crop yields.

B. Fertilizers

In some experiments, fertilizer has increased the uptake of fission products, but in others no increase occurred (14, 47, 117, 120). Fertilizers and manures may affect the availability of the fission products, as discussed in previous sections, but it is possible that better plant growth will obscure any other effect.

C. Cultivation

Cultivation tends to increase strontium uptake by some crops (14), possibly by providing more root contact. Greater uptake of calcium results from a completely mixed calcium application than from a banded application (12). Increased uptake of strontium in the second year of a field experiment has been attributed to a more uniform distribution of strontium (14). In the case of shallow-rooted grasses, as discussed under species differences, it appears that plowing reduces the strontium-90 to calcium ratio of the new planting (72).

Plowing surface-contaminated fields to a depth of seven inches resulted in 50 per cent of the activity in the zero- to four-inch level. Both rotary tillage to four inches and two plowings resulted in >70 per cent of the activity in the top four inches (110). The uptake of fission products placed at a 60-cm depth, compared to a 30-cm depth, was as low as one thirtieth in the tops and one tenth in the seeds of oats and peas (36).

D. Moisture

Variations in the soil moisture tension were found to affect cesium uptake and the cesium-to-potassium ratio, but no effect was noted on calcium and strontium (125). Several observations (128, 129) of a tenfold greater uptake of cesium by lowland rice than by upland rice have been made. Evidence indicates that increased amounts of ammonium ion under the reduced conditions caused the greater uptake.

E. Prolonged Cropping

Little variation in strontium availability is observed over a period of years (14, 43), and the uptake of strontium per unit weight is fairly constant (86). The effect of fixation has not been studied sufficiently in the field to determine whether it is important under prolonged cropping. The uptake of cesium increased under prolonged cropping (86), probably because the potassium became depleted by crop removal.

IX. SUMMARY

Radioisotopes in fallout enter plants by three principal pathways: (1) direct absorption by the aboveground parts; (2) absorption by the stems and roots from the root mat of grass; and (3) absorption by the roots from the soil. Contaminated soil adhering to the aboveground parts of the plants may contribute to the observed uptake of fission products.

Foliar deposition and absorption depend on the surface area of the aboveground portion of the plant and the characteristics of the surface. The greater the surface area, the greater the interception per plant. Pubescence increases the retention of the fallout dust against washing and therefore the period of absorption. Many elements seem to be absorbed, some to a greater extent by this method than through the roots. Fallout particles can be washed from the leaf surface, and even small amounts of absorbed elements can be leached from the leaf.

Plant-base absorption is a relatively recent concept and its general contribution has not been adequately evaluated.

No single crop of plants has been reported to absorb from the soil as much as 10 per cent of the applied dose of fission products. There are two main reasons: (1) the soil has an affinity for the fallout nuclides because most of them are cations; and (2) the plant itself discriminates against them to a certain extent. The uptake of short-lived isotopes, such as barium-140 and iodine-131, through the roots is relatively unimportant as most of the isotope decays during the period required for it to reach the roots.

The uptake of cations by roots is probably by a carrier mechanism. Strontium and calcium compete for the same binding sites on this carrier, whereas cesium and potassium compete for another common site.

With the exception of strontium, and possibly cesium, the longer-lived fission products are taken up in relatively small amounts and therefore are not as important as strontium and cesium with respect to uptake from soils.

Because strontium is chemically similar to calcium, the strontium content of plants is often reported as a strontium-to-calcium ratio as well as an absolute amount of strontium. Both values have some importance in assessing hazards in the subsequent links of the food chain. The usual maximum uptake of strontium appears to be about one per cent of the applied dose per crop. The average DF for strontium to calcium between the soil and plant tops appears to be close to unity. This factor varies among plants and even among different parts of the same plant and by different soil extractants, but the range of variation is considered to be of little practical importance. Variations in the root zone and differences in the vertical distribution of fallout strontium-90 and calcium in the field can have greater effects.

The average maximum uptake of cesium appears to be about one tenth of one per cent of the applied dose. The cesium-to-potassium DF is small—about 0.2 for uptake in nutrient solutions and 0.02 for additions to the soil.

In general, it appears that grasses accumulate less strontium than legumes. The fruit and seeds contain less strontium than the leaves or stems because strontium tends to accumulate in the vascular tissues of the plants. In contrast with strontium, which only moves readily upward, cesium is easily translocated throughout the plant, with perhaps slightly higher accumulation in young leaves and flowers.

Strontium and cesium are retained in the soil partly by ion-exchange bonds on clay minerals and organic colloids. A part of the strontium may be synthesized into organic compounds by the microbial population. A third means of retention in the soil can involve fixation processes. A large fraction of cesium-137 appears to be fixed irreversibly. Exchangeable strontium is leached through soils at the rate of about one inch per 100 inches of leaching water. The downward movement of strontium, and probably cesium, is essentially an exchange reaction and proceeds by successive desorption-adsorption sequences.

The soil that will provide minimum uptake of fission products usually appears to be one considered ideal for maximum crop production. These requirements include high exchangeable calcium, high exchangeable potassium, high organic-matter content, and a slightly alkaline reaction.

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