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RADIONUCLIDE UPTAKE BY PLANTS

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ABSTRACT

Natural radionuclides are always present in the environment and, together with those made by man in the past thirty or more years, differentially move through geochemical and biological cycles. The general principles governing the uptake, transport, and redistribution in plants of radionuclides are reviewed. Also plant uptake of selected radioisotopes of U, Ra, Th, Sr, I, Cs, Tc, Np, Pu, Am, and Cm are discussed in some detail. These isotopes were selected because of their potential usefulness or harmfulness to man in nuclear power generation and in other contexts.

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INTRODUCTION

About 340 nuclides occur naturally in the environment. Of these about 70 are radioactive and are found mainly among the heavy elements.[1] All elements of atomic number greater than 80 possess radioactive isotopes, and all isotopes of elements heavier than number 82 are radioactive. Traces of naturally occurring radioactivity can be demonstrated in all substances, living and nonliving.

With the mid-twentieth century discovery of methods of releasing atomic energy and the increasing world-wide utilization of nuclear energy, particularly for power generation, increasing amounts of man-made radionuclides are being produced. In the fission of heavy nuclei (for example ^{235}U or ^{239}Pu) approximately 200 radioisotopes with atomic weights from 72 to 166 are produced.[2,3] Aside from fission process, radionuclides are also produced by nuclear fusion and neutron and heavy particle irradiations.

Whether it be intentional or unintentional, a certain amount of the man-made radionuclides is bound to be released into the environment. Once

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released, they are subject to cycling in the biosphere and could lead to man through various food-chain pathways. Thus, interest in the health aspects of radionuclides has increased with the development of nuclear energy, and this has caused considerable attention to the radionuclide content of foods. Uptake of radionuclides by plants is one of the important pathways of the food chain to man.

In considering the uptake by plants, only selected natural and man-made radionuclides are reviewed in this article. These radionuclides include ^{238}U, ^{226}Ra, ^{232}Th, ^{90}Sr, ^{131}I, ^{137}Cs, ^{99}Tc, and several transuranic elements (^{237}Np, ^{238}Pu, $^{239,240}Pu$, ^{241}Am, ^{242}Cm, and ^{244}Cm). They were selected on the basis of their potential significance, particularly from the standpoint of their usefulness or potential hazard to man on a long-term basis and their importance in the application of nuclear energy for power generation. Several fission products not listed here have been reviewed previously by Nishita, Romney, and Larson.[4] A number of the excluded radionuclides have applications in agricultural, medical, and biological research. These radionuclides are discussed in many references.[5-17]

This article reviews first the factors that generally influence radionuclide uptake by plants grown in contaminated soils and then the uptake of specific radionuclides.

PATHWAYS OF RADIONUCLIDE UPTAKE BY PLANTS

In order to understand the manner in which radionuclides in the environment are taken up by plants and transferred to man, it is necessary to be aware of the principal food chain pathways through which this can occur. These transport pathways may be represented diagrammatically as illustrated in Figure 1.

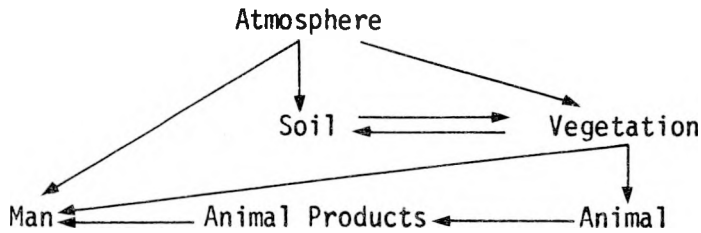


Figure 1. Principal pathways through which radionuclides transfer to man in terrestrial ecosystems.

Material deposited from the atmosphere may lodge on aerial parts of plants or enter the soil. The transfer between soil and plant may proceed in either direction. Inhalation of radionuclides is the most direct transfer to the human body. The consumption of contaminated food is the most direct route for transport through human diet. Animals grazing on contaminated vegetation constitute an indirect route. Food chain pathways, including transfer coefficients, for ^{90}Sr, ^{137}Cs, and ^{239}Pu are discussed in several references.[18-21]

Most plants consumed by man are grown on land. The complicated terrestrial ecological system of which man is a part is nourished mainly by plants. All elements essential for plant growth, except for those like carbon, hydrogen, and oxygen which may be obtained from the atmosphere, are taken up as ions through plant roots growing in soil. In general, ions of any element present in the soil will pass into the root system whether or not they are essential for plant growth. Radioisotopes of elements present in soil will pass into the root in the same manner as their nonradioactive isotopes. The amount of any element, radioactive or nonradioactive, taken up by plants is determined by a number of factors, including the physical and chemical properties of the elements and the soil, microbial activity, and the

characteristics of the plants themselves.

The transfer of a number of radionuclides from soils to plants is often very small. In such cases, direct deposition on vegetation may be the more important mode of transfer to man, since that present as surface contamination together with whatever would be taken up by plants through absorption by the aerial parts might be consumed. With certain perennial plant species, directly deposited material may also collect by entrapment in basal parts of the plant or in the superficial root mat. The absorption of radionuclides from the base of the plant or the root mat is conveniently called "plant-base" absorption to distinguish it from direct foliar uptake or root absorption from the soil proper.[22, 23]

SOIL FACTORS INFLUENCING RADIONUCLIDE UPTAKE

Soils consist of minerals, organic matter, water, and air arranged in a complicated physicochemical system that provides the mechanical foothold for plants in addition to supplying their nutritive requirements.[24] In very broad terms, vertical profile of soils from top to bottom may be separated into three zones, e.g. surface, subsoil, and parent material. Most of the life processes take place in the surface zone, which varies in depth down to an arbitrary limit of around 60-centimeter depth. The subsoil is usually less fertile for plant growth. The parent material zone generally consists of loose and partly decayed rock from which the soil was derived. In closer detail, vertical profiles through soils consist of horizontal layers (horizons) which differ in their physical and chemical characteristics. Depending on the soil type these horizons may or may not be well differentiated. An extreme case of differentiation is an indurated horizon impermeable to water and

plant roots. This may be of some significance in the downward movement of radionuclides in the profile. For example, profile analyses of a Pu-contaminated soil at Nevada Test Site have shown a substantial accumulation of Pu above indurated silica and/or lime lenses.[25] Detailed profile descriptions of different soil types and their relationship to native vegetation may be found in books on soil classification.[26-28]

The major factor governing the availability of radionuclides to plants in soils is the solubility of the nuclides in the soil matrix. This follows from the fact that plant roots, in general, absorb only the soluble species of metals. The stability of the radionuclides in the soluble forms in soil solutions determines their movement in soils as well as their rates and amounts of uptake by plants. The factors governing the solubility and the soluble forms in soils are outlined in Figure 2.

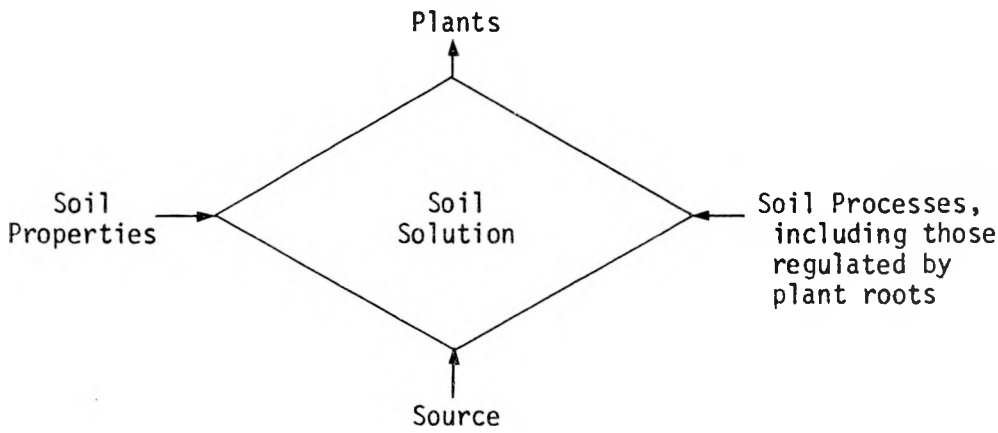


Figure 2. Factors influencing radionuclide behavior in soils.
(Adapted from Wildung, Drucker, and Au)[29]

The forms in which the radionuclides are deposited in the soil control to an appreciable extent their solubility and mobility in the soil. Depending on the source, the radionuclides could be deposited as insoluble organic or inorganic particulate forms, inorganic ionic solutions, or soluble organo-complexes. For example, the transuranics in the nuclear waste stream from a spent fuel separation facility might be mainly in the highly soluble organo-complex forms. Plutonium dioxide released with the involvement of combustion or high temperature is refractory and highly insoluble. The degree of PuO_2 insolubility depends on the oxidation temperature.[30] The PuO_2 formed at high temperatures is the most highly insoluble, while the oxide produced by oxidation at ambient temperatures produces the least insoluble form. The latter form might be considered by some to be even slightly soluble.[31] Once deposited, these materials are subject to complicated interacting soil processes, which in turn are conditioned by soil properties. The solubility of the metal-organo-complexes will be a function of their stability to substitution by major competing ions such as Ca and H[32-34] and the stability of the organic ligand to microbial decomposition.[35] In the first mechanism, a straight chemical change takes place, but in the second, a series of interacting changes of the trace metal may occur. These include indirect mechanisms resulting from metal interaction with microbial metabolites (changes in pH and Eh), direct transformations such as alkylation and alteration of the valence state through microbial oxidation or reduction, immobilization by incorporation into microbial tissues, and release of metals on decomposition of organic residues.[29,36,37] The metal ions released by microbial decomposition then may interact with various soil components. Some of these interactions may increase the availability of the trace radionuclide to plants,

whereas others may decrease it. Formation of soluble metal-organo-complexes may increase the availability. The decrease of availability may result from precipitation or fixation on clay minerals in the nonexchangeable form. The ions adsorbed on clay minerals as well as on solid phase organic matter in the exchangeable form (exchangeable to another ion) are generally considered to be available to plants, whereas the nonexchangeable form is only slowly available. In general, the soil physicochemical parameters that may influence the solubility of the radionuclides deposited in the soil include: soil solution composition, Eh, and pH; type and density of charge on soil colloids, and reactive surface area. These parameters are determined by soil properties, including the kind and amount of different elemental species, organic matter content, mineral composition (including clay minerals), degree of aeration, and microbial activity. The relative influence of these factors on the ionic uptake by plants depends on the particular radionuclide in question. These factors which relate to ion uptake by plants growing in soils are discussed in various detail in numerous references.[36,38-47]

PLANT FACTORS INFLUENCING RADIONUCLIDE UPTAKE

In conjunction with soil properties, plant characteristics play an important role in the uptake of radionuclides. One important characteristic is the root system. In grasses and many other monocotyledons, the main roots form a cluster and are approximately of the same size. The main roots give off side roots which in turn may also branch. This type of root system is called a "fibrous root system." Beets, carrot, and most other dicotyledons have a main root which grows downward perpendicular to the soil surface, and the branches arise from this main root. This type is called the "tap-root

system."[48,49] The type of root system determines the root area in contact with the soil and the depth and expanse of the soil profile penetrated. Root distribution is of considerable significance, since radioactive pollutants are usually inhomogeneously distributed in the soil.

The principal functions of roots are to provide anchor and the absorption of water and nutrients to the plant. Comprehensive discussions of transport of ions into the root are given in several references.[50-53] Roots also function in the upward transport of water and nutrients and extraneous elements that are carried in the transpiration stream and in the downward transport of carbohydrates and other photosynthetic products from the aerial parts of the plant where photosynthesis occurs.[48-50,54] Roots may also store nutrients and accumulate extraneous ions absorbed.

In the absorption of ions, a simultaneous chemical change takes place in the immediate environs of the root. The net result of excess cation absorption is the net release of H^+ from the root, while the result of net excess of anion absorption is the release of OH^- or HCO_3^-.[55,56] This phenomenon arises from the fact that electrical neutrality and cation-anion balance in plant tissues and the attendant biochemical processes must be maintained.[52,57] Aside from the reactions inside the root, the pH change of the external root environment may also have profound effects in that the solubility of ions is affected by pH and the specific forms of the pH-dependent ions may be changed.[56] This is of significance since different ionic forms of a particular element may occur not only in different proportions at different pH's, but they may also be absorbed by roots at different rates. For example, Moore[56] has pointed out that at pH 4, 98.6 percent of the phosphate is in the $H_2PO_4^-$ form and only 0.06 percent is in the divalent form, HPO_4^{2-}.

At pH 9, only 1.5 percent of the phosphate is $H_2PO_4^-$ and 98.5 percent is HPO_4^{2-}. The monovalent form is absorbed by roots more rapidly than the divalent form.[58]

In the root there is continuous uptake of ions and also continuous losses through excretion back to the surrounding medium.[59] Excretion may be a factor in redistributing radioactive pollutants in the soil profile. With contaminants like ^{137}Cs, which is readily translocated in the plant,[60,61] the ions absorbed by roots near the surface of soil profile or translocated from the aerial parts of plants may be transferred to the lower horizons of soil to which roots have penetrated. Wildung and Garland[62] showed that Pu moved downward in the root system and the root is a better vehicle for the movement of Pu in the soil profile than the downward movement with soil water. The tendency for absorption and excretion of salts changes with the growth cycle of a plant.[63] As a rule, old plants and those in the flowering or fruiting phase show increased excretion compared to young plants.[59] Injured and dead roots release salts due to breakdown of cells. Plant roots growing under normal conditions may be divided into two groups, e.g., those which do not excrete salts (grasses, most vegetables, and potato) and those which excrete (plants belonging to Papilionaceae and Polygonaceae, mustard, and hemp).[59,64] It is also well known that root exudates contain certain organic acids and other compounds which contribute to the reducing capacity of plant roots.[65] Absorption by roots of some inorganic nutrients is dependent upon this reducing capacity. For example, there is an obligatory need for reduction of iron in soils before it can be taken up efficiently by plant roots.[66] Among these organic root exudates are powerful chelating compounds which are beneficial in the uptake of metal ions, including iron.[67,68] These natural

chelating compounds in like manner should enhance plant uptake of radionuclide pollutants contaminating soils.

Associated with plant roots is the rhizosphere, which is a zone of soil in which microflora is influenced by roots.[69] This is the zone surrounding the plant roots in which the microflora are different both quantitatively and qualitatively than they are in the soil beyond the zone of influence of the root. All nutrients and extraneous ions that plants absorb from soil must pass through the rhizosphere; consequently this zone could affect the quantity and the form of the ions absorbed. Four general mechanisms[29] through which microorganisms might affect metal ion uptake by plants were listed above in the section on soil factors.

The aerial portion of the plant also has significance in that it provides surfaces on which deposition of radioactive as well as nonradioactive substances occurs. Plants vary from microscopic size to giants like the redwoods (Sequoia sempervirens), but leaves are perhaps the most important part of plants for the interception of aerial contaminants. Leaves vary widely in size and form depending on the plant species.[70] The leaf surface texture also varies widely from flat to convoluted, smooth to hairy, woolly or felt-like, and waxy to sticky with viscid exudates. Obviously, these characteristics affect the amount of aerially deposited substances retained on the surface against washing by precipitations or by deliberate laboratory treatments. Thus, with plants grown in natural environments, it is often difficult to differentiate surface contaminants from those inside the plant due to absorption by roots and aerial parts of the plant.

All plant parts absorb from their environments water, solutes, and gases.[71] Of the aerial parts, the leaves generally are the most important

absorbing organ. Foliar absorption is of particular significance in view of the reports of high radioactivity of the tops in relation to the roots of food crops exposed to fallout from nearby atomic blasts.[72]

Various aspects of foliar absorption have been comprehensively reviewed by Wittwer and Teubner,[71] Boynton,[73] Van Overbeek,[74] and Franke.[75] According to Franke,[75] the overall process of foliar absorption takes place in three stages. In the first stage, the solutes on the leaf surface penetrate the cuticle and then the cellulose wall by limited or free diffusion in the free spaces. In the second stage, these solutes are adsorbed at the surface of the plasma membrane, while in the third stage, adsorbed solutes are taken up into the cytoplasm in a process requiring metabolically derived energy. The passage of solutions through stomatal pores, if such occurs, has only the effect that the solutions enter the stomatal chambers and intercellular spaces. Since the free surfaces of the leaf mesophyll cells and the epidermal cell walls exposed to the internal air spaces are coated with cutin[76] and waxy material[77] the absorption of solutes under natural conditions is considered to take the regular course of penetration of the cuticle.[75] The factors that influence foliar absorption include contact and surface wetting, temperature, humidity, age and nutritional status of leaves, and the chemical form of the substance deposited.[73]

Foliar excretion and leaching by rainfall or heavy dews may reduce to some extent the plant content of substances taken up by plants via root and foliar absorption pathways.[59] Although mineral losses occur from aerial parts other than leaves the losses are much less than from foliage.[59] Excretion may occur in several ways. Guttation is a widespread phenomenon among plants and the presence of both organic and inorganic salts in the exuded

liquids is well established. Plants in which salt glands occur excrete excess salts by metabolically active mechanism.[78] Excretion through leaf surfaces and especially through guard cells has been shown by many investigators.[75] Among the factors influencing the rate of excretion are oxygen, temperature, age of tissue, and the kind of solute.[59] Oxygen deficiency inhibits the transport of solutes in the plant, while the temperature influences the rate of transport. In contrast to the foliar absorptive phenomenon, leaching from old leaves and upper leaf surfaces is usually much greater than from young leaves and lower surfaces. According to Tukey, Wittwer, and Tukey, Jr.,[79] the leaching of radionuclides absorbed by roots and translocated to the young foliage of herbaceous plants was as follows: ^{22}Na and ^{54}Mn, most readily; ^{45}Ca, ^{28}Mg, ^{42}K, ^{35}S, and ^{90}Sr, moderately; and $^{55\text{-}59}Fe$, ^{65}Zn, ^{32}P, and ^{36}Cl, very difficultly.

UPTAKE OF INDIVIDUAL RADIONUCLIDES BY PLANTS

Uranium

Uranium is one of a number of primordial radionuclides (radionuclides associated with formation of the earth) that occur naturally.[80] The U normally found in nature consists of three isotopes, e.g. 2.44×10^5-year ^{234}U, 7.04×10^8-year ^{235}U, and 4.47×10^9-year ^{238}U. The isotopic abundance is 0.0055, 0.720, and 99.28 percent, respectively.[81] Uranium-235, ^{238}U, and ^{232}Th are the long-lived parents of the three naturally occurring radioactive decay series.[1,80] The importance of U as nuclear fuel is well known.

Uranium is ubiquitous in nature and thus finds its way to human beings through the various food chains. The average dietary intake of U in three

large U.S. cities has been estimated as about 1.3 μg/day.[82] The maximum permissible total body burden is 0.4-0.5 μCi, depending on the isotope.[83] The critical organ for natural U is kidney for which the maximum permissible burden is 0.005 μCi.

Uranium is released in nature through weathering of rocks or ore deposits. In the weathering process, bicarbonate ions are significant since they cause the formation of readily soluble uranyl complexes of the type $UO_2(HCO_3)_3^-$ [84] and $UO_2(CO_3)_3^{4-}$. [85] These soluble complexes migrate with water. The extent of migration depends on their stability in solution. Gera[84] has outlined the various processes and chemical reactions that cause the removal of soluble U from solution. In soils, the major part of U is in the difficultly soluble forms. Soils normally contain trace amounts of U (Table 1). The relatively high U content of soils from Niue Island and the Colorado Plateau are due to the presence of U minerals and/or to their proximity to deposits of U minerals.

Plant content of natural U has been reported by a number of investigators.[80,82,86-96] Cannon,[88] for example, found that the leaves of many plant species rooted in ore deposits of Colorado Plateau contained 2 to 100 ppm U. The normal background content of plants collected from barren sandstone and shale was < 1 ppm. Hoffman[86,87] found < 0.14 ppm U in the plants that he examined. In general, the U concentration in plants is of the order of a few ppm.

Uranium uptake by plants is influenced by several factors. Generally, the U concentration in the plant is higher in soils with higher U content.[97] The concentration in the plant also depends on the stage of growth. Depending on plant species, soil U concentration above a very low level is retarding to growth or even toxic.[88,95] Zhukov and Zudilkin,[95] for example, found

significant inhibition of growth of wheat plants when the soil U concentration was raised from 10 ppm to 50 ppm. The influence of U concentration depended on the type of soil and fertilizers applied.[95] Cannon[88] has reviewed the effects of excessive amounts of U on plant growth. Among the effects are suppression of seed germination, yellowing and deformation of roots, leaves and inflorescence, inhibition of root development, and lowering of yield. In the beneficial range of concentration, growth and maturing of the plant are accelerated. Cannon[88] has also provided a list of uranium-tolerant plants growing on the Colorado Plateau.

A number of reports give concentrations of U in plants and/or in soils, but only few have quantified the accumulation in plants in relation to that in soils. This relationship is generally expressed as the ratio of plant content to soil content. Prister[98] calculated this ratio in two ways and designated them as A_x and K_h as follows:

$$A_x = \frac{\text{U content in plant (mg/kg ash)}}{\text{Total U content in soil (mg/kg soil)}}$$

$$K_h = \frac{\text{U content in plant (}\mu\text{g/kg dry matter)}}{\text{Soluble U content in soil (}\mu\text{g/kg dry soil)}}$$

Prister's[98] A_x values ranged from 0.3 to 1.4 and 0.3 to 1.3 for seeds and stems, respectively, of oats, wheat, millet, and peas. The K_h values ranged from 0.7 to 2.3 and 1.5 to 4.2 for seeds and stems, respectively. The lower A_x values as compared to the K_h values were attributed to the fact that major fractions of the soil U were in the difficultly soluble form. The soluble form comprised < 3 percent of the total U content (1.8 ppm) of the soil. The

plant ash to dry soil concentration ratios (CR's) calculated from the data of Yamamoto, Masuda, and Onishi[93] for leaves of vegetable crops ranged from 0 to 0.29 for soils containing 1.10 to 4.00 ppm U. Bondietti and Sweeton[99] calculated their CR values based on the 8 M acid extractable soil concentration of U and obtained ratios from 0.01 to 0.02 for snap beans, millet, soybeans, and tomatoes. These results indicate that the CR depends not only on plant species and parts, but also on the soil concentration basis on which they are calculated.

Radium

Radium-226 (1600 year half-life) is a daughter product of the ^{238}U decay series. In radioactive equilibrium, the Ra/U ratio is 3.6×10^{-7}.[84] Radium is the most hazardous nuclide for exposure by ingestion in the natural radioactive series from the radiological point of view.[84] The average daily intake of ^{226}Ra for the U.S. population is probably in the range of 1-3 pCi.[104-108] About 90 percent of this intake is from food and 10 percent is from drinking water.[109] The maximum permissible burden in total body is 0.2 μCi.[83] Bone is the critical organ for which maximum permissible burden is 0.1 μCi.

Radium content of soils of various geographic regions of the world has been reviewed by several investigators.[80,84,85,106] Table 2 gives typical concentrations of ^{226}Ra in soils as presented by Gera.[84] It shows the overall concentration range to be 0.1 - 3.8 pCi/g soil.

Radium, like Ca, Sr, and Ba, is an alkaline earth element. It behaves somewhat like them in soils, although quantitative differences do exist. Hansen, Vidal and Stout[110] have shown that the relative sorption affinity of these elements on soil is Ra > Ba > Ca. Mistry[111] determined the Ra/Ca and

Ra/Sr soil solution factor according to the method of Russell, Schofield and Newbould[112] and found that the degree of retention of Ra by soil surfaces is much higher than Ca or Sr. Geochemically Ra reacts more like Ba than the other alkaline earth elements, as might be expected from the relative sorption affinity series of alkaline earth metals.[84] In nature, Ra occurs in a very dispersed state and no minerals are known.[84] Its solubility increases at low and high pH values. It is influenced by redox potential less than U and shows greater stability in ground water solutions of low redox potential.[113]

The ^{226}Ra absorbed by plants varies over a wide range. Table 3 shows that its concentration in plant materials may range from < 1 to 2730 pCi/g fresh weight. The highest ^{226}Ra concentration that has been encountered is in Brazil nut.[114] In any specific plant, it usually accumulates in the roots. Plants grown in solution culture have CR's of roots usually 2 to 3 orders of magnitude greater than those of shoots.[111]

Thorium

Thorium is three to four times more abundant in the earth's crust than U[119-121] and may ultimately become an important source of nuclear energy as methods are developed for converting the ^{232}Th to ^{233}U in breeder reactors.[120, 121] Thorium-232 (1.4×10^{10} year half-life) occurs in nature almost entirely as a single nuclear species. It is the parent of one of the three naturally occurring radioactive decay series.[1,80] The maximum permissible total body burden of ^{232}Th for humans is 0.3 μCi[83] with the critical organ being bone for which the maximum permissible burden is 0.04 μCi.

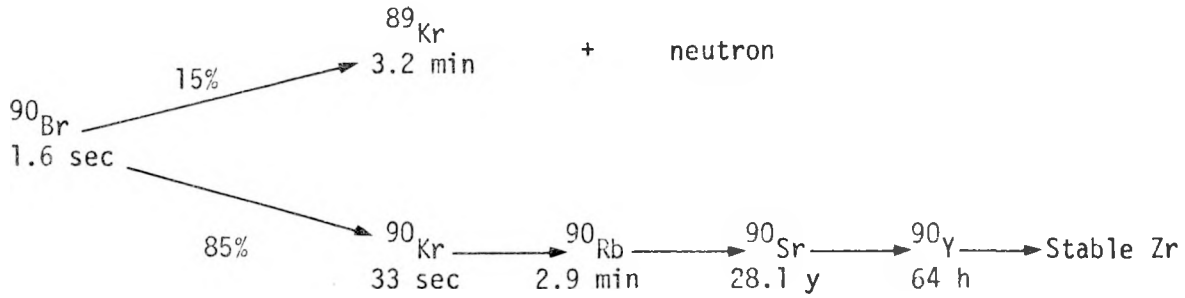
The content of naturally occurring ^{232}Th in soils varies with geographic location. Moore's[122] measurements of 5 soils in the United States show a

concentration range from 33 to 56 μg Th/g soil. Gorski and Zymslowska[123] reported 9.3 μg Th/g at a depth of 10 cm and 8.6 at 50 cm in one Polish soil, and 11 μg Th/g at 5 - 10 cm and 12 at 27 - 33 cm in another soil. Soils of the Russian plain and the Crimea range from 2.3 to 14 μg Th/g.[85] According to Mitchell,[124] the Th content of Scottish soils may range up to 10^3 μg Th/g.

Although ^{232}Th is present in soils, its uptake by plants is generally very small. Mayneord, Turner, and Radley[125] reported that of the number of human, animal, and plant tissues that have been analyzed, no evidence has yet been found for the presence of ^{232}Th in such materials. The ^{228}Th that was found in these materials is presumed to originate from the decay of ^{228}Ra (1.9 year half-life) rather than by metabolic absorption of the element Th. This was supported partly by the fact that Ra is taken up more readily than Th by plants. Later work, however, showed Th may be taken up by plants. In a solution culture experiment using ^{230}Th, D'Souza and Mistry[126] found that Th accumulation in bean plants occurs predominantly in roots and only a very small amount of it is translocated to shoots. Bondietti and Sweeten[99] found that ^{232}Th may be taken up by plants from soils. Based on 8 M acid extractable soil concentration of ^{232}Th, they found CR's from 0.0001 to 0.007 for snap beans, millet, soybeans, and tomatoes.

Strontium

Radiostrontium is perhaps best known of the fission products. It is formed by the following nuclear decay reactions:[127]



The fission yield (the proportion of nuclear fissions yielding a particular product) of ^{90}Sr in thermal neutron fission of ^{235}U and ^{239}Pu is 5.9 and 2.12 percent, respectively.[81] The significance of ^{90}Sr arises from the fact that it has relatively high yield, moderately long half-life, and enters the food chain relatively readily. Langham[18] has discussed the potential hazard of biospheric contamination with ^{90}Sr. In man, the maximum permissible total body burden is 20 μCi.[83] The critical organ is bone for which the maximum permissible concentration is 2 μCi.

Radiostrontium (^{90}Sr and ^{89}Sr) reacts in the same manner as the stable Sr normally present in biological systems, and in a manner generally similar to that of Ca to which it is chemically related.[128] The transfer of ^{90}Sr through food chains is largely determined by the amount of Ca that is present. For this reason, concentration of ^{90}Sr in biological materials is frequently expressed as picocuries of ^{90}Sr per gram of Ca. This ratio, which is called "strontium unit" (SU), is useful in following ^{90}Sr from one biological step to the next, but it may be meaningless when applied to soil under normal environmental conditions.[1] This follows from the fact that ^{90}Sr is not uniformly mixed in the soil, and the plant roots are not exposed to uniform $^{90}Sr/Ca$ ratio. When the ^{90}Sr is uniformly distributed in the soil or in the

water culture, the rate of transfer to the plant might be expressed as "observed ratio" (OR).[128-130]

$$OR = \frac{\text{Sr/Ca ratio in plant}}{\text{Sr/Ca ratio in medium}}$$

The OR (plant/solution) of plants grown in water culture is close to 1.[128] This is generally true also when plants are grown in soil in which the Sr is uniformly distributed.[4,130,131]

In the absorption of ^{90}Sr via the roots, the quantity of readily available Ca (Ca present in soil solution together with readily exchangeable Ca) in the soil is the most important factor in determining the extent of ^{90}Sr absorbed. Numerous investigators have shown the effect of Ca on ^{90}Sr uptake.[132-146] In general, the absorption of ^{90}Sr depends on soil type and is the greatest from soils low in available Ca. The addition of lime or gypsum to soils low in available Ca reduces the plant uptake of ^{90}Sr. Table 4 gives an example of this effect in Sassafras sandy loam, an acidic soil very low in exchangeable Ca. The reduction of ^{90}Sr absorption is largely a dilution effect. Wide differences exist in the ability of plant species to absorb ^{90}Sr, but it has been established that this characteristic correlated with the ability of the plants to absorb Ca.[128] The ability of different crops to increase Ca concentration in the plant with liming has been shown by Bender and Eisenmenger.[147]

The addition of lime or gypsum to a soil has a large effect only in soils of low Ca status. When soil contains adequate Ca for plant growth and the cation exchange capacity is largely saturated with Ca the addition of Ca has little or no effect. In common agricultural practice, the application of lime to acid soils can be effective in reducing ^{90}Sr absorption by plants, but it generally is ineffective with neutral or alkaline soils. Another point of consideration is that the dilution effect of liming often is a

reduction in the $^{90}Sr/Ca$ ratio in the plant, and not an absolute decrease in the ^{90}Sr content.[148] This effect is of value, since in the transfer of radiostrontium in the food chain, the significant factor is the ratio. The reduction of $^{90}Sr/Ca$ ratio in the plant is usually less than one-third of its value without liming.[143]

An increase of Ca supply is only one of several effects of liming of acid soils, and a simple relationship between Ca supply and radiostrontium content of plants is not always evident. Among the other factors of importance are the change of pH and improved plant growth due to improved nutritional status, and the reduction of toxic concentrations of some metal ions such as Al and Mn. Aside from the effects of liming, there are also other soil factors that influence the uptake of Sr and Ca by plants. These factors include clay and organic matter content, electrolyte concentration other than Ca (fertilizers), and moisture content.[4,134,143,146,148] Clays have some influence in that they, together with organic matter, determine the ion exchange properties of soils, which in turn influence the uptake of Sr and Ca by plants.[149-154] There are some indications that a small amount of ^{90}Sr becomes less available with time,[155-158] but the effect of this change on ^{90}Sr absorption by plants is considered to be too small to be of any significance in assessing ^{90}Sr transfer through food chains. The addition of organic matter in large quantities to soil lowers the radiostrontium concentration in the plant.[134,159,160] This effect appeared to be due to the upsurge of microbial population, which reduced the available radiostrontium by absorption, and to the change of K, Mg, and Ca status of the soil. The application of fertilizers might increase or decrease the radiostrontium uptake by plants.[134,137,143,146,161,162] In general, the effect of organic matter

and fertilizers under normal agricultural practices has only small effect on the radiostrontium to Ca ratio in plants, since any influence that these factors might have on radiostrontium uptake would have comparable effect on Ca uptake. Small application of stable Sr to ^{90}Sr-contaminated soil increases the ^{90}Sr uptake by plants as a result of exchange displacement of ^{90}Sr into the soil solution where it is more readily available to the plant.[163] Large application of stable Sr can reduce the plant uptake of ^{90}Sr by isotopic dilution.

Three modes of ^{90}Sr uptake from direct contamination of the above-ground parts of the plant have been recognized, e.g., foliar, floral, and plant-base absorption.[23,128,148,164] Foliar entry and distribution of ions in the plant have been reviewed by Biddulph.[164] Foliar absorption of ^{89}Sr under laboratory conditions was shown by Middleton[61,165,166] for wheat, potatoes, beans, cabbage, sugar beets and swedes. Absorption into the leaves is relatively slow, and the superficial material can be readily lost, especially by rain.[166] Absorption by the plant requires that the fission products be soluble. Thus absorption is likely to be influenced by volume of an aqueous deposit, efficiency of wetting of surface, atmospheric humidity, and dew formation. Translocation of the absorbed ^{89}Sr was only slight compared to ^{137}Cs. Floral absorption, which is the entry into inflorescences, was noticed from the fact that the ratio of ^{90}Sr to Ca in grain and flour usually exceeded that in vegetables.[167-169] Plant-base absorption is of some significance in that ^{90}Sr absorbed from leaf axils, crown of plants, or root mats is relatively undiluted with Ca as compared to absorption from the soil.[148,167] The relative significance of the various routes of absorption by the above-ground parts depends on the growth form of plants.[23] Foliar absorption is of

practical importance with grain crops only, whereas plant-base absorption is important in perennial pastures. The relative importance of absorption from direct contamination and absorption from soil has been discussed in several references.[23,148]

The difference of ^{90}Sr uptake by different plant species mentioned above has been shown by many investigators.[132,134-136,139,145,146] Aside from the biological peculiarities of plants, soil type appears to have an influence, for example, the data of Gulyakin and Yudintseva[134] show that for plants grown in clayey sand, the relative extent of ^{90}Sr uptake was clover > pea > timothy grass > oats, whereas for those grown in heavy loam, the order was clover > oats > pea > timothy grass. The distribution of radiostrontium among the organs and tissues also varies with plant species.[132,134-136,139,146] In the aerial parts, the ^{90}Sr accumulation tends to be the greatest in the leaves or stems. It is appreciably greater, for example, in the stem than in the leaves of diploid buckwheat, but it is vice versa in tetraploid buckwheat.[135] In any case, it is usually the least in the seeds or the grains of plants. These differences between different organs and tissues are explained by a lower mobility of Sr than of Ca inside the plant.[135,170-172]

The plant accumulation of long-life radionuclides (such as ^{90}Sr) from fallout under field conditions can be described by the following formula.[173]

$$C = \underline{a}\, D_p + \underline{b}\, D_t$$

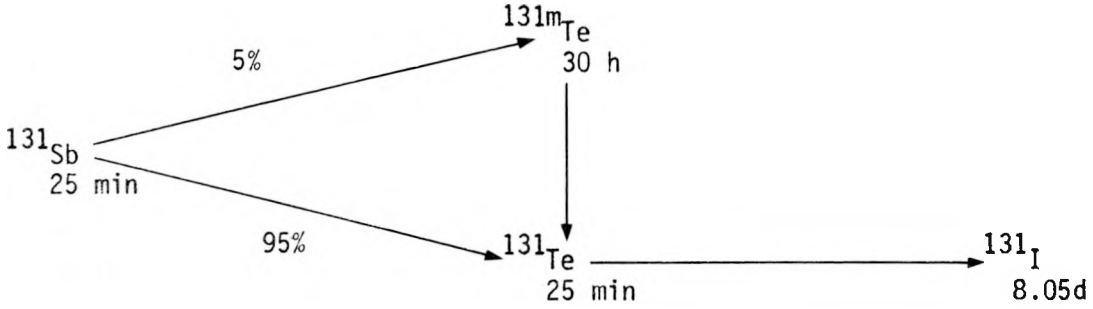
where C is the ^{90}Sr in plant, D_p is the deposition rate during the life time of the plant, D_t is the total cumulative deposition in the soil, and $\underline{a}$ and $\underline{b}$ are proportionally factors. The value of $\underline{a}$ depends on the uptake by the above-ground parts, and can be affected by atmospheric conditions,

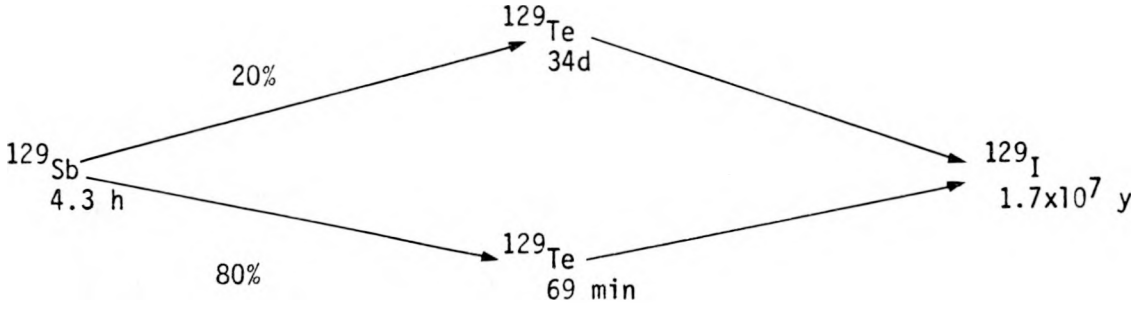
type of vegetation, plant species, etc. The value $\underline{b}$ depends on soil properties and indicates the "availability" of ^{90}Sr to plants. Thus, $\underline{a}\ D_p$ represents the fallout ^{90}Sr retained on the above-ground plant parts, while $\underline{b}\ D_t$ represents the ^{90}Sr absorbed from the soil through the plant roots. Some cautions in the use of this equation have been discussed by Russell.[173] On the basis of this eqution, Russell[173] in 1958 estimated the mean value of ^{90}Sr per gram of Ca in plants to be 1.1 pCi when the mean deposition level was 1 mc/km^2 in the United Kingdom in 1957. Depending on soil type, the ^{90}Sr/Ca ratio in plants is likely to be in the range of 1 to 10 pCi ^{90}Sr/g Ca when the soil level is 1 mc/km^2.[128]

Iodine

Eleven radioisotopes of I are formed in nuclear fission of uranium, but only four (8.05-day ^{131}I, 2.3-hour ^{132}I, 20.8-hour ^{133}I, and 6.7-hour ^{135}I) need be considered from the viewpoint of contamination of food soon after a nuclear detonation or accident.[174] Within one or two weeks only ^{131}I remains significant. The accompanying long-lived ^{129}I (1.59×10^7 year half-life) is considered to be an insignificant source of ingestion hazard because of its low activity.[174] However, because of its extremely long half-life, it has the potential for accumulation in the environment from long-term, chronic releases from nuclear facilities.[175] Russell and Hahn[176] have estimated that by the year 2000 about 10^4 Ci of ^{129}I will have been produced by nuclear power with perhaps one percent of it released to the environment. The concentration of radioiodine in the small volume of the thyroid is the primary hazard. The maximum permissible burden in the thyroid is 0.7 μCi and 3 μCi for ^{131}I and ^{129}I, respectively.

Iodine-129 and ^{131}I are decay products of ^{129}Te and ^{131}Te, respectively.[127]





The fission yield from ^{235}U is 3.1 and 0.9 percent for ^{131}I and ^{129}I, respectively.[81]

The radioiodine uptake by plants from soils is influenced by several factors as with any element. It has been shown with stable I that (a) uptake increased with increasing concentration in the substrate, (b) uptake increased when soil pH was reduced down to about 4 from higher pH, and (c) relative order of uptake by plant species was bean > tomato > barley > Russian thistle.[177] The CR's (μCi/g dry plant / μCi/g dry soil) of radioiodine ranged from 0.1 for barley leaves to nearly 2 for radish leaves.[178] The radioiodine uptake is greater, the greater the stable I content of the soil.[178]

The plant absorption of short-lived radioisotopes of I, including ^{131}I,

is not considered to be a serious problem in their transport to man. This follows from the following facts. Due to fixation of radioiodine contaminant on soil organic matter,[179] it is held mostly in the top few centimeters of the soil even against large leaching by water. By the time the radioiodine can reach the plant root zone, be absorbed into the edible portion of plants, and go through the food chain pathway to man, it is of little consequence as a radioactive entity because of radioactive decay.[1,174,180] Furthermore, because of the very small translocation (less than 5 percent) of the root-absorbed ^{131}I to the aerial parts of plants, the contamination of the aerial parts through the roots by fallout ^{131}I has been shown to be of little importance.[180]

The extent of direct foliar contamination and the state of I deposited are the major factors that control the entry of ^{131}I into food chains. Iodine-131 in fallout may be present as vapor, in aerosols, or attached to particles of different types.[134,181-188] For example, Perkins[186] has shown that a large and varying fraction (10-90 percent) of the radioiodine in fallout resulting from nuclear detonations is in the gaseous form. The chemical state of radioiodine in the particulate material in fallout was about two-thirds in the reduced state (I_2 or I^-), about one-third in the iodate form, and less than 5 percent in the periodate form. Hungate et al.[187] found that up to about 40 percent of the total iodine vapor associated with the leaf was taken up relatively readily. The percentage of absorbed I in the leaf remained constant over an exposure range of 4 pCi to 1 nCi $^{131}I/cm^3$ of air. Foliar absorption of I was considerably less from $Na^{131}I$ solution than from iodine vapor. Only a small fraction of the absorbed ^{131}I (no more than 5 percent) was translocated to parts other than the exposed leaf. The very

slow translocation of absorbed I has been observed also by other investigators.[177,180] In field studies in which plants were exposed to ^{131}I released by burning reactor fuel elements, Hungate et al.[187] found that 90 percent or more of the deposit remained on the surfaces of leaves and over half of this was readily removed by washing with water.

The amount of radioiodine that is transferred in the food chains is the amount of deposition minus the amount of loss before its transfer along the food chains. Thus, the loss of I from plants is as important as its deposition on plants. The effective half-life on plants, which is the cumulative effect of both radioactive decay and all other loss processes, has been reviewed by Thompson,[189] and Chamberlain and Chadwick,[190] and McFarlane and Mason.[180] The reported values of effective half-life on plants, which range from 3.5 to 6.5 days[180] are less than the radioactive half-life (8.05 days) of ^{131}I.

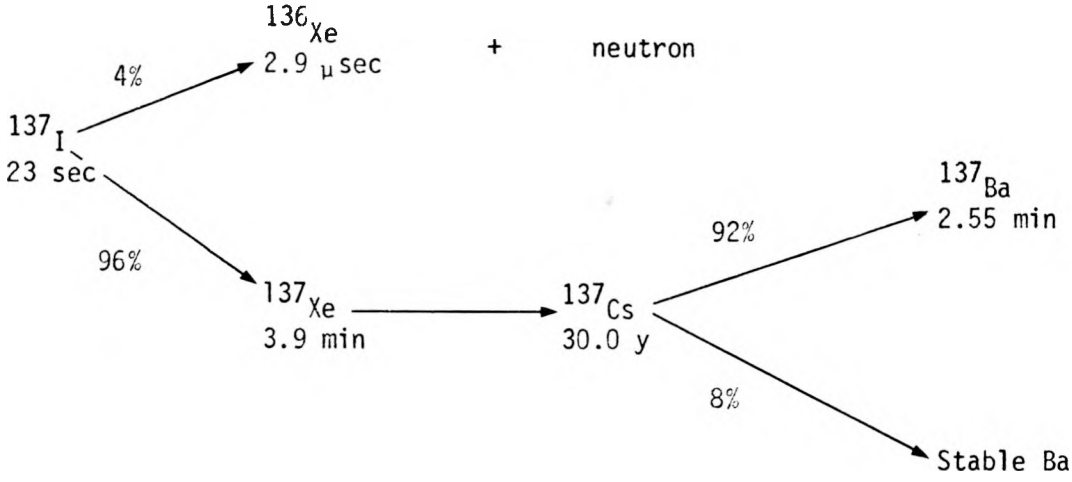
Taking all factors into consideration, ^{131}I is only of short-term hazard under special conditions. Iodine-129 in the biosphere as a whole is too low in concentration to be of hazard on a short-term basis, but on a long-term basis, it may be of potential hazard in the environs of nuclear fuel reprocessing and waste disposal sites.

Cesium

Although ^{134}Cs (2.07-year half-life) and ^{136}Cs (12.9-day half-life) have been detected in the biosphere, only ^{137}Cs (30.1-year half-life) contributes appreciably to the radiation dose to man.[191] Thus, only ^{137}Cs is considered here. The potential hazard of biospheric contamination with ^{137}Cs has been discussed by Langham and Anderson.[20] Cesium-137 is distributed mainly in

soft tissues of human body and is considered primarily as a source of genetic injury.[192] The maximum permissible total body burden of ^{137}Cs is 30 μCi.[83]

The fission product decay chain that produces ^{137}Cs is as follows:[127]



The fission yield in the thermal neutron fission of ^{235}U and ^{239}Pu is 6.23 and 6.5 percent, respectively.[81]

Cesium-137, like ^{90}Sr, can be absorbed by plants by direct contamination and from the soil. It, however, is considerably less readily absorbed from soil by plants than ^{90}Sr.[60,193,194] This difference between ^{137}Cs and ^{90}Sr is strongly dependent on soil type.[195] Among the various soil types, ^{137}Cs is taken up by plants considerably more readily from organic and tropical lateritic soils than from mineral soils of temperate climatic regions. The uptake of ^{137}Cs from freshly contaminated mineral soils is not more than one tenth of the extent of ^{90}Sr.[128,135,148,196-198] Also, the availability of ^{137}Cs relative to ^{90}Sr decreases with passage of time after they have been added to the soil.[199,200] After three years, the quantity of ^{137}Cs absorbed is no more than one twenty-fifth of that of ^{90}Sr.[128,199,200] At least, part of this effect appears to be due to increasing fixation of ^{137}Cs with time.[201-203]

Several mechanisms of fixation are thought to be involved.[198,204] One mechanism is "interlayer" fixation, which involves the diffusion of ions to interlayer sites where they are entrapped on the collapse of the clay lattice.[205-207] Another mechanism is "edge" fixation, which is due to the high affinity of exchange sites at the edges of the interlayer spaces.[208-210] This can occur in both expanded and unexpanded clay lattice structures. Other mechanisms that are more or less interrelated to these have been considered also.[135,201,211-216] The principal factors that influence the fixation of ^{137}Cs are the type of clay mineral and the species of the competing ion (K, NH_4, Rb, Mg, Ca). The relative effectiveness of the competing ions depends on the clay mineral type.[209,213,215] Of the clay minerals, illites and vermiculites in general fix ^{137}Cs particularly strongly while montmorillonite and kaolinite do so to a much lesser extent.[198,210,213,215,217-219] The effect of mineral type on the plant uptake of ^{137}Cs has been demonstrated by several investigators.[203,220] Cesium-137 absorption by plants was greatest from kaolinitic soils and least from illitic soils.

The absorption of ^{137}Cs by plants is influenced strongly by K in two ways, e.g., its effect on the structural condition of the micaceous minerals as discussed above and its antagonistic effect on ^{137}Cs absorption by plant roots. The latter effect has been demonstrated by solution culture experiments.[221] Cesium-137 uptake by plants from soils decreases with increasing concentration of exchangeable K.[60,197,212,221-229] It is increased when soils are depleted of exchangeable K by continued cropping.[197,227] The application of K to Cs137-contaminated soil low in exchangeable K decreases the absorption of ^{137}Cs by plants, but it has only slight effect on soils already high in exchangeable K.[135,197,212,222,223,225,228] It is in the soil that

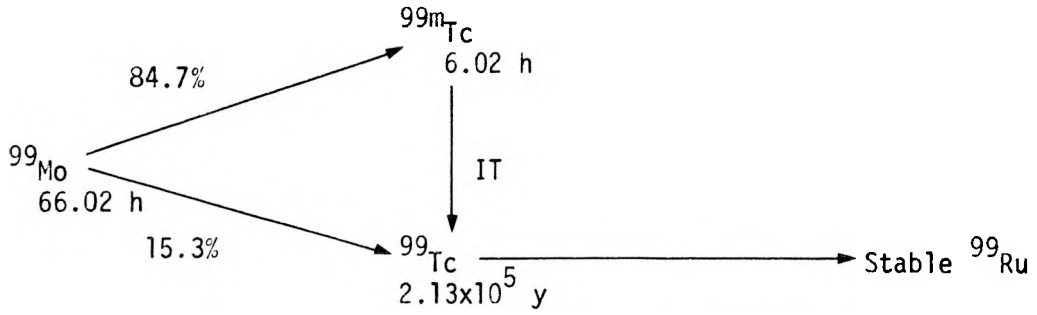
has relatively low capacity for fixing ^{137}Cs that the variation in exchangeable K has the greatest effect.[198] Aside from K, other ions may influence ^{137}Cs uptake by plants. Small applications of stable Cs to ^{137}Cs-contaminated soil cause a greatly increased uptake of ^{137}Cs by plants, presumably by carrier effect.[221] Large applications of stable Cs reduce ^{137}Cs uptake by isotopic dilution. Calcium, Mg^{++}, and NH_4^+ may also influence ^{137}Cs uptake, but their influence is generally lower than of K.[195,198,230-233] The ^{137}Cs fixed (nonexchangeable ^{137}Cs) in the soil is slowly, but definitely taken up by plants.[197,202]

Cesium-137 may be absorbed by plants by foliar, floral, or plant-base absorption.[61,165,166] It differs from ^{90}Sr in that the absorbed ions are more readily translocated within certain crop plants.[166,234-237] This has been demonstrated in such crop plants as wheat, cabbage, potato, sugar beet, pea, and beans. In three genera of woody shrubs (Ceanothus, Adenostoma, and and Quercus), however, foliarly applied ^{137}Cs was no more mobile than Sr.[238] The low mobility of foliarly applied ^{137}Cs in these xerophytic shrubs was later ascribed to adsorption on or within the leaf cuticle.[237] The ready translocation of ^{137}Cs within plants and its accumulation in the newly developing parts of the plant, in seeds, and in storage organs is generally similar to that of K.[166,239] Because of the similarity of behavior of ^{137}Cs and K the plant uptake data are often reported as $^{137}Cs/^{40}K$ or Cs^{137}/g K ratios.[240] The ready translocation of ^{137}Cs provides a mechanism whereby its distribution in the soil profile may be changed. This follows from the fact that ^{137}Cs absorbed by the above-ground parts and translocated to the roots could be released by excretion, injury, or death in different horizons of the soil profile.

The entry of ^{137}Cs from nuclear fallout debris into food and man was demonstrated to be real.[241] In 1957, Anderson et al.[240] presented the ^{137}Cs content of foodstuffs and estimated that the amount of ^{137}Cs in the population of United States averaged 0.006 microcurie. The average radiation dose received from ^{137}Cs was one-twentieth of that received from natural radiopotassium and 1 percent of the average total dose from all natural sources. Langham[242] estimated that ^{137}Cs burden of United States population at 0.01 microcurie from nuclear weapons test fallout through October 1958 and that the potential genetic hazard to the population will not be more than 1 to 2 percent of the natural radiation. The ^{137}Cs concentration in man rose to a maximum of about 180 pCi/g K in the second half of 1964 in the Northern Hemisphere, between 30^{o} and 60^{o} North Latitude.[243] The fallout ^{137}Cs in man continued to increase for about a year and a half after the cessation of atmospheric tests of nuclear weapons by USSR and US in the beginning of 1963.

Technetium

Technetium is produced by spontaneous fission of ^{238}U in nature[244] and by the slow neutron fission of ^{235}U, ^{232}Th, and ^{239}Pu in nuclear reactors either directly or via the mother isotopes of molybdenum.[245] During the fission of ^{235}U in a reactor, the yield of the various Tc isotopes is approximately 1 to 6 percent. All isotopes of Tc are radioactive. The main isotope, ^{99}Tc (6.06 percent yield from ^{235}U), is formed in one of the highest yields during the fission of other fissionable materials also.[245] From molybdenum, ^{99}Tc is formed as follows:[127]



A recent interest in ^{99}Tc stems from a transport analysis of radionuclides from a hypothetical arid storage site of high-level radioactive wastes of nuclear industry.[246] This analysis showed that ^{99}Tc, ^{3}H, ^{129}I, ^{237}Np, and ^{79}Se were the five radionuclides contributing the greatest potential dose to man from a release event from high-level waste repository. Of these radionuclides, ^{99}Tc was thought to provide the greatest potential dose to man. The maximum permissible burden of ^{99}Tc in total body is 200 μCi.[83]

The total worldwide inventory of ^{99}Tc from atmospheric nuclear weapons testing is estimated to be 0.05 mCi as of 1962.[247] Soil environmental levels resulting from fallout are estimated to be 10^{-6} to 10^{-8} μg/g.[248] In view of the high fission yield, large quantities can be expected in radioactive wastes of nuclear industry. Kotegov, Pavlov, and Shvedov[249] estimated that in excess of 170,000 Ci of Tc will have been accumulated worldwide during the period of 1963 to 1980.

The most stable and characteristic oxidation state of Tc in slightly acid, neutral, or basic aqueous solution in equilibrium with the atmosphere is the pertechnetate ion (TcO_4^{-1}) in which Tc is in the heptavalent state.[249,250] Another stable oxidized form of Tc is the tetravalent state. It, however, is oxidized to Tc (VII) by atmospheric oxygen.[251] Thus, Tc (VII) is considered

to be the most likely form under oxidative conditions. Fallout Tc that enters the surface soil in natural environment is most likely to be in the Tc (VII) form. Several investigators have examined the behavior of pertechnetate in soils. Wildung et al,[252] using a batch equilibrium technique, found the distribution coefficient (K_d = (solute sorbed per gram of soil)/(solute per milliliter of equilibrating solution)) for pertechnetate form of ^{99}Tc to range from 0.007 to 2.8 in 22 different soils. Routson, Jansen, and Robinson[253] measured K_d for pertechnetate as a function of HCO_3^- ion concentration (0.002 to 0.200 M) for a South Carolina subsoil and concluded that within the error of measurement, no sorption of pertechnetate form of ^{99}Tc occurred in this soil. Landa, Thorvig, and Gast,[254] extracted 11 pertechnetate-contaminated soils with 0.01 M $CaCl_2$ after they had been moist incubated for 2 months. They found that 31 to 100 percent of the applied ^{99}Tc was extractable, depending on the soil type. The extractable fraction was considered to be "free" ^{99}Tc in the soils, since 0.01 M $CaCl_2$ was found to be a poor extractant of sorbed ^{99}Tc. In general, the pertechnetate form of ^{99}Tc is readily extractable from soils. The mechanism of the nonextractability of relatively small fraction of ^{99}Tc in most soils has not been clearly defined. Wildung et al[252] indicated that pertechnetate K_d values were positively correlated with soil organic carbon and negatively correlated with soil pH. Gast[255] reported that Tc was almost totally "sorbed" by several high organic matter Minnesota soils. Landa, Thorvig, and Gast,[252] however, indicated 100 percent extractability from peat. Some possible mechanisms have been suggested, but they have not been verified.[255,256]

The few plant uptake studies that have been done to date show that the apparent high solubility of pertechnetate in soils is reflected in the ready

uptake of ^{99}Tc by plants. Wildung, Garland, and Cataldo[257] found that at the 0.7 μg ^{99}Tc/g soil level, the CR of soybean ranged from 65 to 100 (stems) and 330 to 1,000 (cotyledons) and from 80 to 180 (stems) and 200 to 270 (cotyledons) at the 5.1 μg ^{99}Tc/g soil level. Some evidences of toxicity on growth were observed on these plants. Under the intensive cropping technique (split-root system) that was used most of the ^{99}Tc was removed from the low level of application. The calculated quantities of ^{99}Tc remaining in the soil after 10 days of cropping amounted to < 5 percent of that applied. Landa, Thorvig, and Gast,[254] growing wheat seedlings in 11 soils by a modified Neubauer technique, found that plant uptake ranged from 42 to 67 and 17 to 77 percent of the applied ^{99}Tc (6.0 μCi per pot of 300 g soil) for the unfertilized and fertilized soils, respectively. In terms of CR, the plant uptake ranged from 590 to 1200 and from 250 to 995 with the unfertilized and fertilized soil, respectively. The application of N-P-K reduced the ^{99}Tc concentrations of plants in all but two of the soils. No apparent toxicity effect was observed in these plants. Compared with monocots (corn, oats, barley, and wheat), high root/shoot CR was found for the dicotyledon species (radishes and soybeans). Routson and Cataldo,[256] growing plants in five different soils (1,000 g per container) found that the percentage uptake of the applied ^{99}Tc ranged from 23 to 82 and 10 to 69 for tumbleweed and cheatgrass, respectively, at the three month's harvest time. The range of CR values was 76 to 390 and 54 to 357 for tumbleweed and cheatgrass, respectively. The plant uptake decreased as the soil pool was depleted. No apparent toxicity was observed in these plants. The results of these investigators indicate a CR range of 54 to 1200, depending on the ^{99}Tc dose level, plant species, age of plant, soil type, and culture method.

Some concentration-toxicity relationships have been examined. In a solution-culture experiment (50 ml of 1/3-strength Hoagland solution), a significant reduction of shoot growth began between ^{99}Tc addition of 0.25 and 1.0 μCi.[254] Measureable effects of ^{99}Tc toxicity on wheat seedlings first occurred at shoot tissue concentrations between 0.68 and 2.8 μCi/g. Whether the toxicity is chemical or radiological in nature remains to be determined. In view of the fact that ^{99}Tc is readily taken up by plants, the effect of toxicity on the absorption and translocation processes needs consideration.

Transuranic Elements

Many actinides are present in the nuclear fuel cycle and, of these, at least seven (U, Th, Np, Pu, Am, and Cm) are of considerable importance in plant uptake as a result of environmental contamination. The significant isotopes of the transuranic elements include ^{237}Np, ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and ^{244}Cm. The series of neutron capture and radioactive decay reactions which lead to their formation are outlined by Pigford and Ang.[258] The plant uptake of U and Th has been discussed above. Because of the limited number of studies on transuranic element uptake by plants, these elements will be discussed together here.

The significance of these elements, particularly Pu, arises from the fact that they have a long radioactive half-life and are extremely toxic.[259-261] The critical organ for these elements is bone for which the maximum permissible burden is 0.04, 0.05, 0.06, and 0.1 μCi for $^{239,240}Pu$, ^{241}Am, ^{237}Np, and ^{244}Cm, respectively.[83] Because of the severe biological toxicity of transuranic elements it is important to prevent, insofar as possible, an indiscriminate dissemination of them into the environment where they might be transported to

mankind and grazing animals through inhalation and food-chain pathways.[262]

The global inventory of Pu was 324 ± 26 kCi ^{239,240}Pu and 216 ± 3.1 kCi ^{238}Pu in 1973.[263] For ^{239,240}Pu, that is 0.6 nCi for each M^2 of the earth's surface if evenly distributed. Worldwide fallout Pu distribution pattern is inferred to be similar to that of fallout ^{90}Sr, which is characterized by a marked peaking in the North Temperate Zone and by much lower values throughout the Southern Hemisphere.[18,264-268] The concentrations of fallout Pu in the surface soils of United States are in the range of 20 fCi/g.[269] At this writing, the concentrations of other transuranic elements had not been well established.

Transuranic element uptake by plants has been reviewed by a number of investigators.[270-275] To date, most work pertaining to transuranic element uptake by plants has been done with Pu and Am. Consequently, most of the discussion here will involve these transuranics. In general, the Pu uptake by plants from contaminated soil is very small.[4,276-280] The review by Schulz[275] on plant uptake shows Pu CR range from 10^{-10} to 10^{-3}, depending on soil type, plant species, and the chemical form of Pu. Langham[19] used a CR of 5×10^{-5} in making estimates of Pu hazard to man. Bennett[281] and Hanson[282] generalized the CR as 10^{-4}. The CR for ^{241}Am, according to Schulz,[275] ranged from 10^{-7} to 10^{+1}. The ^{241}Am uptake by plants is generally considerably greater than that of Pu. For example, Cline[278] reported ^{241}Am CR's 15 to 30 times larger than the Pu CR's in barley plants grown in similar soils. Adams et al[280] found that CR's of ^{241}Am in alfalfa were 20 to 30 times greater than the CR of Pu in similar experimental plots. The investigators[278,280] above worked with soils that had been contaminated with Pu and Am in the solution form. Schulz et al[283] working with Nevada Test Site soils in which Pu

isotopes were present mainly in the particulate form (particles in the range of 0.2 to 0.7 μm diameter) found that Am was taken up 4 to 11 times more readily than Pu by barley plants. Romney et al,[284] also working with Nevada Test Site soils, found the average Am/Pu ratio was 4 for barley, 10 for alfalfa, and 22 for soybeans. The Am/Pu concentration ratios in the plants grown in Nevada Test Site soils generally tended to be lower than those grown in solution spiked soils used by Cline[278] and Adams et al.[280] This tendency was perhaps linked to the fact that the ^{241}Am in the Nevada Test Site soil was, at least in part, inside and/or on a Pu-bearing particle as a direct result of its production by decay of ^{241}Pu.[283] Like ^{241}Am, ^{237}Np and ^{244}Cm also are more readily taken up by plants than is Pu. Price[285,286] working with tumbleweed and cheatgrass observed that < 2, < 0.05, < 0.05, and < 0.001 percent of dose of ^{237}Np, ^{241}Am, ^{244}Cm, and ^{239}Pu, respectively, were taken up. Thomas and Healy[273] came to the same order of uptake in their review. Dahlman, Bondietti, and Eyman[274] included U and reported that the relative order of plant uptake of these actinides appeared to be Np > Am ≈ Cm ≈ U > Pu. A CR greater than 10^{-1} was suggested for Np.[274] The ^{239}Pu CR based on 8 M HNO_3 extractable soil concentrations was found to be only slightly less or about the same as that of ^{232}Th.[99] The ^{239}Pu uptake is also considerably less than significant fission products that have been investigated. Cummings and Bankert[279] indicated that ^{238}Pu uptake by oat plants may be as much as six orders of magnitude lower than ^{85}Sr and ^{137}Cs uptakes by oat plants from the same soils. The ^{239}Pu uptake relative to that of several significant fission products was considered to be in the order $^{89,90}Sr$ >> ^{131}I > ^{140}Ba > ^{137}Cs, ^{106}Ru > ^{144}Ce, ^{91}Y, ^{147}Pm, ^{95}Zr-Nb > ^{239}Pu by Nishita, Romney and Larson.[4]

Many factors contribute to the wide ranges of CR values observed for transuranic elements. As generalized above, these factors are due to varying chemical characteristics of the element in question, soil properties, and plant characteristics. A number of investigations explain some of the variability due to these factors.

Chemical characteristics that influence the transuranic element uptake by plants include concentration, chemical form, particle size, isotopic form, and oxidation state. Some effect of concentration has been illustrated. Wildung and Garland [62] found that the concentration of Pu in barley shoots and roots increased with increased soil Pu concentration. The CR, however, decreased with increased soil Pu. With ^{241}Am, an increase of soil concentration from 10,000 to 40,000 dpm/g resulted in a 3-fold decrease in the CR of barley shoots.[287] The causes of these effects of transuranic elements has not been clearly defined. One factor may be the injurious effect of the transuranics on plant roots at higher soil concentrations. Wildung and Garland[62] suggested a change in Pu availability due to the susceptibility of microbes involved in the solubilization of transuranics at the higher soil concentration levels. Some evidences of microbial action in increasing the availability of Pu to higher plants have been presented by several investigators.[62,288-291] The chemical form of Pu is also known to influence plant uptake. Schulz, Tompkins, and Babcock[292] grew wheat on three soils previously contaminated by the addition of Pu in dilute chloride or nitrate solutions. In the alkaline calcareous soil, the nitrate form of the Pu was taken up about 100 times greater than the chloride form. Adams et al[280] found higher Pu CR's in alfalfa when soils were contaminated by Pu in the nitrate solution form than when it was grown in Pu-contaminated soils heated

to 300° C and 900° C. The decrease of Pu CR was much greater for 900° C soil than for 300° C soil. A major part of this effect was considered to be due to the increasing refractory nature of PuO_2 with increasing temperature. Some effect of the change of chemical state of the heated soil may also be involved. Since PuO_2 is highly insoluble, its particle size may have an effect on its availability to plants. Little et al[293] reported that Pu uptake by plants was negatively correlated with the size of the particles containing Pu. Variation in particle size resulted in several orders of magnitude variations in the concentration of Pu in vegetation. Bio-availaability from relatively large particles is low. According to Adams et al,[280] the CR in ash from plants grown in soil with 100 μm $^{238}PuO_2$ particles ranged from 10^{-7} to 10^{-8}. Isotopic difference between ^{238}Pu and ^{239}Pu in ecosystems has been suggested, but to date this does not appear to be unequivocally established. Little[294] reported that ^{238}Pu was more soluble than ^{239}Pu in a grassland ecosystem. Hakonson and Johnson[295] and Wildung and Garland[296] reported that ^{238}Pu was more soluble than ^{239}Pu when the inorganic salts are added to soil. On the other hand, others report no difference in plant uptake of ^{238}Pu or ^{239}Pu. The oxidation state of an element represents still another aspect. Early work of Jacobson and Overstreet[276] indicated a relative magnitude of plant uptake of Pu in different oxidation states to be $PuO_2^{++} > Pu^{++++} > Pu^{+++}$. Their relative sorption on clay (bentonite) was in the same order. Thus, differences in the availability of the different oxidation states may be related to their relative reactivity with soil components or their relative insolubilities. Dahlman, Bondietti, and Eyman[274] pointed out that plant uptake appears to be consistent with the relative tendencies of the different oxidation states to hydrolyze. Strictly

speaking, elements in the same oxidation states should be compared when considering their relative uptake by plants. However, elements of different oxidation states are often compared against each other. As an illustration, Price[285,286] compared the plant uptake of Np (V), Pu (IV), Am (III), and Cm (III) and found that Np (V) was taken up most readily, while that of Pu (IV) occurred least readily. This manner of comparison was done in formulating the relative plant uptake series presented above. This type of comparison has practical basis, since the most prevalent oxidation states of these elements in nature are likely to be those that were used. This is supported by the fact that the most stable oxidation state in aqueous solution is V for Np, IV for Pu, and III for Am and Cm.[297] Bondietti, Reynolds, and Shanks[298] indicated that Pu (IV) is the most likely stable oxidation state in the environment, since Pu (VI) is reduced to Pu (IV) by natural organic matter and soil clays containing organic coatings and Pu (V) is an intermediate in the reduction of Pu (VI). Pu (III) is oxidized to Pu (IV) by atmospheric oxygen.[297,299,300]

Several soil factors are known to influence the uptake of transuranic elements by plants. The pH of the soil or growth medium is one of the properties that is often related to plant uptake. Rediske, Cline, and Selders[301] found the CR of ^{239}Pu increased from 10^{-4} to 10^{-3} with decrease of pH of root medium from 7 to 4. Wilson and Cline's[277] data showed that ^{239}Pu uptake for an acid soil by barley was three times that of an alkaline calcareous soil. Comparisons of this kind are often made and the results ascribed to pH. The pH, however, is usually accompanied by other factor(s) that may be different for different situations. These factors may be the dominant ones rather than pH per se. For example, the pH effect observed by Rediske, Cline, and

Selders[301] may well be due mainly to the change of degree of hydrolysis and polymerization of Pu with pH. The difference between acidic and alkaline calcareous soils may be due to the strong interaction of Pu with insoluble $CaCO_3$ in the alkaline calcareous soil rather than pH per se. Organic matter in the soil is another factor of considerable importance. A number of organic compounds can complex or chelate with transuranics.[297,302,303] Information discussed by Gel'man et al[303] indicates the complexing tendency for the transuranic elements considered here is in the order $Pu^{+4} > Am^{+3} \approx Cm^{+3} > NpO_2^{+}$. Complex formation can influence the availability of these elements to plants. Price[286] found that plant uptake of these transuranics added to the soil as organic acid complexes (acetate, glycolate, oxalate, and citrate) was in the same order as uptake from the nitrate forms, i.e. Np > Am ≈ Cm > Pu. The order of uptake compared among the transuranics was unchanged, but uptake compared among chemical forms varied with the kind of transuranic element and complexing agent used. Transuranics with oxidation state of IV and V (Pu and Np respectively) generally showed increased uptake with complexation, and the III elements (Am and Cm) showed little change or a decreased uptake relative to nitrate forms. The cause of the latter effect is still to be determined. Several investigators have determined the effect of chelating agent, diethylenetriamine pentaacetic acid (DTPA), on the plant uptake of ^{239}Pu[304] and ^{241}Am.[305-310] In a sand culture, Lipton and Goldin[304] found that DTPA increased the ^{239}Pu uptake by pea plants by a factor of 1.3×10^3. Hale and Wallace[308] found that DTPA increased the uptake of ^{241}Am by two orders of magnitude. This, however, depends upon concentration of DTPA and other factors. A 10-fold increase of ^{241}Am concentration (from 10,000 to 40,000 dpm/g soil) in the soil without applied DTPA resulted in a 3-fold decrease in

the CR of barley shoots. On the other hand, with DTPA applied to the contaminated soil, only slight, if any, change of CR occurred over the same ^{241}Am concentration range.[310] A 16-fold increase of ^{241}Am (from 3475 to 55,600 dpm/g soil) in the soil without applied DTPA decreased the CR of bush bean leaves 3.9 times, while under the same condition but with applied DTPA, the CR of the leaves was about doubled.[287] Also, the +DTPA/-DTPA ratio of ^{241}Am uptake increased 7.5 times in the leaves and 64.8 times in stems, indicating the partitioning of ^{241}Am between stems and leaves as well as increase in concentration. The pH has an effect because of the pH to metal chelate stability relationship. Wallace[307] found that more ^{241}Am is taken up at pH 7.7 than at higher and lower pH. DTPA may also change the translocation gradient between organs. When bush bean plants were grown in solution culture with DTPA, less accumulation of ^{241}Am on roots and greater transfer to shoots from roots occurred than those grown in -DTPA solution.[287] Aside from DTPA, cyanide, which is a chelator of many heavy metals, has also been studied and found to increase the transport of ^{241}Am into shoots of bush beans.[311] In general, organic chelate formation is an important reaction, since soils contain many different organic compounds.[41,312-314] Organic chelation or complexation, however, does not necessarily increase the availability of transuranics to plants. Cataldo et al,[315] growing soybeans on seven soils by the split-root technique, found some evidence that the rate of ^{238}Pu accumulation (dpm accumulated in shoot/day) is inversely related to soil organic C content. The influence of natural organic compounds in soils on transuranic element uptake by plants needs to be investigated further.

Plant characteristics are an important factor that influence transuranic element uptake by plants. Thomas and Jacobs [316] found that ^{242}Cm was taken up

in appreciable amounts by bean plants, but it was not taken up by tall fescue. Price[286] observed that ^{237}Np uptake by tumbleweed was at about 2 percent of dose and that by cheatgrass was an order of magnitude lower. Cline and Hinds[317] reported that monocots took up less ^{238}Pu than did dicots. Aside from plant species effect, a root to shoot to seed concentration gradient in transuranic element accumulation is generally observed. Rickard et al[318] observed differences among crop species in their ability to accumulate ^{238}Pu in shoots and also in seeds. Peas, for example, had about 10 times greater amount of Pu in shoots as in cheatgrass, but only about 1/16 as much Pu in seeds as in cheatgrass grain. Jacobson and Overstreet[276] found that although large amounts (20 to 38 percent) of ^{239}Pu were sorbed on plant roots, very little (about 0.00045 to 0.01 percent depending on the oxidation state of Pu) was translocated to the aerial part of the plants. Rediske and Selders[319] noted that the sorption of ^{239}Pu (IV) to the root surfaces from culture solutions but found very small concentrations of Pu translocated to other parts of the plant. Wildung and Garland[62] observed that barley root Pu concentration exceeded that of the shoots by a factor of 3 to 8, depending on soil Pu concentration. In considering root concentration, surface contamination could be an important factor. Adams et al[280] found that radish roots contained 10 times as much ^{239}Pu as tops but 99 percent of the root radioactivity was removed by peeling. Shoot to seed gradient also occurs. According to Adams et al[280] bean seeds contain 200 times less Pu tnan did leaves. Cline and Hinds[317] obtained similar results with other plant species. Plutonium was less mobile than ^{241}Am in retranslocation from soybean shoots to seed pods, and their concentration in seed pods was highly positively correlated with that in the shoots.[284] Shoot to fruit gradient could be influenced by

surface contamination. Cataldo and Vaughan[320] found that foliar applied aerosols of ^{238}Pu were not only tenaciously held by leaves, but also that the Pu was available for transport to fruits with a CR of shoots to fruits about like that for soil to plants. Plant age can be another factor since it can determine the elemental concentration in the tissues. Soybeans grown on seven soils by split-root technique by Cataldo et al[315] accumulated ^{238}Pu linearly for the first 32 to 53 days following germination. After this time period, the rate of accumulation by shoot tissues decreased slightly or remained relatively constant, depending on the soil on which the plants were grown. The cotyledons and primary leaves attained near maximum Pu content at 20 days. The Pu content of the stem tissue reached an initial maximum at 4 days, decreased rapidly by 10 days, and remained relatively constant to 40 days. Increased accumulation occurred at a time of rapid dry matter production and growth of shoot tissues. The Pu accumulation in shoot tissues decreased from older, basipetal to younger, acropetal tissues.

Aside from the factors discussed above, there are several others that need consideration. One is the change of bio-availability of contaminants with prolonged cropping of a soil. Newbould[321] saw a time-related change in uptake. Growing rye-grass in three different soils, he found somewhat greater Pu uptake by plants the second year than the first. Romney, Mork, and Larson[322] found a 5-fold increase in the Pu concentration of ladino clover grown on a Nevada Test Site soil in containers under greenhouse conditions during a 5-year period. From the soil of 10^5 dpm Pu/g, the plant concentration increased from 3.1 dpm/g in 1958 to 22.6 dpm/g in 1962. This apparent increase was considered to be due to natural chelation of the residual Pu. Concentration of the contaminating transuranic elements in the

soil needs further consideration. Most of the work considered above has been done at relatively high concentrations of transuranics in soils. More observations of plant uptake at environmental levels are needed, since there is an indication that concentration ratio of Pu increases as its concentration in the soil decreases.[62]

CONCLUSIONS

Natural radionuclides are always present in the environment and, together with those radionuclides made by man in past thirty plus years, they differentially move through geochemical and biological cycles. One important limitation on the use of nuclear resources is the level of radionuclides at which various environments may become contaminated. To cope with this limitation, knowledge concerning the principles governing the uptake, transport and redistribution in plants of various radionuclides is of utmost importance. Principles discussed in this review can help control uptake by plants and will also help dictate tolerance levels under various situations.

Adverse impacts from radioactive contamination of foodstuffs vary with the individual radionuclides involved. Reaction mechanisms regulating food chain transport are sufficiently understood to sort out those radionuclides which are of potential concern to health and welfare of mankind. A wealth of information has been gathered but this review indicates important gaps. Information on uptake, transport, and redistribution of transuranics, particularly ^{237}Np and ^{244}Cm, is still quite limited. Most of the plant uptake studies on transuranics have been done at relatively high concentrations. More work must be done at environmental levels. Fission product ^{99}Tc needs more attention.

Differentiation between radiological and chemical toxicity on the absorption of ions by plants needs to be clearly defined. Research in these areas as well as environmental monitoring should be continued as the world moves further into the nuclear age.

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Table 1

Uranium Contents of Soils

Soil Location	Concentration Range ppm	Reference*
Ispra, Italy	1.5 - 3	100
Several Areas in United States	1.92 - 5.06	101
Several Areas in California	0.78 - 3.6	102
Several Areas in Russia	2.6 - 4	85
Japan	1.9 - 4	92
Niue Island, Pacific Ocean	60	103
Colorado Plateau, United States	∿ 100	85

* Refers to references of this review.

Table 2

Typical Concentration of ^{226}Ra in Soil Excluding High-Background Areas

(From Gera, 1975 (84))

Region	Soil (dry weight basis) (pCi/g)	Reference*
British Guiana	0.3 - 1.6	106
Czechoslovakia	0.1 - 3.8	85
Germany	0.15 - 1.3	80
Ireland	1.3 - 2.9	85
Italy (zone of Varese)	0.72 $\pm$ 0.11	115
United Kingdom	0.08 - 1.5	85, 106
U.S.A.	0.8 - 2.8	85
U.S.S.R.	0.1 - 1.3	85

* Refers to references of this review.

Table 3

Radium Content of Plants Used as Foods

Plant	^{226}Ra concentration		Reference*
carrots	1.6 - 6.1	pCi/kg	109
cabbage (2 samples)	1.0 , 2.4	fresh weight	
apples	0.9		
potatoes (N. Germany)	1.0		
potatoes (S. Germany)	0.6		
Brazil nuts	2730	pCi/kg	114
peanuts	120	fresh weight	
pears	1.1		
cabbage	1.7 - 3.8	pCi/kg	116
lettuce	4.5 - 5.5	fresh weight	
tomatoes	1.2		
corn	1.5		
apples	1.8		
pea	10.5	10^{-16} g/g	117
lima beans	10.5	dry plant	
kidney beans	79		
peas, green	19		
peas, yellow	39		
wheat (2 samples)	60,50	10^{-14} g/g ash	118
potatoes (2 samples)	46,87		

* Refers to references of this review.

Table 4

Strontium-90 Concentration in Ladino Clover Grown on Sassafras Sandy Loam Treated with $CaCO_3$ and $CaSO_4$ (from data of Romney et al)[139]

Amount of Ca added (meq/100 g soil)	^{90}Sr concentration $CaCO_3$ (pCi/g dry tissue)	$CaSO_4$ (pCi/g dry tissue)
1.0	2454	1244
5.0	1143	556
10.0	824	498

Sassafras sandy loam pH = 4.6, cation exchange capacity = 6.0 meq/100 g, exchangeable Ca = 0.4 meq/100 g.