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UPTAKE BY PLANTS OF RADIONUCLIDES FROM FUSRAP WASTE MATERIALS

by

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ABSTRACT

Radionuclides from FUSRAP wastes potentially may be taken up by plants during remedial action activities and permanent near-surface burial of contaminated materials. In order to better understand the propensity of radionuclides to accumulate in plant tissue, soil and plant factors influencing the uptake and accumulation of radionuclides by plants are reviewed. In addition, data describing the uptake of the principal radionuclides present in FUSRAP wastes (uranium-238, thorium-230, radium-226, lead-210, and polonium-210) are summarized. All five radionuclides can accumulate in plant root tissue to some extent, and there is potential for the translocation and accumulation of these radionuclides in plant shoot tissue. Of these five radionuclides, radium-226 appears to have the greatest potential for translocation and accumulation in plant shoot tissue.

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1. INTRODUCTION

1.1 DESCRIPTION OF THE FUSRAP PROGRAM

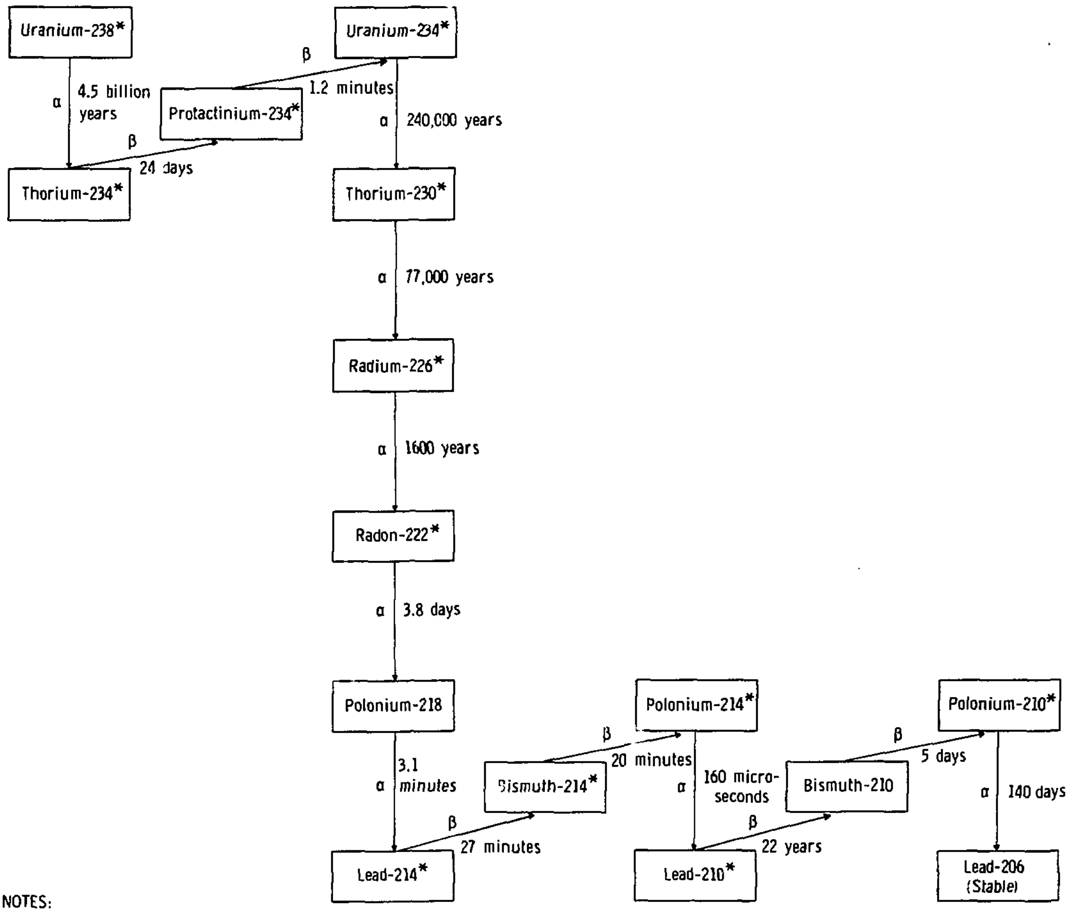
During the 1940s and 1950s, the U.S. Army Corps of Engineers Manhattan Engineer District (MED) and the U.S. Atomic Energy Commission (AEC) conducted programs that resulted in a number of sites throughout the continental United States becoming radioactively contaminated. When MED/AEC activities were terminated, many of these sites were decontaminated in accordance with health and safety criteria and guidelines then in use. Recent radiological surveys of these formerly utilized sites have indicated that residual radioactive contamination exists at levels that may require remedial action at a number of the sites. The U.S. Department of Energy's (DOE) Formerly Utilized Sites Remedial Action Program (FUSRAP) was initiated for the purpose of decontaminating these sites to permit their unrestricted use or to stabilize and/or otherwise control residual radioactivity at the sites to meet current criteria for the protection of public health and safety.

Large volumes of soil and rubble containing trace amounts of radionuclides could be generated by the decontamination activities and require subsequent disposal. The potential for plant uptake of radionuclides from these FUSRAP wastes exists during the remedial action activities and permanent near-surface burial of the contaminated materials. Therefore, when assessing the efficacy of methods intended to isolate FUSRAP materials from the surrounding environment, it is essential to understand the process of radionuclide uptake by plants and the degree to which the radionuclides present in FUSRAP wastes may be accumulated in plant tissue. The purpose of this discussion is to: (1) introduce the reader to the FUSRAP program, (2) review the soil and plant factors influencing the uptake and accumulation of radionuclides by plants, and (3) summarize currently available data describing the plant uptake of specific radionuclides present in FUSRAP wastes.

Radioactive material at FUSRAP sites consists primarily of waste by-products from uranium and thorium processing that are present as contaminants in building materials and soil (U.S. Dep. Energy 1981). Because more than 90% of these wastes resulted from uranium processing activities, this review focuses on radioactive isotopes of the uranium-238 decay series (Figure 1.1). Due to their potential radiotoxicity, the length of their half-lives, and their predominance in FUSRAP wastes, the specific radioactive elements discussed in this review are uranium-238, thorium-230, radium-226, lead-210, and polonium-210.

1.2 FACTORS INFLUENCING RADIONUCLIDE UPTAKE BY PLANTS

The interaction of plants with radionuclides from FUSRAP wastes can occur via two pathways: (1) by foliar absorption of radionuclides deposited upon



NOTES:

Only the dominant decay mode is shown.
The times shown are half-lives.
The symbols α and β indicate alpha and beta decay.
An asterisk indicates that the isotope is also a gamma emitter.

Figure 1.1. Uranium-238 Decay Series.

leaf and stem surfaces or (2) by uptake from the plant root zone in the soil. Radionuclides could reach foliar surfaces by means of fugitive dust emissions from remedial action activities with subsequent deposition of contaminated particulate matter on adjacent vegetation. Radionuclides adsorbed on the surface of particles retained on plant foliar surfaces are likely to be available for foliar uptake and subsequent accumulation (Koranda and Robison 1978). However, techniques intended to minimize fugitive dust emissions (e.g., water sprays) are likely to be employed during remedial action activities. Thus, it appears that the principal pathway by which plants might accumulate radionuclides from FUSRAP wastes would be root uptake. This could occur both at interim-storage sites (through plant establishment directly in contaminated materials) and at permanent disposal facilities (by root intrusion of buried waste materials).

Uptake by plant roots of uranium, thorium, radium, lead, and polonium from soil is influenced by a number of factors, including properties of the contaminated soil, local climatic conditions, and the biosynthetic activity of the plant. The major factor governing the availability to plants of these elements in soils will likely be the solubility and thermodynamic activity of uncomplexed ions (Jenne and Luoma 1977). In order for root uptake to occur, soluble ionic forms must exist in the soil solution adjacent to the root membrane for some finite period (Cataldo and Wildung 1978). The chemical form of the soluble species will have a strong influence on its longevity in the soil solution, its mobility in soil, and the rate and extent of plant uptake.

On the basis of research with various trace metals, it can be concluded that the solubilization of radionuclides is determined largely by the form and concentration of the radionuclides involved as well as by the physicochemical characteristics of soil. Important soil characteristics include: soil solution composition; conductivity; pH; quantities of various clays and humic, fulvic, and other organic complexes present; type and density of charge on soil colloids; soil organic matter concentration and composition; and amount of reactive surface area (Cataldo and Wildung 1978; Koranda and Robison 1978). These phenomena are in turn dependent upon other properties of soil such as: particle size distribution; quantity and reactivity of hydrous oxides, carbonates, phosphates, sulfates, and other anions; mineralogy; degree of aeration; and microbial activity. Geographically, these physical and chemical properties of soil are extremely variable and are the result of the combined effects of soil parent material, local topography, climate, biological processes, time, and human activities (Brady 1974).

The uptake and accumulation of ions from the soil solution are dependent on a number of plant factors as well. These include: the plant species involved, plant age, stage of growth, rate of physiological activity, plant rooting characteristics, plant nutritional status, moisture availability, the kinetics of ion transport across membranes, the metabolic fate of absorbed ions, and ion interactions within the plant (Cataldo and Wildung 1978; Dvorak et al. 1978).

The relative efficiency with which plants take up radionuclides from soil is also dependent on the interrelationships between the soil factors and plant factors discussed above. It is clear from this discussion that the factors influencing the chemical form and plant availability of radionuclides in the soil solution and their subsequent uptake by plants are highly complex.

2. PLANT UPTAKE OF RADIONUCLIDES PRESENT IN FUSRAP WASTE

Information describing the uptake and accumulation of uranium, thorium, radium, lead, and polonium by plants is limited. Much of the available information is based on short-term, relatively high-exposure laboratory studies. The results of plant uptake and translocation experiments conducted under artificial laboratory conditions may not be applicable to the long-term, low-level exposure conditions likely to occur at FUSRAP interim-storage sites or near-surface burial sites. The manner in which such short-term experiments are conducted often precludes growing the plants to maturity, thereby excluding important plant parts (e.g., grain, fruits) from study (Schulz 1977). In the discussion that follows, field studies and laboratory experiments in which plants were grown to maturity are emphasized.

Due to the complexity of the processes by which these radionuclides are taken up by plants and the limitations of available data, it is not possible to determine in a precise manner the degree to which uranium-238, thorium-230, radium-226, lead-210, and polonium-210 will accumulate in plants at FUSRAP interim-storage sites or near-surface burial sites. However, the literature review presented below will give an indication of the propensity for uptake and accumulation of these radionuclides by plants.

2.1 URANIUM

Relatively few plant uptake studies have been conducted with uranium, other than observations related to geobotanical prospecting for this element (Price 1973). For example, trees rooted in uraniferous soils associated with uranium ores commonly contain 1 to 2 μg U/g of tissue compared to an average of 0.5 μg U/g for trees growing on adjacent nonuraniferous soil (Gough et al. 1979). The uranium content of vegetables grown in soil containing "natural" or background amounts of this element (2 pCi U/g of soil) ranged from 5.6×10^{-4} to $<7.0 \times 10^{-4}$ pCi U/g (Laul et al. 1977). Several laboratory pot-, sand- and solution-culture studies have been conducted to determine (1) the effects of water availability on uranium uptake, and (2) the concentrations of soil uranium that are toxic to plants. Prister and Prister (1970) found that for corn (*Zea mays*) and bean (*Phaseolus* spp.) plants grown in sand-culture until each plant had four pairs of leaves, doses of up to 200 mg U/kg of sand (uranium isotope not specified) had no effect on plant dry weight. In a second experiment, the authors reported that uranium accumulation by 13-day-old corn plants grown in solution-culture occurred primarily in root tissue; less than 1% of the uranium taken up by the corn seedlings was translocated into shoots. In this solution-culture experiment, symptoms of uranium toxicity (e.g., loss of turgor, leaf curling) were reported in seedlings exposed to uranium concentrations greater than 50 mg U/L.

In greenhouse soil-culture studies using a sandy-loam soil (pH = 8.5), Gulati et al. (1980) demonstrated that uranium uptake by plants depended not only on the uranium concentration in the soil but also on the amount of water supplied. The biomass of wheat (Triticum aestivum) grown until maturity actually increased as soil uranium content increased from 0 to 3 μg U/g dry wt. of soil. When the amount of uranium added to soil was increased to 6 μg U/g, wheat biomass was significantly depressed. In contrast, the biomass of tomato (Lycopersicon esculentum) continuously decreased with increasing additions of from 0 to 6 μg U/g dry wt. of soil. The uranium content of mature wheat shoot tissue increased from 0.07 to 0.22 μg U/g dry wt., and the uranium content of mature tomato shoot tissue increased from 0.05 to 0.23 μg U/g dry wt. with increasing soil-uranium concentrations. Uranium uptake was also slightly increased in both wheat and tomato when the amount of water supplied to plants over the course of the study was increased for all uranium exposure levels. For mature wheat plants, shoot uranium concentrations increased from 0.11 to 0.18 μg U/g dry wt. as the amount of water applied increased from 11.5 to 19.2 L. Tomato uranium uptake increased from 0.13 to 0.18 μg U/g dry wt. as water applications increased from 15.1 to 25.2 L.

In recent years, a number of field investigations have been reported concerning uranium uptake from uranium mill tailings or contaminated soils. However, it should be noted that because the uranium milling process removes >90% of the uranium in the ore, the concentration of uranium present in mill tailings may be somewhat low relative to certain FUSRAP wastes. Moffett and Tellier (1977) established stands of creeping red fescue (Festuca rubra), redtop (Agrostis alba), reed canarygrass (Phalaris arundinacea), and 'climax' timothy (Phleum pratense) on a series of field plots in both coarse- and fine-textured uranium mill tailings in the Elliot Lake area of Ontario, Canada (average annual precipitation of 81 cm). The accumulation of uranium by the aboveground biomass of these perennial species was analyzed after four growing seasons. The analytical techniques employed to measure the uranium content of plant tissue did not discriminate between the various isotopes of uranium. The average uranium content of the four plant species ranged from 0.02 μg U/g dry wt. ('climax' timothy) to 0.04 μg U/g dry wt. (redtop). The uranium concentration in the mill tailings of the various field plots averaged 17.45 μg U/g of tailings material.

One measure of a plant's ability to accumulate (or concentrate) in its tissue a specific element from a growth medium (e.g., soil) is the concentration ratio (CR):

$$CR = \frac{\text{Element's Concentration in Plant Tissue}}{\text{Element's Concentration in Growth Medium}}$$

The CRs for the study of Moffett and Tellier ranged from 0.001 to 0.003. With CR values this low, these plants do not appear to be accumulating large amounts of uranium in their aboveground parts. However, the authors gave no indication of the nutritional status or general condition of the vegetation established on the mill tailings. The unusual edaphic conditions (e.g., low pH) of the tailings may be adversely affecting plant growth, which may in turn reduce the rate of uranium uptake and/or translocation by plants. Indeed, for shoot tissue of a number of rangeland grasses and shrubs growing on contaminated soils adjacent to a uranium mill tailings pond in New Mexico, the uranium CR values reported by Rayno et al. (1980) varied from 0.01 to 0.12 (Table 2.1).

Table 2.1. Concentration Ratios of Uranium for a Variety of Plant Species

Plant Species	Plant Part	Growth Medium†[1]	CRs from Various Data Sources: Moffett and Tellier (1977)	Rayno et al. (1980)	Garten (1980)
Creeping red fescue (Festuca rubra)	Shoot	UMT	0.002	-	-
Reed canary grass (Phalaris arundinacea)	Shoot	UMT	0.001	-	-
Redtop (Agrostis alba)	Shoot	UMT	0.003	-	-
'Climax' timothy (Phleum pratense)	Shoot	UMT	0.001	-	-
Russian thistle (Salsola paulsenii)	Shoot	CS	-	0.11	-
Indian ricegrass (Oryzopsis hymenoides)	Shoot	CS	-	0.12	-
Four-wing saltbush (Atriplex canescans)	Shoot	CS	-	0.01	-
Boxelder (Acer negundo)	Leaves	CS	-	-	0.0007-0.011

†[1] UMT = uranium mill tailings; CS = contaminated soil.

Rayno et al. (1980) also collected and analyzed root tissue samples for several rangeland grasses, i.e., galleta grass (Hilaria jamesii), blue gramma grass (Bouteloua gracilis), Indian ricegrass (Oryzopsis hymenoides), and dropseed grass (Sporobolis asper). Uranium concentrations in roots ranged from 0.63 to 12.5 pCi U/g dry wt. of tissue. The distribution index (ratio of root radionuclide concentration or specific activity to shoot radionuclide concentration or specific activity) was calculated for these species. The values calculated were 0.82, 1.65, 3.30, and 4.69 for galleta grass, blue gramma grass, Indian ricegrass, and dropseed grass, respectively.

Recent studies have examined the uptake of uranium by trees and grasses from the soils of a Tennessee floodplain contaminated when the site was used as a temporary retention pond for low-level liquid radioactive wastes in 1944. Garten (1980) investigated the pattern of seasonal change in the concentrations of various actinides in the leaves of box elder trees (Acer negundo). The concentrations of both uranium-234 and uranium-238 in leaf tissue increased throughout the growing season (May-September) and far exceeded those of the other actinide elements examined--plutonium-239, americium-241, and curium-244. Uranium-238 concentration ratios varied from 0.0007 to 0.011 over the growing season. Garten et al. (1981) studied radionuclide uptake by Kentucky-31 fescue (Festuca arundinacea) grown on the same contaminated floodplain. This species accumulated proportionally 10-fold more uranium than plutonium or thorium from these soils. Although the manner in which Garten et al. presented the data (i.e., radionuclide ratios) does not allow the determination of uranium accumulation by fescue, the uranium content in fescue shoot tissue was probably on the order of 10^{-2} to 10^{-3} ng U/g dry wt.

2.2 THORIUM

Few studies have been conducted to determine the uptake and translocation of thorium by plants. The uptake of thorium-230 by red kidney beans (Phaseolus vulgaris) grown in solution-culture has been reported by D'Souza and Mistry (1970). Fifteen-day-old bean seedlings were placed in a nutrient solution containing 0.25 µCi Th-230/L and grown for a period of 15 days. After this exposure period, root and shoot tissues were assayed for alpha activity. Only 0.09% of the thorium-230 added to the nutrient solution had been translocated to shoot tissue, whereas 76.35% had been taken up by root tissue or adsorbed onto root surfaces. The CRs for bean shoot and root tissues were 0.91 and 4185, respectively. It should be noted, however, that no effort was made by the authors to remove thorium adsorbed to root surfaces (e.g., by rinsing or washing) before radioassay. Therefore, the CR calculated from these data is likely to be an overestimate of bean root thorium-230 uptake. In addition, it should be emphasized that this study employed a solution-culture growth medium and, thus, thorium added to the nutrient solution was more available for plant uptake than in a soil-culture system. Soil binding may be the principal factor inhibiting the movement of thorium into plants (Russell and Smith 1966).

Thorium-230 uptake and translocation into aboveground, field-grown vegetation has also been investigated (Moffett and Tellier 1977). The thorium content of the aboveground parts of plants grown on uranium mill tailings ranged from 0.003 to 0.022 µg Th-230/g dry wt. The CRs for this study ranged from 0.0006 to 0.004 (Table 2.2).

In a study by Rayno et al. (1980), the thorium-230 content for forage grasses growing in the vicinity of a uranium mill tailings pond ranged from 0.12 to 13.2 pCi Th-230/g dry wt. The only CR calculated on the basis of thorium-230 in shoot tissue was 0.11 for Indian ricegrass. This CR approaches or exceeds those calculated in this study for uranium-238 and radium-226, radionuclides considered more likely to be taken up by plants than thorium-230. The distribution index for thorium in the forage species sampled varied from 0.30 to 10.50.

Some workers (Garten et al. 1981; D.A. Cataldo, personal communication) believe that thorium tends to behave environmentally like plutonium. The few thorium uptake studies found in the literature tend to substantiate this position. [The mobility of plutonium in soils and its uptake by plants has been reviewed by Francis (1973) and Price (1973).] Garten et al. (1981) reported that there were no significant differences between the uptake of plutonium-239 and thorium-232 by Kentucky-31 fescue. Shoot tissue concentrations of thorium-232 were on the order of 10^{-4} ng Th/g dry wt. Further statistical analysis indicated that no differential transfer (i.e., CR $\geqq$ 1.0) of thorium-232 from soil to fescue shoot tissue occurred.

Table 2.2. Concentration Ratios of Thorium-230 for a Variety of Plant Species

Plant Species	Plant Part	Growth Medium†[1]	CRs from Various Data Sources: Moffett and Tellier (1977)	Rayno et al. (1980)	D'Souza and Mistry (1970)
Creeping red fescue (Festuca rubra)	Shoot	UMT	0.0006	-	-
Reed canary grass (Phalaris arundinacea)	Shoot	UMT	0.002	-	-
Redtop (Agrostis alba)	Shoot	UMT	0.004	-	-
'Climax' timothy (Phleum pratense)	Shoot	UMT	0.001	-	-
Indian ricegrass (Oryzopsis hymenoides)	Shoot	CS	-	0.11	-
Red kidney beans (Phaseolus vulgaris)	Shoot	SC	-	-	0.91
	Root	SC	-	-	4185.0

†[1] UMT = uranium mill tailings; CS = contaminated soil; SC = solution culture.

2.3 RADIUM

Of the radionuclides considered in this review, radium-226 appears to have the greatest propensity for uptake and accumulation by plants because it serves as an analog for calcium, an essential plant nutrient. In fact, D'Souza and Mistry (1970) suggest that radium-226 is likely to be the major contributor of radioactivity to the aerial portions of plants grown under conditions where root absorption is the principal route of radionuclide entry into plants. This conclusion is based upon the results of radium-uptake experiments using red kidney bean seedlings grown in solution-culture. During a 15-day exposure period, bean shoot tissue accumulated 9.61% of the 0.25 μCi of radium-226 present in the nutrient solution used--an accumulation equal to a CR of 84.60.

However, Moffett and Tellier (1977) reported substantially less radium-226 uptake by plants in their field studies. The CRs of radium-226 calculated from their data vary from 0.002 to 0.04 (Table 2.3), four or five orders of magnitude less than that determined by D'Souza and Mistry. It appears that, like thorium, much of the radium in uranium mill tailings and soil may not be available for plant uptake. Kirchmann and Boulenger (1966) showed an inverse relationship between the logarithm of radium-226 concentrations of both plant shoots and roots and the amount of sorptive material in soil. The specific type of sorptive material (e.g., soil organic matter, clay content) was not specified. However, it appears that soil radium availability to plants is highly dependent upon soil organic matter content (Rayno 1982). The radium-226 CR values reported by Rayno et al. (1980) are also quite low (0.01-0.25) and in general agreement with those of Moffett and Tellier (1977).

Table 2.3. Concentration Ratios of Radium-226 for a Variety of Plant Species

Plant Species	Plant Part	Growth Medium†[1]	CRs from Various Data Sources		
			Moffett and Tellier (1977)	Rayno et al. (1980)	D'Souza and Mistry (1970)
Creeping red fescue (*Festuca rubra*)	Shoot	CMT	0.04	-	-
	Shoot	FMT	0.03	-	-
Reed canary grass (*Phalaris arundinacea*)	Shoot	CMT	0.03	-	-
	Shoot	FMT	0.002	-	-
Redtop (*Agrostis alba*)	Shoot	CMT	0.04	-	-
	Shoot	FMT	0.004	-	-
'Climax' timothy (*Phleum pratense*)	Shoot	CMT	0.04	-	-
	Shoot	FMT	0.006	-	-
Russian thistle (*Salsola paulsenii*)	Shoot	CS	-	0.04-0.25	-
Indian ricegrass (*Oryzopsis hymenoides*)	Shoot	CS	-	0.01	-
Sticky snakeweed (*Gutierrezia lucida*)	Shoot	CS	-	0.11	-
Red kidney beans (*Phaseolus vulgaris*)	Shoot	SC	-	-	84.60
	Root	SC	-	-	1787.0

†[1] CMT = coarse-textured uranium mill tailings; FMT = fine-textured uranium mill tailings; CS = contaminated soil; SC = solution culture.

2.4 LEAD

Due to the toxicity of lead to animals and man, a large body of literature has developed describing the uptake of lead by plants and assessing whether crops and vegetables are a significant pathway for lead exposure. Although little of this effort has focused specifically on the radioisotope lead-210, it is very likely that the pattern of plant uptake and translocation of this isotope will be quite similar to nonradioactive isotopes of lead. Therefore, only a brief discussion of the available information will be presented here. For further detail, the reader is referred to the reviews of Cannon (1976) and Demayo et al. (1982).

As discussed earlier, the uptake of trace elements or radionuclides depends upon a number of physical and chemical characteristics of soil. In the case of lead, soil pH and organic matter content appear to be the parameters controlling lead availability in soil (Demayo et al. 1982). Evidence suggests that the availability of lead to plants is greatest in soils with an acidic pH (< 6.0) and very low organic matter content. Under other soil conditions, the capacity of soil to bind lead by precipitation, sorption, and chelation indicates that probably very little of the total lead content of soil is available for plant uptake. Of the amount available for uptake, it appears that much of the lead taken up by plant roots remains in the roots, with little translocation to aboveground plant parts. Lead levels 2 to 20 times greater in root tissues than in leaf tissues have been reported in alfalfa and brome grass (Demayo et al. 1982). Similar results have been obtained in experiments specifically focused upon the uptake of lead-210 by plants. Tobacco plants grown in 3 kg [illegible] soil amended with 100 μCi of lead-210 accumulated less than 3% of the lead-210 addition in their tissues during an exposure period of two weeks (Tso and Fisenne 1968). Less than 1% of the lead-210 accumulated in plant tissue was found in leaves and stems, with the remainder occurring in the roots. Athalye and Mistry (1972) reported little movement of lead-210 from roots into shoots of tobacco plants grown in solution-culture.

In a field investigation, Rayno et al. (1980) reported CRs of lead-210 ranging from 0.08 to 0.54 in the aboveground portions of several western desert plants.

2.5 POLONIUM

Polonium-210 has a half-life sufficient to allow significant accumulation (food-chain transfer); however, little effort has been made to understand the uptake of this radioisotope by plants. Berger et al. (1965) reported the polonium-210 content of a number of field-grown vegetables and crops (i.e., corn, tobacco, cabbage, carrots, red beets, cucumbers, radishes, snap beans, and potatoes) and associated soils. No detectable concentrations of polonium-210 were found in any of the plant species sampled, although levels of 1.9 to 5.9 pCi/g were measured in soil.

Tso and Fisenne (1968) found that the uptake and translocation of polonium-210 occurs to a certain extent in tobacco plants. In a two-week period, tobacco plants grown in 3-kg pots containing 50 μCi of polonium-210 took up approximately 6% of the polonium present. Of this amount, roughly half remained in root tissue, with the remainder present principally in the tissue of stems and older leaves. In experiments where 17 μCi of polonium-210 was fed directly into the stems of tobacco plants, older leaves contained approximately five times the polonium concentration found in younger leaves. More recently, Athalye and Mistry (1972) reported a similar pattern of polonium-210 uptake in the leaves of tobacco grown in solution-culture.

3. DISCUSSION

Uranium-238, thorium-230, radium-226, lead-210, and polonium-210 are all accumulated in root tissue to some extent. Thus, root intrusion of buried FUSRAP wastes could potentially result in the movement of these radionuclides from the waste material into overlying uncontaminated soil cover layers. However, insufficient information is available to determine the rate of radionuclide movement into overlying cover layers.

There is potential for translocation and accumulation of radionuclides from root to shoot tissue. Of the radionuclides, radium-226 appears to have the greatest potential for translocation and accumulation in plant shoot tissue. Information describing the mobility of these radionuclides within plant tissue is needed to determine the extent of this phenomenon.

Plant uptake of these radionuclides into root or shoot tissue could potentially result in subsequent human exposure to the nuclides and their decay products.

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