NVO-178
UC-11

TRANSURANICS IN NATURAL ENVIRONMENTS

JUNE 1977

EDITED BY

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NEVADA APPLIED ECOLOGY GROUP
UNITED STATES
ENERGY RESEARCH & DEVELOPMENT ADMINISTRATION
LAS VEGAS, NEVADA

5-4

CONF-761065--

TRANSURANICS IN NATURAL ENVIRONMENTS

A SYMPOSIUM AT GATLINBURG
OCTOBER 1976

Nevada Applied Ecology Group
U.S. Energy Research & Development Administration

PLUTONIUM CONTAMINATION OF VEGETATION IN DUSTY FIELD ENVIRONMENTS

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ABSTRACT

Transport of plutonium in food chains of grazing animals and mankind by vegetation carriers becomes an important avenue of contamination in dusty field environments. Findings indicate that most of the activity present in vegetation of such areas at the Nevada Test Site (NTS) is superficial contamination resulting from the attachment of particles to foliage surfaces during resuspension. We suspect, however, that the root uptake pathway eventually will become more significant as the result of natural concentration and recycling processes at work in the field within the plant root zone.

INTRODUCTION

Our assignment for this symposium is the consideration of the problem of plutonium contamination of vegetation in dusty field environments. In accepting this challenge, we acknowledge several timely reviews and discussions concerning various aspects of transuranic element behavior in ecosystems which have appeared in recent literature (Bernhardt and Eadie 1976; Brown 1976; Carfagno and Westendorf 1973; Dahlman et al. 1976; Francis 1973; Hakonson 1975; Hanson 1975; Healy 1974; Mullen and Mosley 1976; Price 1973; Romney and Davis 1972; Stannard 1973). We shall address the problem of vegetation-carrier transport of plutonium in relation to certain radionuclide cycling processes at work in the desert environment as illustrated in Fig. 1. The pathways of ingestion and inhalation are included in this illustration to give symmetry to the overall problem, but discussion of them is assigned to other participants in this symposium.

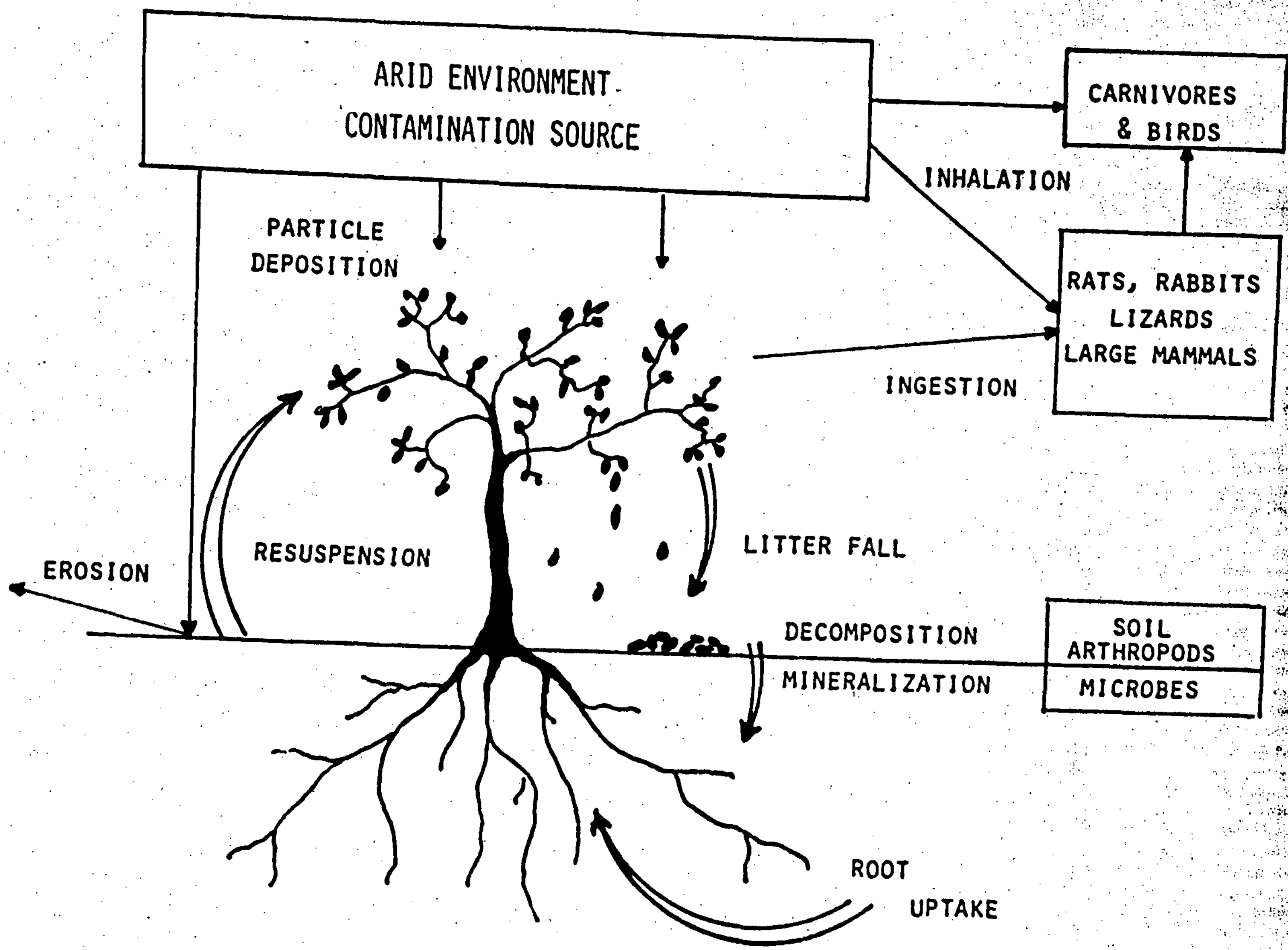

Fig. 1. Simplified illustration of pathways of natural cycling and concentration processes at work in the desert ecosystem.

AIRBORNE FALLOUT DEPOSITION

Earlier studies of fallout from nuclear weapon tests disclosed the biological significance of superficial contamination on plant surfaces resulting from radioactive airborne particle deposition (Fowler (ed) 1965; Larson et al. 1966; Romney et al. 1963; Russell (ed) 1966). Radioactive materials may reach the tissue of plants in two principal ways: First, airborne materials may be deposited upon the above-ground parts of plants and either adhere to their surfaces or be absorbed. Second, materials which have entered the soil may be absorbed by roots along with the nutrients on which plants depend for growth (Russell (ed) 1966). The significance of the root uptake-pathway is governed largely by the biological availability of each given radionuclide during its interactions in soil. Thus, if radioactive materials are insoluble they will most likely contaminate plant tissues only superficially (Romney et al. 1963; Martin 1965).

Early studies done by the UCLA groups on vegetation in fallout contaminated areas at the Nevada Test Site indicated that several natural processes influenced the fate and persistence of fallout debris both from nuclear and non-nuclear contamination events. The period during and shortly after particulate deposition was characterized by conditions of instability largely controlled by wind activity. Rainfall, snow, and periods of calm air movement near ground level hastened later development of a quasi-stable condition wherein particulate movement occurred primarily through processes governing resuspension. Vegetation contamination could be prevented or markedly reduced by protective covering during the unstable and quasi-stable periods after fallout had been deposited (Rhoads et al. 1971; Romney et al. 1971). Measurements of fallout particles on soil and plant material sampled within downwind fallout patterns showed that a partitioning into different sized particles normally occurred during initial fallout deposition. Thus, the mean particle size generally decreased at greater distances downwind from ground zero (Larson et al. 1966).

More recent studies by the NAEG in aged fallout areas at NTS and Tonopah Test Range (TTR), where plutonium was dispersed by chemical explosives, give evidence that this partitioning and patterning has continued to be reflected in the superficial contamination of the indigenous vegetation (Romney et al. 1974, 1975, 1976a, 1976b). The vegetation-to-soil inventory ratios determined in the various activity strata within several different fallout patterns seem to show that a greater proportion of the deposited $^{239-240}Pu$ source material has moved onto vegetation at greater distances away from ground zero. Examples are given in Table 1. Inasmuch as the activity entrapped on plant foliage primarily represents material in the resuspendable particle size range, the amount of contamination on foliage is less in proportion to the total amount of fallout activity deposited on soil at points nearer to ground zero compared to points farther away. Autoradiographs of annual plant leaf tissues collected near ground zero at Area 13 in 1976 showed that the activity present on vegetation was still in discrete particles of suspendable size range nearly 20 years after fallout had occurred (Wallace, unpublished data).

Table 1. Estimated Inventory of $^{239-240}Pu$ for Vegetation in Activity Strata of Aged Fallout Areas at NTS (Romney et al. 1976). Higher numbered activity strata are nearer ground zero.

Activity strata	n	Mean ± S.E. (n Ci/g dry)	Mean[a] ± S.E.[b] (n Ci/m²)	Inventory ± S.E. (millicuries)	Percent	Veg. invent. / Soil invent. ± S.E.[c]
NTS AREA 11, SITE A (1956)						
1	12	.0015 ± .00057	.76 ± .35	.095 ± .045	95	.0029 ± .0018
2	18	.00064 ± .000099	.33 ± .11	.0026 ± .00082	3	.0050 ± .0020
3	6	.0010 ± .00038	.54 ± .24	.00026 ± .00011	0.3	.00031 ± .00033
Total	36			.098 ± .045	100.3	.0028 ± .0017
NTS AREA 11, SITE B (1956)						
3	11	.10 ± .024	60 ± 40	.58 ± .14	24	.0020 ± .00085
2	14	.19 ± .053	110 ± 31	.78 ± .23	33	.00049 ± .00019
4	19	.57 ± .087	330 ± 52	1.0 ± .16	43	.00023 ± .000055
Total	44			2.4 ± .31	100	.00039 ± .000058
NTS AREA 11, SITE C (1956)						
2	12	.14 ± .026	61 ± 12	1.2 ± .23	27	.0018 ± .0014
3	14	.34 ± .054	160 ± 25	1.0 ± .16	24	.0018 ± .00052
4	17	1.1 ± .41	490 ± 180	1.9 ± .72	44	.00035 ± .00013
5	5	1.2 ± .38	530 ± 170	.21 ± .069	5	.000088 ± .000049
Total	48			4.3 ± .78	100	.00048 ± .000089
NTS AREA 11, SITE D (1956)						
2	12	.17 ± .033	72 ± 14	2.5 ± .48	35	.0016 ± .00034
3	13	.24 ± .079	99 ± 33	1.4 ± .46	19	.00045 ± .00025
4	20	.72 ± .14	300 ± 58	1.7 ± .32	23	.00031 ± .00011
5	11	1.3 ± .21	550 ± 89	1.7 ± .28	23	.00020 ± .000088
Total	56			7.3 ± .79	100	.00039 ± .000084

[a] Mean (n Ci/m²) ≅ (mean biomass in g/m²) x mean $^{239-240}Pu$ concentration in n Ci/g

[b] $S.E. \cong [(\text{mean g/m}^2)^2 \times \text{Var (mean n Ci/g)} + (\text{mean n Ci/g})^2 \times \text{Var (mean g/m}^2) - \text{Var (mean n Ci/g)} \times \text{Var (mean g/m}^2)]^{1/2}$

[c] $S.E. \cong R\left[\frac{\sigma^2_x}{x^2} + \frac{\sigma^2_y}{y^2} - \frac{2r\sigma_x\sigma_y}{xy}\right]^{1/2}$ where R = x/y; r = estimated correlation between x, y

Studies of airborne particulates around a contaminated area at the Rocky Flats Plant yielded an average resuspension factor of 10^{-9} m^{-1} during an 8-month sampling period. This factor was near 10^{-6} m^{-1} for particulate material collected upon sticky paper exposed to suspendable fine soil particles from the soil surface. Size distribution studies of all suspended particles containing plutonium indicated a geometric mean diameter of about 10 μm (Volchok 1971, 1972). Results from cyclone and elutricator samples indicated median diameters of about 5 μm. Additional measurements by Sehmel and Orgill (1973) and Sehmel and Lloyd (1974, 1975) in the Rocky Flat area gave resuspension factors ranging from 10^{-9} to 10^{-5} m^{-1}. The Rocky Flats area is subject to winds which are occasionally very strong and gusty. Environmental surveillance data strongly indicate that the movement of contaminated soil particles by wind was a major force causing the original dispersion of plutonium from a barrel storage area (Krey and Hardy 1970; Whicker et al. 1973). The initial source of most of the outlying contamination at Rocky Flats was from leakage of cutting oil containing plutonium. Subsequent studies by Nathans and Holland (1971) of the transformation from this source to suspendable material led to the concept that the resuspendable "hot" particles are agglomerates of small plutonium-containing particles and larger soil grains. Environmental studies at Rocky Flats by the group at Colorado State University have shown some movement of plutonium into several ecosystem compartments within study plots. Data indicate some 96 to 98 percent of the plutonium in the ecosystem is associated with the 0-3 cm depth of soil. About 1 to 3 percent appears to be associated with root and litter samples, while the amounts in standing vegetation and small mammals are generally less than 1 percent. Findings indicate that most of the plutonium present in vegetation samples is superficial contamination resulting from the attachment of particles to biological surfaces. There is also some indication that grass species have higher concentration of plutonium than other species, again probably as the result of the greater surface area per unit mass in the grasses (Whicker 1973; Little 1976).

Healy (1974) completed a noble task of synthesizing into a significant treatise some of the early air sampling data obtained during the safety shots at NTS, where plutonium was dispersed by chemical explosives. Data are given and literature cited which show resuspension factors ranging from 10^{-10} to 10^{-4} m^{-1} for arid or semiarid environments. The higher values represent more unstable, outdoor conditions. Langham (1969) used a value of 10^{-6} m^{-1} in assessing limits for a weapons accident of the type simulated by the safety tests in which plutonium was dispersed at NTS. A continuing study of resuspension at NTS test areas is underway by elements of the Nevada Applied Ecology Group (Anspaugh et al. 1974, 1974a, 1974b, 1974c, 1975). Resuspension factor values calculated from recent experiments now fall within the range of 10^{-11} to 10^{-9} m^{-1} compared to values of 10^{-10} to 10^{-6} m^{-1} measured about 20 years ago at some NTS sites. Most of the total mass of suspendable material is found between diameters of 0.7 μm and 15 μm (Bretthauer et al. 1974; Shinn and Anspaugh 1975). Studies by Tamura (1974, 1975, 1976), have shown that from 50 to 75 percent of the plutonium in soil samples collected from an aged fallout area at NTS is predominantly associated with the coarse silt (20-53 μm) fraction. Ten percent or less usually is associated with the soil fraction less than 5 μm diameter. Highest activities also are associated with heavy mineral fractions (> 2.9 g/cm^3), suggesting the presence of oxides.

Fallout particles intercepted by plant foliage are principally of sizes smaller than 44 μm diameter (Romney et al. 1963). The foliage of vegetation at NTS selectively traps these small-sized particles in the matted hairs and crevices and on resinous glands of the leaf surfaces. The capacity for retaining fallout particles largely depends upon the mechanical-trapping characteristics of plant surfaces. We believe that most of the plutonium contamination now found on vegetation sampled from these aged plutonium fallout areas at NTS is superficial contamination from particulate material. This mechanism of contamination is presently the most important route through which plutonium enters the food chain of grazing animals. However, the relative importance of superficial contamination compared to the root uptake route may diminish with passing time for reasons to be discussed shortly. Fortunately the resuspension factors and vegetation biomass are not great at these sites. The undisturbed desert soil remains quite stable partly because most of the suspendable surface material was removed by wind and water erosion ages ago. Resuspension problems now arise particularly when the stabilized soil surface crust is mechanically disturbed. That is one of the reasons that recommendations have been made to avoid disturbing these aged fallout areas (Wallace and Romney 1975; Rhoads 1976). The purpose for this caution is evident from the data in Table 2, which summarizes values for the estimated inventory of

Table 2. Summary of Estimated Inventory of $^{239-240}Pu$ for Vegetation in Aged Fallout Areas (Romney et al. 1976)

Fallout Area	n	Inventory ± S.E.[a] (millicuries)	Vegetation invent. / Soil inventory ± S.E.[a]
NTS Area 5	113	.86 ± .14	.00034 ± .000046
NTS Area 11A	36	.098 ± .045	.0028 ± .0017
NTS Area 11B	44	2.4 ± .31	.00039 ± .000058
NTS Area 11C	48	4.3 ± .78	.00048 ± .000089
NTS Area 11D	56	7.3 ± .79	.00039 ± .000084
NTS Area 13	141	26.4 ± 3.5	.00060 ± .000076
TTR DT	48	.62 ± .19	.00012 ± .000036
TTR CS1	53	.67 ± .34	.00013 ± .000078
TTR CS2	63	4.3 ± .63	.00015 ± .000030
TTR CS3	41	4.6 ± 1.2	.00016 ± .000037

Time plutonium was dispersed by chemical explosive: NTS Area 5, 1954-1955; NTS Area 11, 1956; NTS Area 13, 1957; TTR Areas, 1963.

[a] See note c Table 1.

plutonium for vegetation in these fallout areas. Only small amounts of the total quantity of plutonium originally deposited presently appear to move to the standing vegetation from fenced-in fallout contaminated soils which are now undisturbed, except for natural wind conditions. Advantage can be gained from keeping these sites as little disturbed as possible. This includes surface erosion by water (Hakonson et al. 1976; Hakonson and Nyhan 1976).

RECYCLING OF PLUTONIUM TO VEGETATION

Most of the standing biomass of vegetation in the aged fallout areas at NTS is contributed by deciduous shrubs (2,000 to 6,000 kg/hectare) which normally yield about 10 percent of their total weight as new annual foliage. The production of grasses, forbs and annual plant species is spasmodic from year to year, depending upon rainfall and climatic conditions. Seldom, however, does the productivity of these annual species exceed 1 percent of the standing shrub biomass. As the result, only from 200 to 600 kg/hectare of new plant foliage is potentially available to undergo the processes of litter fall, decomposition and mineralization. Most of the fallen litter is moved about by wind action to lodge underneath sheltering shrub clumps where much of the initial breakdown is carried out by consumer organisms. Very little is known about the impact of soil arthropods and micro-organisms on plutonium in these areas at NTS, but deductions from well known effects of such organisms on inorganic nutrient elements during mineralization processes would indicate that they should help increase the biological availability (solubility) of plutonium with passing time. The relationships of microbial processes to the fate of transuranic elements in soil is discussed by Wildung et al. (1977, this volume).

First glance at the data in Table 2 gives the impression of an insignificant amount of plutonium in vegetation of the fallout area. However, when one considers that this contaminated plant foliage goes through annual cycles of litter fall and decomposition and mineralization in concentrated areas underneath shrubs, and that the plutonium will persist for tens of thousands of years, one must recognize the possibility of an increasing significance this cycling process will have on increasing root uptake with passing time. The significance of this concentration process should be seen first with ^{241}Am because of its greater biological availability (Romney et al. 1976b). Studies on the difference in edaphic properties underneath shrub clumps and in adjacent bare soil areas at NTS show that highly significant concentration processes have been at work increasing the levels of plant nutrients and organic matter in the root zone underneath shrubs (Romney et al. 1973a, 1973b). Hanson (1975) recentl discussed what appears to be differential biological availability and concentration between ^{238}Pu and ^{239}Pu in soils, vegetation and animal components of the Trinity Site ecosystem. He cited the data of Hakonson and Johnson (1973) showing that ^{238}Pu/^{239}Pu ratios increased from 0.05 (soils) to 0.10 (plants) to 1.0 (mammals), respectively.

RESUSPENSION IN CONTAMINATED AREAS

Once radioactive material has been deposited upon soil, the main concern is to control subsequent resuspension, especially in dry, dusty areas. Whether or not the contaminating radionuclide remains in an original particulate form or undergoes chemical and physical transformations which result in its being carried by soil particles may have no particular biological consequence. Either form deposited upon the surfaces of plants becomes a source of contamination in the diet of grazing animals. The mechanisms of resuspension and its consequences are discussed by other participants in this symposium. The subject of transuranic resuspension and the need for standards for controlling health effects from plutonium in soils is discussed in a recent treatise by Healy (1974).

Optimum conditions for resuspension usually are found in arid, dusty environments, but such movement of contaminated particulate material is not necessarily limited to those areas. In follow-up studies after the accident at Palomares, Spain, Iranzo (1968) called attention to the entrapment of plutonium by external foliage of agricultural crops harvested from contaminated soil. About one half of the contaminant could be removed by washing the foliage of tomato plants compared to from 73 to 95 per cent removed by washing fruit. Deposition on the surfaces of leaves and stems has been identified as the principal mechanism of plutonium contamination of vegetation collected from sampling locations around and adjacent to the Savannah River Plant which is situated in a humid area (McLendon et al. 1976). Some of this contamination could arise either from direct fallout deposited downwind of the source stack or from subsequent resuspension of contaminated soil. A study to measure resuspension during field preparation and planting of winter wheat at the SRP site was reported by Milham et al. (1976). Small, but detectable amounts of airborne plutonium averaged 210 fCi/m^3 at 7.6 m and 10 fCi/m^3 at 30.5 m distances downwind from the edge of the field under cultivation. The air at the tractor operator's face level contained 49 fCi/m^3. The average concentration of plutonium in the 0-5 cm layer of soil was 3100 fCi/g; estimated resuspension factors were of the order of 10^{-8} m^{-1}.

Work by the Los Alamos group at the Trinity site in New Mexico showed that the horizontal distribution of plutonium in soil continued to be largely determined by the original fallout deposition pattern. The Trinity soil samples contained elevated amounts of plutonium in coarser size fractions (> 105 μm) near ground zero and relatively larger amounts in finer fractions at increasing distances from ground zero (Hakonson and Nyhan 1976). Contamination of grasses within the fallout pattern, as a function of distance downwind from ground zero, generally followed the pattern observed in the 0-5 cm soil core fraction. The plutonium concentrations in grasses, lichens and mosses were consistently elevated above the levels observed in forb, shrub and tree samples. This was considered to reflect higher entrapment efficiency for grasses because of greater tissue surface areas (Hakonson and Johnson 1973; Hakonson and Nyhan 1976).

ROOT UPTAKE PATHWAY

Most soil-plant studies focusing upon plutonium movement through plant roots indicate relatively low uptake through this pathway. Concentration ratios for plants grown in potted soil ranged from 10^{-8} to 10^{-3} (Plant Panel 1975). However, in view of the long half-life of plutonium, one is plagued with nagging questions concerning the extent to which the transuranics will become more biologically available through this pathway with passing time. Some evidence already has appeared indicating an increased relative availability (or mobility) of ^{238}Pu and ^{241}Am in given situations (Essington et al. 1976; Hakonson and Johnson 1973; Romney et al. 1975).

It is difficult to differentiate between the amounts of plutonium in vegetation attributable to root uptake and superficial contamination. Attempts, therefore, have been made to accomplish this by pot uptake experiments conducted under air-filtered, glasshouse conditions. Table 3 summarizes some results obtained from soils collected from the aged fallout areas at NTS. Use is made of the Pu/Am ratio in soil and plant samples and also the concentration ratio (CR) for comparing data representing field and glasshouse conditions. The term, CR, is a simple concentration ratio calculated from the activity/g. plant divided by activity/g. soil. For the field data the soil activity is that contained in the 0-5 cm surface layer; the glasshouse soil activity is based on thoroughly blended, potted soil. We are not, therefore, comparing the same conditions in the two systems because the roots of field-grown plants should feed much deeper in the soil; certainly below the top 5 cm layer. What makes this field data most useful in Table 3 is that the 0-5 cm soil Pu/Am ratio more nearly reflects that ratio for the resuspendable material deposited on the leaf surfaces. Considerable difference exists between the plutonium contents in the different soils tested. We should also point out that the field data are overall ratios and CR values obtained from many samples analyzed from the field ($n > 30$) while the glasshouse values are means of six replicates. The important findings from these data are the similarities of the Pu/Am ratios for field soil and vegetation samples which we believe shows principally a superficial source of contamination on vegetation under field conditions. In addition, the calculated CR values for plutonium in the field ranged from 10^{-3} to 10^{0} compared to 10^{-6} to 10^{-3} obtained from our pot uptake tests through the root uptake pathway. The Pu/Am ratios in pot-grown vegetation are much lower than those in the potted soil, indicating uptake of ^{241}Am in much greater amounts than $^{239-240}Pu$. Other data from these tests (Romney et al. 1976d) have shown that synthetic chelating compounds and acidulation agents added to soil significantly (P .05) increase root uptake of these transuranics.

Table 3. Summary of $^{239-240}Pu$ and ^{241}Am Ratios and Pu CR Values for Vegetation and Soil Under Field Conditions Compared to Root Uptake Experiment Under Glasshouse Conditions

Soil Source	Field Conditions			Glasshouse Conditions		
	Soil Pu/Am	Plant Pu/Am	Pu CR**	Soil Pu/Am	Plant Pu/Am	Pu CR*
NTS Area 11B	7.7 ± 0.14	8.5 ± 1.0	1.3E-2 to 1.6E-1	6.5 ± 0.17	0.83 ± 0.11	1.5E-4
NTS Area 11C	6.0 ± 0.08	5.2 ± 0.1	4.5E-2 to 3.4E-1	5.3 ± 0.17	0.35 ± 0.13	1.8E-4
NTS Area 11D	5.8 ± 0.15	4.1 ± 0.18	2.7E-2 to 1.7E-1	5.2 ± 0.12	0.34 ± 0.12	1.1E-3
NTS Area 13	9.4 ± 0.15	7.9 ± 0.2	7.8E-2 to 4.4E-1	5.6 ± 0.27	0.19 ± 0.03	1.1E-4
TTR DT	23.5 ± 0.73	15.8 ± 1.4	1.1E-2 to 9.4E-2	21 ± 0.67	0.97 ± 0.72	2.6E-4
TTR CS1	26.0 ± 0.76	16.2 ± 0.52	7.4E-3 to 4.2E-2	19 ± 0.33	0.57 ± 0.36	4.3E-4
TTR CS2	22.2 ± 0.41	11.6 ± 0.64	1.4E-2 to 9.2E-2	21 ± 1.9	6.3 ± 1.3	7.6E-4
TTR CS3	22.0 ± 0.28	17.0 ± 0.93	3.9E-2 to 5.3E-2	19 ± 0.67	1.9 ± 0.26	5.5E-4

* Data given are for soybean foliage. Seed CR values ranged from 10^{-6} to 10^{-4}. Data are summarized from Romney <u>et al</u>. (1975, 1976).

** Range of mean CR values calculated for activity strata in fallout area. CR values for individual collection sites ranged from 10^{-3} to 10^{0}.

CONCLUSIONS

Two principal incorporation mechanisms are involved in the vegetation-carrier transport of plutonium in the diet of grazing animals and mankind: (1) superficial entrapment of particulate material with possibilities of foliar absorption of soluble contaminant, and (2) root uptake of the contaminant entering soil. It is important to reduce this transport of plutonium as much as possible. Findings from studies in dusty field environments at NTS indicate that superficial contamination is presently the most important route. However, certain natural cycling and concentration processes are underway which, in terms of the long half-life of plutonium, should gradually increase the importance of the root uptake pathway. We suspect that even now the root uptake pathway has increasing importance relative to superficial contamination in humid ecosystems, except at sites subject to local fallout material from such sources as emission stacks of processing facilities. The aged plutonium fallout areas at NTS will continue to be of value as sites in which to make periodic assessments to detect changes in the cycling and concentration of plutonium in various components of the ecosystem. Because of its greater solubility, ^{241}Am at these sites may show an earlier indication of such changes. We believe it always will be important to avoid disturbances that will increase resuspension and erosion in plutonium contaminated sites at NTS and elsewhere. In their present natural states we believe that the plutonium contaminated sites at NTS and TTR present no radiological hazard to grazing animals and mankind so long as residence within the fenced exclusion areas is prohibited.

ACKNOWLEDGMENTS

Work was conducted under Contract E(04-1) GEN-12 between the United States Energy Research and Development Administration and the University of California. Supplemental support was provided by the Nevada Applied Ecology Group, Bioenvironmental Science Division, USERDA Nevada Operations Office. We are indebted to members of the NAEG management and sample collection, preparation and radiochemical analysis teams for their devoted efforts in support of this work.

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