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AN APPRAISAL OF AVAILABLE INFORMATION ON UPTAKE BY PLANTS OF TRANSPLUTONIUM ELEMENTS AND NEPTUNIUM

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ABSTRACT

A critical review was made of reported information from laboratory studies of plant uptake of transplutonic elements plus neptunium. The available data are meager but indicate that the uptake of Np is the greatest followed by Am and Cm. The data are not sufficient to provide recommended values for use in hazard calculations but they do indicate that the actinides other than plutonium will be accumulated in plants to a greater degree than plutonium.

INTRODUCTION

A potentially important parameter in assessing hazards from materials released to the environment is the possibility of such materials entering the human food chain through uptake by plants from soils. With the forecasted potential use of nuclear energy, it is anticipated that small quantities of transuranic elements will be released to the environment. In planning for the future and to assure that proper precautions will be exercised, it is important to have information on the behavior of those elements in soil-plant systems.

It has frequently been assumed that all actinides behave like plutonium in the environment; however, the chemical properties of some differ significantly from those of plutonium. Initial experiments with plants, as well as studies with animals, confirm that these elements behave differently in living systems and that direct comparison with plutonium may result in errors in estimating human hazards. This report reviews the available experiments to provide the best information for assessing this pathway.

In this study, only uptake and incorporation into plant tissue (or in some cases absorption onto roots) from the growing medium has been considered. A sizable body of data exists which indicates that uptake of this nature may be insignificant in the field when compared to direct accumulation and retention on plant surfaces. Any accumulation is dependent, of course, upon climatic conditions and the particular plant type in question, and is expected to vary with those parameters. However, it is not clear at this time as to whether the uptake and retention of the metabolized isotope in animals will be the same as that deposited directly on the plant.

ACTINIDE ELEMENTS ASSOCIATED WITH PLUTONIUM FUEL CYCLES

The potential environmental importance of transuranic nuclides associated with plutonium fuel cycles depends not only upon their apparent behavior in the environment but also upon their abundance in the environment. Thus, in assessing the relative hazard one must consider not only the uptake of each element but also the relative quantity of each of the isotopes.

Table I lists the abundance of a number of the longer lived nuclides of importance relative to ^{239}Pu at three times after discharge. These data were calculated from the levels reported by Bell[1] for nuclides in spent fuel discharged from the Atomics International Reference Oxide LMFBR. While other reactors will have somewhat different ratios, these values will indicate the relative abundances in a, more or less, typical fuel. It must be realized that, for the environmental situation, these ratios may be altered depending upon the process used in separation of the U and Pu and the point in the process where releases may occur.

UPTAKE OF AMERICIUM, CURIUM, AND NEPTUNIUM IN PLANTS

A series of experiments has been performed by Price[2-6] which allows comparison of plant uptake of Am, Cm, Np, and Pu. The first study was performed to determine uptake of these actinides in the nitrate form, and served as a basis for subsequent studies on the influence of various complexing materials on uptake. Soil, plant types, and methods used were constant for the series of experiments. The soil used was Burbank loamy sand local to the Hanford, Washington area. Two introduced and locally growing plant varieties, tumbleweed and cheatgrass, were grown for two months, harvested, dried, and analyzed by liquid scintillation. One-quart plastic-lined pots were each packed with 1 kg of dry soil, and 50 μCi

TABLE I

CHARACTERISTICS OF TRANSURANIC NUCLIDES
ASSOCIATED WITH PLUTONIUM FUEL CYCLES

Isotope	Half-Life	Specific Activity (Ci/g)	Type of Decay	Abundance Relative to ^{239}Pu		
				3y	10^2y	10^3y
^{236}Pu	2.85y	5.32×10^{2}	α	3.30×10^{-4}	0	0
^{238}Pu	86y	1.75×10^{1}	α	6.79×10^{0}	3.13×10^{0}	3.04×10^{-3}
^{239}Pu	2.44×10^{4}y	6.13×10^{-2}	α	10^{0}	10^{0}	10^{0}
^{240}Pu	6580y	2.26×10^{-1}	α	1.66×10^{0}	1.65×10^{0}	1.53×10^{0}
^{241}Pu	13.2y	1.12×10^{2}	β^-	1.93×10^{2}	1.19×10^{0}	0
^{242}Pu	3.79×10^{5}y	3.90×10^{-3}	α	4.77×10^{-3}	4.79×10^{-3}	4.89×10^{-3}
^{241}Am	458y	3.24×10^{0}	α	1.49×10^{0}	6.22×10^{0}	1.65×10^{0}
^{242}Am	16h	8.09×10^{5}	β^-	3.12×10^{-2}	2.01×10^{-2}	3.40×10^{-4}
^{242m}Am	152y	9.76×10^{0}	α	3.12×10^{-2}	2.01×10^{-2}	3.40×10^{-4}
^{243}Am	7950y	1.85×10^{-1}	α	1.83×10^{-2}	1.83×10^{-2}	1.73×10^{-2}
^{242}Cm	163d	3.31×10^{3}	α	2.81×10^{-1}	2.02×10^{-2}	3.42×10^{-4}
^{243}Cm	32y	4.60×10^{1}	α	1.30×10^{-2}	1.59×10^{-3}	0
^{244}Cm	17.6y	8.33×10^{1}	α	4.04×10^{-1}	8.90×10^{-3}	0
^{237}Np	2.14×10^{6}y	7.05×10^{-4}	α	1.86×10^{-5}	1.95×10^{-3}	1.22×10^{-3}
^{235}Np	2.1d	2.61×10^{5}	β^-	0	0	0
^{239}Np	2.35d	2.33×10^{5}	β^-	1.83×10^{-2}	1.82×10^{-2}	1.73×10^{-2}

$^{237}Np(NO_3)_5$, 50 µCi $^{239}Pu(NO_3)_4$, 25 µCi $^{241}Am(NO_3)_3$ or 25 µCi $^{244}Cm(NO_3)_3$ in 0.33 N HNO_3 was evenly distributed on the surface from a 10-ml pipette. This was allowed to dry overnight at room temperature and 1-cm layer of soil was added. Six seeds, later thinned to three tumbleweed or four cheatgrass seedlings, were planted and covered with 2 cm of coarse sand. Nitrogen, as NH_4NO_3, was provided and the total, including that in the HNO_3 spike, was equivalent to 172 µgN/g of soil. The cation exchange capacity of the soil was reported as 6.4 meq/100 g, organic matter as 0.3% and pH at saturation as 7.8 prior to adding radionuclides. Harvested plants were dried for 24 hours in paper bags in a forced draft oven at 60° C. Dried plant tissue was pre-ashed in H_2SO_4 and ashed in a muffle furnace. One hundred-mg samples of ash were dissolved in 2 ml of 2 N HNO_3 with heat. A scintillation gel was added and analyses were made using an ambient temperature liquid scintillation counter. Data were presented as average uptake of the nuclide added to each pot, and as average concentration factor (CF) defined as

$$CF = \frac{\mu Ci/g \text{ oven dry plant}}{\mu Ci/g \text{ oven dry soil}} ,$$

where the weight of oven dry soil actually contaminated was determined to be about 40 g. This value was determined by adding 10 ml of solution to air-dried soil and removing the wetted soil as soon as flow was noted to have ceased. It was assumed that addition of the nuclide in this manner for the uptake studies would result in the same 40-g value.

In subsequent experiments[3,4,6] Price studied the effect of organic acids, EDTA and DTPA, on uptake of ^{241}Am, ^{244}Cm, ^{237}Np, and ^{239}Pu. The organic acid salts, ammonium acetate, sodium glycolate, ammonium oxalate, and ammonium citrate, were prepared in dilute (0.15M) solutions to which ∿2.5 µCi of Np, Cm, or Am/ml or ∿5 µCi/ml was then added. Due to the specific activities of the isotopes (see Table I), the molar ratios of organic acid salt-to-nuclide were different by a factor of about 10^5. Approximately 10 ml of these solutions or of the nuclides chelated with EDTA or DPTA (amounts not known) were added to pots as described for the nitrates. Soil pH's determined after addition of the contaminated solutions were 5.1 for the nitrate treatment, 7.4 to 8.5 for the acetate, 6.6 to 9.1 for the glycolate, 6.5 to 8.6 for the oxalate, and 6.8 to 8.3 for the

citrate. Where a range is given, the lower pH corresponded to Np and the higher to Pu and Cm with Am values slightly less than the higher value. Reported pH values were from the treated soil, estimated by the author to represent about a 5-mm layer of soil within the pot.

The growth period was two months in all cases. Five replicates were prepared for the nitrates and three for the organic acid complexes. The number of replicate pots planted for the chelates was not reported. Concentration factors for all soil treatments are shown in Table II.

Price[5] also reported ratios of root nuclide concentration to shoot nuclide concentration for the nitrate solutions, and taking those values as unity, he reported relative root-to-shoot ratios for EDTA and DTPA treatments. Since a method for cleaning the plant parts prior to analysis was not reported, the extent to which root concentrations represent adsorption as opposed to absorption is not known. Reported data, which were based on nuclide concentrations in plant ash, are shown in Table III.

Both chelating agents appear to increase the uptake of Pu, Am, and Cm in the roots to a greater extent than they increase the uptake in the shoots. With Np, which has a high concentration factor as the nitrate, the increase in either root or shoot due to chelating agents is small if it does occur at all. The author concluded that, "In the case of Pu, chelation apparently not only increases plant uptake from soil but also may facilitate transport from root to shoot. Chelation of Am and Cm increases plant uptake but may not have an influence on root-to-shoot transport. Chelation apparently results in reduced uptake of Np into roots or shoots."

The results of this experiment were undoubtedly affected by the technique. Although the plants were grown in about 1 kg of soil, the actinide was applied to only about 40 g of soil and the latter value was used in the calculation of concentration factors. It is not clear whether the same result would have been obtained if the soil were uniformly contaminated with the same amount of actinide although to be the same, the following conditions must be fulfilled in both systems: (1) The absorption through the root must be the same in all volumes of the soil implying, essentially, uniform distribution of the active root area through the mass; and (2) the rate of absorption must be linear with concentration of the nuclide. Differential movement of the nuclide due to movement with watering was not measured but if the above conditions were not met and there was a differential

TABLE II

CONCENTRATION FACTORS FOR UPTAKE OF ^{237}Np, ^{239}Pu, ^{241}Am, AND ^{244}Cm IN TUMBLEWEED AND CHEATGRASS PLANTS (ABOVEGROUND PARTS)

DATA ARE MEAN ± 1 STANDARD ERROR OF MEAN[a]

	Nitrate	Acetate	Glycolate	Oxalate
Tumbleweed				
^{237}Np	$1.1\times10^{-1}\pm2.0\times10^{-2}$	$2.4\times10^{-1}\pm5.0\times10^{-2}$	$2.3\times10^{-1}\pm7.0\times10^{-2}$	$2.8\times10^{-1}\pm5.0\times10^{-2}$
^{239}Pu	$4.6\times10^{-5}\pm7.0\times10^{-6}$	$4.8\times10^{-5}\pm4.0\times10^{-6}$	$2.5\times10^{-4}\pm6.0\times10^{-5}$	$2.7\times10^{-4}\pm5.0\times10^{-5}$
^{241}Am	$1.4\times10^{-3}\pm2.0\times10^{-4}$	$1.7\times10^{-3}\pm3.0\times10^{-4}$	$2.1\times10^{-3}\pm4.0\times10^{-4}$	$1.5\times10^{-3}\pm3.0\times10^{-4}$
^{244}Cm	$2.2\times10^{-3}\pm3.0\times10^{-4}$	$1.2\times10^{-3}\pm1.0\times10^{-4}$	$4.2\times10^{-3}\pm7.0\times10^{-4}$	$1.5\times10^{-3}\pm3.0\times10^{-4}$
Cheatgrass				
^{237}Np	$1.2\times10^{-2}\pm1.0\times10^{-3}$	$1.5\times10^{-2}\pm3.0\times10^{-3}$	$1.3\times10^{-2}\pm2.0\times10^{-3}$	$7.0\times10^{-3}\pm1.0\times10^{-3}$
^{239}Pu	$1.7\times10^{-5}\pm2.0\times10^{-6}$	$1.4\times10^{-5}\pm3.0\times10^{-6}$	$4.3\times10^{-5}\pm4.0\times10^{-6}$	$5.3\times10^{-5}\pm5.0\times10^{-6}$
^{241}Am	$6.0\times10^{-4}\pm1.0\times10^{-4}$	$2.3\times10^{-4}\pm2.0\times10^{-5}$	$8.0\times10^{-5}\pm1.0\times10^{-5}$	$6.0\times10^{-5}\pm1.0\times10^{-5}$
^{244}Cm	$4.8\times10^{-4}\pm5.0\times10^{-5}$	$3.3\times10^{-4}\pm3.0\times10^{-5}$	$1.9\times10^{-4}\pm2.0\times10^{-5}$	$8.0\times10^{-5}\pm1.0\times10^{-5}$

	Citrate	EDTA	DTPA
Tumbleweed			
^{237}Np	$2.8\times10^{-1}\pm2.0\times10^{-2}$	$4.5\times10^{-2}\pm2.0\times10^{-3}$	$5.3\times10^{-2}\pm3.0\times10^{-3}$
^{239}Pu	$3.1\times10^{-4}\pm8.0\times10^{-6}$	$1.7\times10^{-3}\pm3.0\times10^{-5}$	$1.4\times10^{-1}\pm1.3\times10^{-2}$
^{241}Am	$1.5\times10^{-3}\pm2.0\times10^{-4}$	$8.3\times10^{-3}\pm4.0\times10^{-4}$	$5.6\times10^{-1}\pm2.7\times10^{-2}$
^{244}Cm	$1.4\times10^{-3}\pm2.0\times10^{-4}$	$9.7\times10^{-3}\pm13.\times10^{-3}$	$5.6\times10^{-1}\pm7.3\times10^{-2}$
Cheatgrass			
^{237}Np	$1.1\times10^{-2}\pm2.0\times10^{-3}$	$2.5\times10^{-2}\pm3.0\times10^{-3}$	$1.7\times10^{-2}\pm3.0\times10^{-3}$
^{239}Pu	$5.1\times10^{-5}\pm2.0\times10^{-6}$	$3.9\times10^{-4}\pm4.0\times10^{-5}$	$3.1\times10^{-2}\pm3.0\times10^{-3}$
^{241}Am	$1.0\times10^{-4}\pm2.0\times10^{-5}$	$8.0\times10^{-4}\pm1.0\times10^{-4}$	$1.0\times10^{-1}\pm1.8\times10^{-2}$
^{244}Cm	$1.6\times10^{-4}\pm1.0\times10^{-5}$	$1.2\times10^{-3}\pm1.0\times10^{-4}$	$1.5\times10^{-1}\pm9.0\times10^{-3}$

[a]See Price 1975.[6]

TABLE III

RELATIONSHIP OF ROOT AND SHOOT CONCENTRATIONS IN PLANT ASH OF ^{237}Np, ^{239}Pu, ^{241}Am, AND ^{244}Cm IN CHEATGRASS AND TUMBLEWEED PLANTS

	Root-to-Shoot Ratio Relative to Nitrate		
	Nitrate	EDTA	DTPA
Tumbleweed			
^{237}Np	4.0	~0.3	---
^{239}Pu	~600	<0.1	<0.1
^{241}Am	0.5-1.0	5.0	41.0
^{244}Cm	0.5-1.0	21.3	20.0
Cheatgrass			
^{237}Np	4.0	<0.1	---
^{239}Pu	~600	0.3	0.4
^{241}Am	0.5-1.0	68.0	22.0
^{244}Cm	0.5-1.0	58.0	22.0

mobility among the actinides or treated actinides, then a portion of the observed results could have been due to this movement. Martell[7] has noted that, in the field situation, leaching by precipitation and ground water will proceed more rapidly for Am and Cm than for Pu. He, therefore, postulates that such movement away from the root zone may partially offset the increased uptake of these actinides. Particularly with Np, where the ratio of the CF's of the Np to Pu as the nitrate is ∿2400 for tumbleweed and ∿700 for cheatgrass, a portion of this difference could be due to Np moving away from the relatively shallow cheatgrass roots but remaining available to the deeper tumbleweed roots.

The importance of contaminant depth was also noted by Price.[5] He added nitrate and oxalate solutions of the four nuclides at mid-depth of large pots. Tumbleweed and cheatgrass were grown in the pots and harvested after two months. Roots above and below the depth of nuclide addition were collected and analyzed separately. Except for ^{237}Np, only root tissue below the nuclide layer showed radionuclide accumulation. The author stated it was unlikely that the transuranics had leached through the soil to contaminate roots. Since no soil analysis data were reported, it can only be said with certainty that those nuclides did not move upward to contaminate root tissue. It is assumed that ^{237}Np was detected in roots from above and below the contaminant layer, which possibly indicates greater mobility of Np than of the other nuclides and perhaps provides an explanation of the apparent greater uptake of Np by these plants.

Thomas and Jacobs[8] studied uptake of ^{242}Cm by tall fescue grass and giant stringless bean plants. The +3 state of Cm was implied although the chemical compound was not specified. Curium was added to Knop's nutrient solution in a concentration of 5000 dis/min per ml solution, and 200 ml (0.45 μCi ^{242}Cm) of the nutrient solution was used in each growth situation. Young bean plants were grown in polyethylene bottles of a size that held 200 ml of ^{242}Cm-tagged solution or Fullerton loam soil maintained at field capacity with 200 ml of tagged solution (about 1300 gm of soil if field capacity was 15% as noted for Burbank soil).[2] It is assumed that plants were first grown in solution or soil not containing ^{242}Cm and then transferred to the tagged growing medium. Two dozen plants were grown in solution, 16 in soil, and 10 in solution but with the roots removed 1 cm below the root collar. Fescue grass, assumed to be of a mature age, was clipped to 8 cm in height and transplanted to four plastic trays each containing 200 ml of tagged nutrient solution such that the root mat was in the solution. The growth period of all plants in contaminated media was 28 days, during which

time bean plants were reported to have increased in height by a factor of 4 and in weight by a factor of 6 but did not produce fruit. Grass leaves grew to a length of 25 cm. Control plants of both types were grown in uncontaminated nutrient solution and exhibited no differences in growth or vigor compared to experimental plants. Harvest included all grass greater than 4 cm in height and all aboveground parts of bean plants.

Analyses for ^{242}Cm were performed on plant samples weighing about 200 mg (oven dry weight), and results were reported for entire plant parts the weights of which were not reported. Samples were dried for 24 h (105°C), ashed overnight (600° C), dissolved in 1 ml of 1 N HCl, filtered into liquid scintillation counting vials with a 10-ml distilled water rinse, evaporated to near dryness and counted following addition of 20 ml scintillation solution. Control plant samples, experimental plant samples, and standards were prepared similarly and a detection limit of 0.002% uptake per whole plant was determined. The authors reported that grass contained no activity in excess of that in control samples, and a physical or physiological barrier in grass was postulated since grasses do contain other heavy metals.

Results of analyses of bean plants are shown in Table IV. Of 16 plants grown in soil, 3 did not have significant radioactivity. The authors suggested that, based on the apparent relative ease with which ^{242}Cm entered the transpiration system of rootless plants, abrasion of roots by soil and the greater activity of microorganisms in soil than in nutrient solution could have increased uptake by soil-grown rooted plants over nutrient-grown rooted plants. The possible influence of soil microorganisms on transport of radioactive contaminants from soil to plants was not discussed but may be an influence which may not be present in systems using a sterile medium or oven-dried soil. Some studies are currently under way, with preliminary results indicating that plutonium (^{238}Pu), even as an oxide, can be taken up and transported to the spores of Aspergillus niger, implying that certain nuclides could be made more available to the food chain as a result of microbial activity.[9,10]

Bean and barley plants were grown by Cline[11] to determine comparative uptake of ^{241}Am and ^{239}Pu. Bean plants were grown for 18 days in six liters of Hoagland's nutrient to which 1 μCi ^{239}Pu/l or 0.9 μCi ^{241}Am/l was added at 14 days (4 days before harvest).[12] Barley was grown for 18 days [13] in Cinebar clay loam (pH 4.5) or in Ephrata sandy loam (pH 7.5) to which 10 μCi ^{239}Pu/g or 1.8 μCi ^{241}Am/g had been added at the start. One hundred barley plants were grown

TABLE IV

UPTAKE OF ^{242}CM IN GIANT STRINGLESS BEAN PLANTS

Growth Medium	Root Condition	^{242}Cm in Plant (pCi)	Plant Uptake (%)
Solution	Intact	135 ± 4[a]	0.030
Soil	Intact	306 ± 22	0.068
Solution	Severed	387 ± 14	0.086

[a] ± values are 1 standard error.

TABLE V

UPTAKE OF ^{241}AM AND ^{239}PU IN BEAN AND BARLEY PLANTS (CONCENTRATION FACTORS)

Isotope	Bean (Nutrient Solution)	Barley (Cinebar)	Barley (Ephrata)
^{241}Am	3×10^{-3}	3×10^{-3}	2×10^{-3}
^{241}Am[a]	3×10^{-3}	3×10^{-3}	3×10^{-3}
^{239}Pu	6×10^{-5}	2×10^{-4}	1×10^{-4}

[a] From 2% Am contaminant in ^{239}Pu solution.

in 100 g of soil. The nuclide solutions were either essentially pure ^{241}Am or with an activity distribution of 95.1% ^{239}Pu plus 2.9% ^{238}Pu and 2.0% ^{241}Am. Stems and leaves of bean plants were oven-dried for 48 hours at 68°C. Barley leaves were clipped about one-quarter inch above the soil surface (the soil had been covered with 1/4 inch of sand) and oven-dried. Analyses were performed by direct plant tissue counting for ^{241}Am, using a NaI crystal, and by chemical separation and liquid scintillation counting of ^{239}Pu. Plant uptake of the nuclides was reported as concentration factors, activity per gram barley/activity per gram soil, or activity per gram bean plant/activity per ml of solution (Table V). Americium-241 uptake was 15-50 times that of ^{239}Pu. A meaningful comparison of plant types for the two nuclides is not possible because the growing media were different as were the times of exposure to nuclide.

Americium-241 uptake in bush bean leaves was studied by Hale and Wallace,[14] using Hacienda loam (pH 7.5; calcareous) and Yolo loam (pH 5.7; non-calcareous) as growing media. Soils were prepared by mixing the loam with vermiculite one-to-one by volume, and KNO_3 equivalent to 200 pounds N per acre was included.

Bush beans were germinated in sand and seedlings were transplanted to pots containing 500 g of the soil mixture to which 10 μCi ^{241}Am in a volume of ∿50 ml HNO_3 had been added in increments while the pots were being filled so that the distribution through the soil was reasonably uniform. Chelates, DTPA, EDDHA, and FeEDDHA were added to soil in amounts equivalent to 10 pounds Fe per acre. Two pots containing two plants each were grown for each treatment and as controls, for each soil. Leaves were harvested after 14, 31, and 45 days and analyzed separately for each plant in a sodium iodide (NaI) well counter and were then oven-dried and weighed. The approximate weight of leaves was 200 mg per plant.[15] Results, as reported and as derived, are given in Table VI. Counting efficiency was not reported, but it has been determined by discussion with one of the authors that a value of about 8% was used at the time of the experiment and this value was used in estimating the concentration factors.

TABLE VI

UPTAKE OF ^{241}AM IN BUSH BEAN LEAVES

Treatment	Hacienda Loam			Yolo Loam		
	14 Days	31 Days	45 Days	14 Days	31 Days	45 Days
Measured Uptake (Counts $min^{-1} mg^{-1}$)						
Control	0.28	0.2	0.8	0.73	0.3	0.1
DTPA	193.5	17.7	40.7	457.4	19.6	40.5
EDDHA	5.7	0.7	0.2	330	5.3	3.2
FeEDDHA	2.0	0.0	0.1	17	1.9	1.4
Concentration Factors						
Control	0.075	0.054	0.216	0.198	0.081	0.027
DTPA	51	4.8	11.1	123	5.4	10.8
EDDHA	1.53	0.189	0.054	90	1.44	0.87
FeEDDHA	0.54	0	0.027	4.5	0.51	0.39
Relative Effect of Chelating Agents[a]						
DTPA	470	44	100	1100	50	100
EDDHA	14	1.7	0.5	830	13	8
FeEDDHA	5	<1	0.2	42	5	4

[a]Ratio of chelate CF at each sampling time to control CF averaged for three sampling times and two soils.

Table VI indicates a greater influence of DTPA than the other two agents and it appears that the presence of the iron on the FeEDDHA has decreased the uptake over that of the EDDHA for both soils. However, the effect of the chelating agent appears to be somewhat greater for the Yolo loam, a non-calcareous soil. There is also an apparent decrease in effectiveness of the chelating agent following the first cropping although this does not seem to continue further to the third cropping.

Wallace[16] performed a series of experiments to determine the joint influence of DTPA and added micronutrients on ^{241}Am uptake by plants. Soybeans were germinated in sand and transplanted to plastic-lined pots containing 500 g Hacienda loam that had been premixed with 8 μCi ^{241}Am. The loam had been prepared by mixing with an equal volume of vermiculite to facilitate drainage. Nitrogen, as NH_4NO_3, was supplied at a rate of 100 ppm N on a dry soil weight basis. In DTPA treatments, equal molar amounts of Fe and Na_2 DTPA were used, 2 ppm Fe and 13 ppm DTPA. Micronutrients applied with and without Fe + DTPA were 25 or 50 ppm Zn as $ZnSO_4$ and 50 or 100 ppm Mn as $MnSO_4$. Plants were harvested 13 days after transplanting, and stems and leaves were handled separately. Plant tissues were washed in 0.1 N HCl followed by distilled water, dried, ground, weighed, and analyzed in a NaI well counter. The results, means of six replicates, are given in Table VII. In communication with Wallace[15] he indicated that the results, as reported, were calculated using an efficiency for the counter of 8% whereas a value of 25% may be more accurate. However, we have reported the values given by Wallace on the assumption that the spike solution was also measured on this device. It was noted that the plants were all iron chlorotic, as determined by leaf color and only slight improvement was noted as a result of the FeDTPA treatment. Wallace noted the increases in ^{244}Am in the plant caused by the FeDTPA but that the increase was not as great as noted with bush beans for the application without iron.[14] The addition of Zn and Mn resulted in slight decreases in uptake. It is also of interest to note that the increased uptake with chelation was primarily in the leaves. The stems, although at about the same concentration as the leaves in the controls, showed little change with chelation although the addition of Mn apparently decreased the uptake.

The effect of temperature on root uptake by bush beans was investigated by Wallace.[16] Seedlings pregrown for 7 days in 10^{-4} M $CaCl_2$ were transferred to 300 ml tubes containing 10^{-4} M $CaCl_2$, 10^{-4} M DTPA and 26 μCi ^{241}Am. After 48 hours at

TABLE VII

INFLUENCE OF DTPA AND MICRONUTRIENT TREATMENT ON UPTAKE OF ^{241}AM BY SOYBEAN PLANTS

Treatment	Weight		Plant Content		Concentration Factor		
	Stems (mg)	Leaves (mg)	Stems (dis·sec^{-1}g^{-1})	Leaves (dis·sec^{-1}g^{-1})	Stems	Leaves	Total[a]
Control	63	124	130	72	0.22	0.12	0.15
25 ppm Zn	66	116	83	66	0.14	0.11	0.12
50 ppm Zn	70	129	92	30	0.16	0.051	0.089
2 ppm Fe + 13 ppm DTPA	77	152	123	1526	0.21	2.6	1.8
2 ppm Fe + 13 ppm DTPA + 25 ppm Zn	77	151	121	885	0.20	1.5	1.1
2 ppm Fe + 13 ppm DTPA + 50 ppm Zn	67	129	8	820	0.014	1.4	0.93
50 ppm Mn	61	127	26	26	0.044	0.044	0.044
100 ppm Mn	66	106	33	33	0.056	0.056	0.056
2 ppm Fe + 13 ppm DTPA + 50 ppm Mn	62	124	127	891	0.21	1.5	1.1
2 ppm Fe + 13 ppm DTPA + 100 ppm Mn	69	135	77	702	0.13	1.2	0.84

[a]Total CF is weighted average of stems and leaves.

TABLE VIII

EFFECT OF ROOT TEMPERATURE - MEANS OF 6 REPLICATES

Root Temp (°C)	^{241}Am Concentration in Plants (dis·sec^{-1}g^{-1})			Concentration Factor		
	Root	Stem	Leaf	Root	Stem	Leaf
26	1027	65	141	0.32	0.02	0.044
16	882	64	680	0.28	0.02	0.21
6	951	32	420	0.30	0.01	0.13

three different temperatures, the plants were separated into leaves, stems, and roots, and roots were washed in 10^{-3} M Na_2 DTPA. Samples were dried and measured for ^{241}Am. Results are given in Table VIII. Wallace indicated that the differences with temperature are not significant which would indicate that this uptake may not be related to the plant metabolism.

In another experiment with bush beans he placed seedlings in aerated nutrient solution containing 10^{-5} M Na_2 DTPA and about 2 μCi ^{241}Am per jar. After seven days, one set was removed for measurement and two other sets were placed in nutrient jars one with and one without DTPA. Both sets were allowed to grow to maturity (21 days additional) and were then measured. The results indicated that, in the presence of DTPA, the plants retained much of the ^{241}Am in the old growth with, however, some translocation to new leaves. In the absence of DTPA more ^{241}Am was translocated to new growth with much of it coming from old leaf tissue. In both cases there was marked redistribution from old leaves and stems back into the roots. He concluded that DTPA had little effect in promoting translocation.

In a final study Wallace[16] determined the influence of DTPA and RA 157 (the toluene derivative of EDDHA) without iron on ^{241}Am uptake in Valencia orange, scions of which had been whip-grafted to trifoliate orange or rough lemon roots. Seventy days after grafting, single plants were placed in pots containing 500 g Hacienda loam "previously equilibrated with 125 μCi each, in 6000-g lots," or presumably about 10.4 μCi ^{241}Am per pot. At the start of the experiment, 35 ppm of DTPA or RA 157 were added to the soil. Suckers were harvested 85 days after transplanting and shoots above the graft at 119 days. After one year, 135 ppm DTPA was applied to soil of pots containing rough lemon rootstock, including the control. New growth was harvested 30 days following that treatment (13 months from transplanting). Plant tissues were washed with 0.1 N HCl, rinsed, dried, divided into leaves and stems, and analyzed for ^{241}Am using a NaI well detector. Results represented 5-8 replicates at 85 and 119 days, and 3 replicates at 13 months and are given in Table IX.

In discussion of these results, Wallace noted that the DTPA increased the transport of ^{241}Am to the shoots but there was little effect of rootstock. This suggests that the mechanism for absorption of ^{241}Am may be different than that for iron or zinc since large differences had been observed depending on rootstock for these elements. It is of interest that the RA 157 chelating agent had little or no effect on uptake. The increase in uptake following the DTPA application

TABLE IX

UPTAKE OF ^{241}AM BY VALENCIA ORANGE

GRAFTED TO TWO ROOTSTOCKS

Rootstock	Treatment	85-Day Suckers		119-Day Scions			13-Month
		Leaf	Stem	Old Leaf	New Leaf	Stem	New Growth[a]
Plant Concentration (dis•sec^{-1}g^{-1})							
Rough Lemon	None	0	0	4	4	8	79 ± 28[b]
Rough Lemon	DTPA	530	0	1582	3520	178	145 ± 73
Rough Lemon	RA 157	34	22	2	4	13	120 ± 6
Trifoliate Orange	None	3	0	4	2	2	
Trifoliate Orange	DTPA	2260	37	894	788	32	
Trifoliate Orange	RA 157	52	3	6	3	8	
Concentration Factors							
Rough Lemon	None	0	0	0.0052	0.0052	0.010	0.10
Rough Lemon	DTPA	0.69	0	2.1	4.6	0.23	0.19
Rough Lemon	RA 157	0.044	0.029	0.0026	0.0052	0.017	0.16
Trifoliate Orange	None	0.0039	0	0.0052	0.0026	0.0026	
Trifoliate Orange	DTPA	2.9	0.048	1.2	1.0	0.042	
Trifoliate Orange	RA 157	0.068	0.0039	0.0078	0.0039	0.010	

[a] 30 days after application of DTPA.

[b] ± values are 1 S.D.

superimposed on the other treatments indicates that the ^{241}Am was still available to the chelating agent after 1 year.

Schulz et al.,[17] studied the uptake of plutonium and americium in wheat grain from three soil types with isotope added as both the chloride and the nitrate. Soils used were Aiken, a high-yielding, acid, forest soil; Yolo, a neutral, agricultural soil; and Panoche, a calcareous, alkaline soil. The soils were blended with stock solutions of Pu(III) chloride, Pu(IV) nitrate, Am(III) chloride, and Am(III) nitrate, to give about 10^6 dis/min per gram of ^{239}Pu and 10^5 dis/min per gram of ^{241}Am. Six wheat seedlings were planted in 3-kg portions of the soil in

pots designed to control any leakage. The wheat was thinned to 4 plants per pot at one week and grown to harvest (120 days later) in a controlled environment chamber. Results are given in Table X.

The differences in uptake serve to illustrate the importance of soil type and form of administration. The authors note that they expected the Aiken (forest) soil to have the highest uptake. Instead it was the lowest for both nuclides. The importance of the chemical form in the calcareous Panoche soil is not explained since for both nuclides the highest uptake observed was in this soil with the nitrate and the lowest with the chloride. It is of interest that the use of the edible seeds in this experiment resulted in lower concentration ratios than were observed in the previously described experiment.

TOXICITY OF NEPTUNIUM AND AMERICIUM TO PLANTS

In his experiments to determine plant uptake of ^{241}Am, ^{244}Cm, ^{237}Np, and ^{239}Pu Price[2] used 50 µCi of Np or Pu and 25 µCi of Am or Cm per quart container. Greater uptake of ^{237}Np than the other isotopes and the low specific activity of ^{237}Np resulted in 183 µg and 21 µg of Np in tumbleweed and cheatgrass, respectively. Leaf tip burning was noted in Np-contaminated seedlings and was considered to be a possible toxicity symptom since burning was not observed in any other plants including controls.

Cline[11] specifically studied toxicity of ^{241}Am relative to that of ^{239}Pu in pea seedlings. Seedlings were grown in solutions containing 0.1 mCi ^{241}Am or ^{239}Pu per liter. Seedling radicles grew about 1 cm after ^{241}Am treatment, but there was no secondary root formation and no mitotic figures were observed in

TABLE X

CONCENTRATION FACTORS FOR WHEAT GRAINS

Soil Type	Chemical Form of Nuclide	Concentration Factors Pu	Concentration Factors Am	Ratio Am/Pu
Aiken	Nitrate	1.1×10^{-7}	6.1×10^{-6}	55
	Chloride	1.2×10^{-7}	5.3×10^{-6}	44
Yolo	Nitrate	5.5×10^{-7}	1.6×10^{-5}	29
	Chloride	7.8×10^{-7}	3.0×10^{-5}	38
Panoche	Nitrate	3.8×10^{-6}	6.3×10^{-6}	1.7
	Chloride	4.4×10^{-8}	2.6×10^{-7}	5.9

root tissue. Seedlings treated with ^{239}Pu showed diminished cell division for about a day, but nearly normal secondary root growth occurred. The author suggested solubility differences based on specific activity of the two isotopes as a possible cause; he also suggested that differences in chemical behavior which allowed greater uptake of ^{241}Am also allowed greater toxicity of Am to be exerted upon the plant.

DISCUSSION

The data from experiments using no treatment, such as chelating agents, are given in Table XI. The plutonium values are primarily those resulting from comparative experiments and are included to provide some relative comparison of uptakes. They, by no means, are intended to be an exhaustive survey of the data available. Percentage uptake values (i.e., percent of the nuclide in the total soil mass taken up by the plant or plant part) are included since concentration factors could not be obtained in some cases. Although percentage uptake is of little value for our purposes because it depends on mass of soil and mass of plant, the values can be used for comparison in those cases where concentration factors cannot be estimated.

Concentration factors seem to provide the best means of comparison between plants or soils or experiments; however, as we have pointed out with regard to studies of Price,[2-6] experimental methods and related assumptions may cause the calculation and interpretation of CF's to be less straightforward than one might presume. The method of layering used by Price[2] as a means of simulating a surface waste storage situation, as opposed to uniformly mixing a radionuclide with soil, may be the more realistic approach to some circumstances in which plants can become contaminated through their roots. The results of the experiments, which indicate that grass is less likely to accumulate these radionuclides than plants such as tumbleweed, may be influenced by the technique used and the characteristics of the nuclides relative to soil. In the case of experiments performed by Hale and Wallace[14] and by Wallace,[16] it is not known to what extent the availability of nuclides for plant uptake may have been altered by their having added vermiculite to the soil, thus increasing the volume of a given weight of soil.

In attempting to compare the available uptake data shown in Table XI and to assess their general applicability, one must recognize the possible influence of root structure and plant type. The difference between cheatgrass and tumbleweed roots was noted earlier. Although quantitative information has not been found relating tumbleweed age to root length, personal knowledge of the plant suggests

TABLE XI

SUMMARY OF AVAILABLE DATA ON UPTAKE

Nuclide	Plant	Soil	Part Analyzed	Growth Period	Uptake	Concentration Factor	Reference
^{237}Np	Tumbleweed	Burbank	[a]All	2 mo	2.1	0.11	2
	Cheatgrass	Burbank	All	2 mo	0.12	0.013	2
^{239}Pu	Tumbleweed	Burbank	All	2 mo	9.3×10^{-4}	4.6×10^{-5}	2
	Cheatgrass	Burbank	All	2 mo	1.9×10^{-4}	1.7×10^{-5}	2
	Barley	Cinebar	All	18 da	----	2.0×10^{-4}	11
	Barley	Ephrata	All	18 da	----	1.0×10^{-4}	11
	Bean	Solution	All	4 da	----	6.0×10^{-5}	11
	Wheat	Aiken	Grain	127 da	----	1.2×10^{-7}	18
		Yolo	Grain	127 da	----	6.6×10^{-7}	18
		Panoche	Grain	127 da	----	1.9×10^{-6}	18
^{241}Am	Tumbleweed	Burbank	All	2 mo	0.032	1.4×10^{-3}	2
	Cheatgrass	Burbank	All	2 mo	0.0072	6.0×10^{-4}	2
	Barley	Cinebar	All	18 da	----	3.0×10^{-3}	11
	Barley	Ephrata	All	18 da	----	2.0×10^{-3}	11
	Bean	Solution	All	4 da	----	3.0×10^{-3}	11
	Bush Bean	Hacienda	Leaves	14 da	3.0×10^{-3}	0.075	14
		Hacienda	Leaves	31 da	2.2×10^{-3}	0.054	14
		Hacienda	Leaves	45 da	8.7×10^{-3}	0.22	14
		Yolo	Leaves	14 da	7.8×10^{-3}	0.20	14
		Yolo	Leaves	31 da	3.3×10^{-3}	0.081	14
		Yolo	Leaves	45 da	1.1×10^{-3}	0.027	14
	Soybean	Hacienda	Stems	13 da	5.5×10^{-3}	0.22	16
		Hacienda	Leaves	13 da	6.0×10^{-3}	0.12	16
		Hacienda	All	13 da	1.2×10^{-2}	0.15	16
	[b]Orange	Hacienda	Leaves	85 da	----	0	16
		Hacienda	Stems	85 da	----	0	16
		Hacienda	Old Leaves	119 da	----	5.2×10^{-3}	16
		Hacienda	New Leaves	119 da	----	5.2×10^{-3}	16
		Hacienda	Stems	119 da	----	0.010	16
	[c]Orange	Hacienda	Leaves	85 da	----	3.9×10^{-3}	16
		Hacienda	Stems	85 da	----	0	16
		Hacienda	Old Leaves	119 da	----	5.2×10^{-3}	16
		Hacienda	New Leaves	119 da	----	2.6×10^{-3}	16
		Hacienda	Stems	119 da	----	2.6×10^{-3}	16
	Wheat	Aiken	Grain	127 da	----	5.7×10^{-6}	17
		Yolo	Grain	127 da	----	2.3×10^{-5}	17
		Panoche	Grain	127 da	----	3.3×10^{-6}	17
^{242}Cm	Fescue Grass	Fullerton	All	28 da	<0.003	----	8
	Bean	Solution	All	28 da	0.03	----	8
		Fullerton	All	28 da	0.068	----	8
	[d]Bean	Solution	All	28 da	0.086	----	8
	Tumbleweed	Burbank	All	2 mo	0.047	2.2×10^{-3}	2
	Cheatgrass	Burbank	All	2 mo	0.006	4.8×10^{-4}	2

[a]Designation "All" means all aboveground parts.

[b]Valencia orange grafted to rough lemon rootstock.

[c]Valencia orange grafted to trifoliate orange rootstock.

[d]Roots excised 1 cm below root collar before placing in nutrient solution.

that the root of a two-month-old plant could easily reach the bottom of a standard pint or quart container. Two literature sources were found which indicate that in mature plants of cheatgrass[18] or fescue,[18,19] root depth would not exceed 1.5 inches. A possible extreme in tap root growth may be found in alfalfa, an important forage crop for which we have no uptake data for transuranic nuclides. While alfalfa roots normally reach depths of 6-15 feet, a depth of 129 feet has been recorded.[20] For such a plant, the phenomenon of weathering could serve to maintain the availability of nuclides for uptake, although uptake might be relatively less at greater depths than near the surface. A comparison of plant types studied, with regard to their above ground parts, shows quite a variation, grasses having little stem, tumbleweed having little leaf, and bean and citrus having both stems and leaves. In accepting values for uptake derived from experiments with these plant types, one must recognize the lack of information on such plants as root crops and leafy vegetables, and the fruits of those plants that have been studied.

Recognizing the numerous factors that may have influenced experimental results shown in Table XI, some estimates can be made of probable plant uptake of the four transuranium elements studied. Simple averages (not weighted by number of samples comprising each value) and ranges of CF's, and the ratios of average Np, Am, and Cm CF's to ^{239}Pu CF are shown in Table XII. The wheat seed and orange plant data were not included and only the total plant value for soy beans was used. To place uptake ratios in perspective relative to the potential environmental importance of these nuclides, abundance of the nuclides relative to ^{239}Pu at 3, 100, and 1,000 years in the fuel have been used. Relative abundance values multiplied by the approximate relative uptake (CF) values from Table XII provide numbers relating to the potential importance of these isotopes with regard to their availability to the food chain in plants. Table XIII shows some of these relationships, giving some of those in which importance relative to ^{239}Pu is greater than one. It is clear that some isotopes other than ^{239}Pu are of importance in the environment for long times.

A summary (Table XIV) of the effect of DTPA on ^{241}Am uptake by leaves of bush bean, soybean, and Valencia orange, and by aboveground parts of tumbleweed and cheatgrass shows increases ranging from 22 to 885 times control values. Increase factors of less than 170 are associated with second and third croppings of bush bean leaves and in soybean leaves the control CF of which was comparatively

TABLE XII

UPTAKE OF PU, NP, AM, AND CM BY PLANTS

FROM CONTAMINATED GROWING MEDIA

Isotope	Number Values	Concentration Factors		Av CF
		Average	Range	Av CF of ^{239}Pu
^{237}Np	2	0.062	$1.3 \times 10^{-2} - 1.1 \times 10^{-1}$	730
^{239}Pu	5	8.5×10^{-5}	$1.7 \times 10^{-5} - 2.0 \times 10^{-4}$	1
^{241}Am	12	0.068	$6.0 \times 10^{-4} - 2.2 \times 10^{-1}$	800
^{244}Cm	2	1.3×10^{-3}	$4.8 \times 10^{-4} - 2.2 \times 10^{-3}$	15

TABLE XIII

RELATIVE IMPORTANCE OF SOME TRANSURANIC

NUCLIDES WITH REGARD TO UPTAKE BY PLANTS

Isotope	Time	Abundance Rel. to ^{239}Pu	Plant Uptake (CF) Rel. to ^{239}Pu	Importance Rel. to ^{239}Pu
^{237}Np	3 y	1.86×10^{-5}	730	0.014
	100 y	1.95×10^{-3}	730	1.4
	1,000 y	1.22×10^{-3}	730	0.89
^{241}Am	3 y	1.49×10^{0}	800	1,200
	100 y	6.22×10^{0}	800	5,000
	1,000 y	1.65×10^{0}	800	1,300
^{244}Cm	3 y	4.04×10^{-1}	15	6.1
	100 y	8.90×10^{-3}	15	0.13

high. Amount of DTPA used by Price[5,6] was not reported; that used by Hale and Wallace[14] was equivalent to 10 pounds Fe/acre; Wallace[16] used 13 ppm DTPA with 2 ppm Fe (~3.6 lb/acre) in his soybean experiment and 35 ppm DTPA in his 119-day citrus experiment. Agricultural levels of chelate were reported to be equivalent to 1-4 pounds Fe/acre.[15] Price[5] added chelate to the isotope solution and then to the soil surface; the other investigators mixed isotopes with soil and added chelate to the soil mixture. Increase factors due to DTPA treatments are quite similar, considering experimental differences, and may represent an upper boundary for the effect that might be anticipated for increase in ^{241}Am uptake.

One further point should be considered in use of these uptake data. All concentration factors given were based on the oven-dry weight of plant tissues analyzed. In young plants, such as were used in many of the experiments, the

TABLE XIV

INFLUENCE OF DTPA ON ^{241}AM UPTAKE BY PLANTS

Plant	Soil	Growth Period	Concentration Factors		Increase Factor	Reference
			Control	DTPA		
Tumbleweed	Burbank	2 months	1.4×10^{-3}	5.6×10^{-1}	4.0×10^{2}	6
Cheatgrass	Burbank	2 months	6.0×10^{-4}	1.0×10^{-1}	1.7×10^{2}	6
Bush Bean	Hacienda	14 days	7.5×10^{-2}	5.1×10^{1}	6.8×10^{2}	14
	Hacienda	31 days	5.4×10^{-2}	4.8	8.9×10^{1}	14
	Hacienda	45 days	2.2×10^{-1}	1.1×10^{1}	5.0×10^{1}	14
	Yolo	14 days	2.0×10^{-1}	1.2×10^{2}	6.0×10^{2}	14
	Yolo	31 days	8.1×10^{-2}	5.4	6.7×10^{1}	14
	Yolo	45 days	2.7×10^{-2}	1.1×10^{1}	4.0×10^{2}	14
Soybean[a]	Hacienda	13 days	1.2×10^{-1}	2.6	2.2×10^{1}	16
Orange-Lemon Rootstock	Hacienda	119 days (old)	5.2×10^{-3}	2.1	4.0×10^{2}	16
	Hacienda	119 days (new)	5.2×10^{-3}	4.6	8.8×10^{2}	16
Orange-Orange Rootstock	Hacienda	119 days (old)	5.2×10^{-3}	1.2	2.3×10^{2}	16
	Hacienda	119 days (new)	2.6×10^{-3}	1.0	3.8×10^{2}	16

[a] Soybean data from Fe+DTPA treatment.

water content is fairly high although variable among different plant types. Although this may be a useful method of reporting scientific data, the diets of individuals are generally based upon wet weight. Thus to use these concentration factors in estimating transfer through the food chain, it is necessary to reconvert them to the wet weight basis. Unfortunately, investigators seldom report the wet weight so that approximations are necessary. If we assume 80% as the water content, all concentration factors given should be reduced by a factor of 5. We would recommend that all experiments of this nature which are intended to be used in assessing health risks be based on wet weights.

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