Factors Affecting the Availability of Americium-241 to the Rice Plant

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Since there has been no published transuranic uptake data on the rice plant (*Oryza sativa* L.), greenhouse experiments were conducted to determine the effects of some factors on the uptake of ^{241}Am by this crop. Results indicate that chelated ^{241}Am (in the form of americium-241–diethylenetriaminepentaacetic acid) applied to the flood water was markedly taken up by the rice plant, compared to the nonchelated form. However, most of the accumulation of ^{241}Am occurred in the vegetative parts and only trace amounts, if any, were translocated to the grain. Soil application of ^{241}Am resulted in much lower uptake. Soil amendment with either diethylenetriaminepentaacetic acid (DTPA) or organic matter did not produce a discernible uptake pattern. A synthesis of published data on plant uptake of ^{241}Am indicates that the concentration ratio (CR, a measure of availability of ^{241}Am to the plants) values for ^{241}Am for agricultural crops ranged from 10^{-6} to 10^{1} (from lowest to highest availability). Some factors that appear to influence ^{241}Am uptake are as follows: plant parts (grain usually having lower CR), chelating agents (DTPA usually increasing the CR), organic matter (inconsistent effects although generally decreasing the CR), and lime (usually decreasing the CR).

Proposals in recent years to increase the use and production of plutonium in the nuclear fuel cycle have catalyzed research enthusiasm on the environmental chemistry of the transuranic nuclides. These radionuclides, if released to the environment, may pose a threat to human health either by direct inhalation or ingestion. Airborne radionuclides can reach humans by the inhalation pathway, whereas those deposited in the ground can reach humans through the food-chain pathway. Humans may consume the food crops directly, or indirectly by consumption of meat of animals (wildlife and livestock animals), which feed on these contaminated plants.

Although the inhalation pathway has been considered as the major route in which these contaminants may reach humans (Bennett, 1976), the food-chain pathway may be more important based on a long-term assessment, because some of these radionuclides have extremely long physical half-lives. For example, plutonium is carcinogenic once it gains entry into the body and has a half-life of approximately 24 000 years for ^{239}Pu and about 90 years for ^{238}Pu. On the other hand, ^{241}Am, a transplutonic actinide and daughter product of ^{241}Pu, has a half-life of about 460 years. These elements are alpha-emitters and have been shown to be generally persistent and immobile in the soil–plant system (Price, 1973).

Plant uptake studies can be classified into four general categories: (1) studies using soils contaminated by global (nuclear) fallout (Hardy et al., 1977); (2) studies using soils contaminated by plutonium associated with high explosive (nonnuclear) detonations (Schulz et al., 1976b); (3) studies using soils contaminated by waste disposal as in the White Oak Creek Floodplain at Oak Ridge (Dahlman, 1978) and Los Alamos (Hakonson, 1978); and (4) studies using artificially spiked soils (Adriano et al., 1979; Thomas and Healy, 1976). More research on naturally contaminated areas probably would ensue in view of the most recent findings that ^{60}Co migrated from nuclear waste burial grounds at the Oak Ridge National Laboratory (Means et al., 1978) and some transuranic elements at the Idaho National Engineering Laboratory (Markham et al., 1978). These findings imply that potential contamination of vegetation adjacent to nuclear waste burial sites around the country exists.

In the 1950's and 1960's, some investigators have demonstrated that transuranic elements entered the plant roots in trace quantities and were translocated to aerial parts of plants. In general, plant species differed in their uptake of these elements with ^{241}Am generally found more available than ^{239}Pu or ^{238}Pu. Values for concentration ratios (CR: calculated as the ratio of ^{241}Am radioactivity per unit mass of plant tissue and radioactivity per unit mass of soil, both dry weight), an empirical index of availability, generally ranged from 10^{-7} to 10^{-3} for ^{239}Pu, compared to 10^{-6} to 10^{1} for ^{241}Am (Adriano et al., 1979). These wide variations in availability were caused by numerous factors, which will be discussed later.

Although rice is a major food crop in the world, the literature search revealed that there are no transuranic data on this important crop. The apparent reason is that even though nuclear fuel technology is advanced in some countries, including the United States, rice is not an important food crop in these countries. In the United States, rice is being cultured rather extensively only in the states of Arkansas, California, Louisiana, and Texas (Adair et al., 1962), where in 1975 production approached 6×10^{6} tons, which was equivalent to about 1.5% of the world total. Because of the global importance of rice, studies were conducted to (1) determine the effects of the method of application (water applied vs. soil applied) of ^{241}Am on its uptake by the rice plant, (2) determine the effects of some soil amendments on the uptake of ^{241}Am by the rice plant, and (3) compare the uptake data for rice with the uptake data for other plants published in the open literature.

EXPERIMENTAL METHODS

An uncontaminated Dothan sandy clay loam soil, an ultisol, was collected from the top layer in a field located at the U.S. Department of Energy Savannah River Plant. This soil type is commonly found in the Savannah River Area, site of the Savannah River Plant, near Aiken, South Carolina (where three operational nuclear reactors, two reprocessing facilities, and a nuclear waste burial ground are located) and nuclear waste burial ground in Barnwell, South Carolina. The soil had a pH of about 4.5, total carbon content of 1%, 12% clay, and cation-exchange

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Table I. Americium-241 Concentrations in Various Rice Plant Parts (Experiment 1) [Values (Means of Five Replicates) in the Same Vertical Column (Separately for + DTPA and − DTPA) Sharing the Same Alphabet Are Statistically the Same ($p \leq 0.05$)]

method	concentration, pCi/g dry wt: unshelled grain	grain collar[a]	stem	green blade	dead blade	leaf sheath
			+ DTPA			
water applied						
period 1[b]	BG[ca]	0.88[a]	4.52[a]	1.53[a]	109[ab]	443[a]
period 2	0.46[b]	2.86[a]	10.3[a]	5.37[b]	260[b]	1581[b]
period 3	0.42[b]	4.16[a]	36.5[b]	6.75[b]	631[c]	3715[c]
soil applied[d]	BG[a]	1.20[a]	2.02[a]	0.10[a]	11.2[a]	9.65[a]
			− DTPA			
water applied						
period 1	BG[a]	0.84[a]	3.26[a]	2.13[a]	18.6[a]	42.0[a]
period 2	0.27[b]	2.13[a]	6.45[b]	2.30[a]	19.3[a]	89.0[a]
period 3	BG[a]	0.41[a]	7.15[b]	3.81[a]	169[b]	406[b]
soil applied[d]	BG[a]	1.35[a]	2.74[a]	1.05[a]	9.81[a]	4.27[a]

[a] Mean values of three replications. [b] Periods 1, 2, and 3 correspond to booting stage, flowering stage, and dough (ripening) stage, respectively. [c] Radioactivity was below the detection level. [d] Data taken from experiment 2.

Table II. Americium-241 Concentrations in Various Rice Plant Parts (Experiment 2) [Values in the Same Vertical Column Sharing the Same Alphabet Are Statistically the Same ($p \leq 0.05$)][a]

treatment	concentration, pCi/g dry wt: unshelled grain	grain collar	stem	green blade	dead blade	leaf sheath
			Flooded			
control	BG[ba]	1.35[a]	2.74[ab]	1.05[a]	9.81[a]	4.27[ab]
+ DTPA	BG[a]	1.20[a]	2.02[ab]	0.10[a]	11.23[a]	9.65[c]
+ 5% O.M.	BG[a]	1.18[a]	2.66[b]	3.32[bc]	5.97[a]	3.66[ab]
			Nonflooded			
control	0.28[a]	3.40[a]	5.58[c]	10.66[d]	23.79[b]	7.72[bc]
+ DTPA	0.10[a]	0.25[a]	BG[a]	1.55[ab]	8.73[a]	2.93[a]
+ 5% O.M.	0.11[a]	1.21[a]	4.07[bc]	5.89[c]	6.58[a]	2.56[a]

[a] Values are means of five replicates except for the grain collar where sample $n = 3$. [b] Radioactivity was below the detection level.

capacity of 10 mequiv/100 g of soil.

In the first experiment, a total of 5 kg of soil was placed in each black plastic pot (top diameter = 22 cm; bottom diameter = 18; height = 20 cm; soil column = 13 cm) and 2 μCi of ^{241}Am was added by water application. Chelated or nonchelated ^{241}Am was added to the standing water (flooded condition) at three stages of growth: booting stage, flowering stage, and dough (ripening) stage. For chelated treatments americium was chelated before the application by adding $^{241}Am(NO_3)_3$ solution to 50 mL of 100 ppm diethylenetriaminepentaacetic acid (DTPA) solution, giving 0.001 N HNO_3.

In the second experiment, 5 kg of soil per pot was also used, amended either with DTPA or organic matter (bermuda grass hay). For chelation, the DTPA salt was premixed (before the americium application) with the whole soil to give 40 ppm DTPA and, likewise, the organic matter was premixed at the 5% rate, on a dry weight basis. The "layering" technique (Adriano et al., 1977) was used to spike the top 1 kg of soil with 2 μCi of nonchelated ^{241}Am in both the DTPA and 5% O.M. treatments. The control treatment was spiked similarly.

In the first experiment, a lowland (flood) rice variety from Asia was used. In the second experiment, two varieties [same lowland variety and an upland (nonflood) variety from Asia] were used. Ten rice seeds were germinated directly on the pot and eventually thinned out to only three seedlings per pot. All treatments were replicated five times (number of samples per treatment = 5). The pots were randomly distributed in a metal water bath located in a greenhouse. The water in the bath was circulated using an immersed pump to produce a uniform temperature (27 ± 1 °C) throughout the tank. In flooded treatments, the pots always had about 3 cm of standing water above the soil surface at all times, whereas in nonflooded treatments, the pots were irrigated close to saturation when needed, to produce a partially aerobic condition. During the early stages of growth, all pots were supplemented once with solutions of reagent-grade NH_4NO_3, KH_2PO_4, and KNO_3 to provide 100 ppm N, 50 ppm P, and 100 ppm K (dry soil weight basis), respectively.

At maturity, the water in the pots was allowed to recede, plants were clipped about 5 cm from the soil surface, and the lower 10-cm portions were rinsed in deionized water, then separated into various parts. The vegetative parts were clipped into shorter pieces, oven-dried to constant weight, and placed into counting tubes. The samples were counted in a NaI well crystal interphased with a multichannel analyzer for 140 min. The whole grain was analyzed unshelled because of limited sample size.

Analysis of variance were conducted on the data and sample means compared using Tukey's HSD multiple range test (Steel and Torrie, 1960).

RESULTS AND DISCUSSION

Results presented in Tables I and II indicate that only trace quantities, if any, of ^{241}Am were translocated to the whole (unshelled) grain. The vegetative data also indicate that ^{241}Am-DTPA could be translocated to the grain with water application (Table I). However, the radioactivities in some grain samples were insignificant compared with the radioactivities of the other plant parts. No radioactivity in the grain was detected with soil application, however, except with the upland (nonflood) variety (Table II).

Table III. Percent Distribution of ^{241}Am in the Rice Plant (Experiment 1)[a]

method	distribution as percent of total removed					
	unshelled grain	grain collar	stem	green blade	dead blade	leaf sheath
			+ DTPA			
water applied						
period 1	0 (17.6)[b]	<0.1 (5.5)	0.6 (5.8)	0.2 (7.9)	17.6 (7.1)	81.6 (9.1)
period 2	<0.1 (18.8)	<0.1 (5.8)	0.4 (6.7)	0.2 (7.8)	9.7 (6.7)	89.6 (10.1)
period 3	<0.1 (19.4)	<0.1 (5.9)	0.6 (7.5)	0.1 (8.1)	8.9 (6.4)	90.3 (10.6)
soil applied[c]	0 (24.1)	1.5 (5.9)	5.3 (8.1)	<0.1 (6.7)	44.7 (12.2)	48.5 (15.4)
			− DTPA			
water applied						
period 1	0 (16.9)	0.2 (4.8)	3.6 (6.6)	2.9 (7.9)	19.6 (6.1)	73.7 (9.7)
period 2	0.5 (18.8)	0.6 (5.6)	4.0 (7.5)	1.6 (8.6)	12.1 (6.7)	81.3 (10.7)
period 3	0 (18.1)	<0.1 (4.9)	1.0 (6.8)	0.6 (7.9)	17.9 (6.2)	80.5 (10.1)
soil applied[c]	0 (19.8)	2.6 (5.3)	8.9 (6.6)	3.5 (7.1)	56.6 (11.6)	28.4 (13.6)

[a] Calculated from the concentrations (Table I) and total dry matter of each part. [b] Values in parentheses are for dry matter (grams per pot). [c] Data taken from experiment 2.

Table IV. Percent Distribution of ^{241}Am in the Rice Plant (Experiment 2)[a]

treatment	distribution as percent of total removed					
	unshelled grain	grain collar	stem	green blade	dead blade	leaf sheath
			Flooded			
control	0 (19.8)[a]	2.6 (5.3)	8.9 (6.6)	3.5 (7.1)	56.7 (11.6)	28.3 (13.6)
+ DTPA	0 (24.1)	1.5 (5.9)	5.3 (8.1)	<0.1 (6.7)	44.7 (12.2)	48.5 (15.4)
+ 5% O.M.	0 (13.7)	2.9 (5.1)	17.0 (7.0)	19.4 (6.7)	34.1 (6.6)	26.6 (8.7)
			Nonflooded			
control	~1.0 (21.2)	1.2 (5.3)	10.7 (15.2)	11.8 (8.6)	55.2 (18.0)	20.3 (20.6)
+ DTPA	~1.0 (20.8)	0.5 (5.6)	0 (14.0)	4.2 (7.8)	68.4 (16.5)	25.9 (19.5)
+ 5% O.M.	1.0 (23.1)	1.3 (5.9)	22.2 (13.1)	17.3 (7.9)	39.1 (14.3)	18.6 (17.6)

[a] Calculated from the concentrations (Table II) and total dry matter of each part. [b] Values in parentheses are for dry matter (grams per pot).

When water-applied, Am in various parts of the rice plant decreased in the order: leaf sheath ≫ dead blade > stem > green blade ≥ grain collar > unshelled grain (Table I). However, when soil applied Am decreased in the order: dead blade > leaf sheath ≥ stem ≥ green blade ≥ grain collar > unshelled grain (Table II). With water application, the stems had considerably higher Am contents than the green blades while dead blades had much higher Am than the green blades. With soil application, however, the stem had equal or greater Am contents than the green blade. Thus, based from both experiments (Tables I and II), it appeared that Am accumulation in the older leaves was larger than in the newer leaves. Most of the accumulation occurred in the sheath with water application, especially with the ^{241}Am-DTPA form (Table I). This did not happen with soil application, however (Table II). Thus, it appears that the accumulation in the sheath can be attributed primarily to physical absorption of the radioisotope from the standing water, rather than physiological assimilation, as might be the case for stems and leaves.

Some important trends in Tables I and II need further elaboration. Firstly, the method of placement affected the uptake of ^{241}Am (Table I). With DTPA, the ratios of plant Am from water application to plant Am from soil application ranged from 46 to 385 for the leaf sheath; 10 to 56 for the dead blade; and 15 to 68 for the green blade. Without DTPA, the ratios ranged from 10 to 95 for the leaf sheath; 2 to 17 for the dead blade, and 2 to 3 for the green blade. Similar trends in rice were observed by Myttenaere et al. (1971) elsewhere where they found that flooded rice plants took up 37% of the ^{54}Mn applied to the soil surface and only 14% of that mixed with the soil. They attributed this trend to the numerous adventitious and surface roots of the rice plants above the ground surface. In this study, the Am ions in the water (present most probably in the 3+ valence) were expected to be intercepted directly by the roots. On the other hand, when mixed with the soil, the Am ions were held in the "exchange" complex of the soil particles and were needed to be released first through the "exchange" process prior to interception by the roots through mass diffusion. Thus, it appears that Am ions applied to the standing water of flooded rice would be more

Table V. Total ^{241}Am Accumulated by the Rice Plant (Experiments 1 and 2)

	total dry plant tissues per pot, g[a]	total pCi per pot	as percent of total applied
exp. 1		+ DTPA	
water applied			
period 1	53.0	4634	0.23
period 2	55.9	18389	0.92
period 3	57.9	45077	2.25
soil applied[c]	72.4	311	0.02
		− DTPA	
water applied			
period 1	52.0	608	0.03
period 2	57.9	1221	0.06
period 3	54.0	5085	0.25
soil applied[c]	64.0	206	0.01
exp. 2[b]		Flooded	
control	64.0	206	0.01
+ DTPA	72.4	311	0.02
+ 5% O.M.	47.8	117	0.006
		Nonflooded	
control	88.9	783	0.04
+ DTPA	84.2	217	0.01
+ 5% O.M.	81.9	240	0.01

[a] Sum of dry matter of all parts. [b] For experiment 2, the radioisotope was all soil-applied. [c] Data taken from experiment 2.

Table VI. Concentration Ratio (CR)[a] of Americium-241 for Various Agricultural Crops

plant	part	growth stage	soil and location	comments	CR	reference
alfalfa (*Medicago sativa*)	whole, several cuttings	bloom stage	surface soil from the Nevada Test Site, NV, pH 8.0	grown in potted (3 kg) soil having 1 nCi/g of ^{241}Am from high explosive (nonnuclear) detonations > 20 years ago; soil amended with chelate DTPA, organic matter, sulfur, and nitrogen	10^{-4}–10^{-3}	Adriano et al. (1979)
alfalfa	whole, several cuttings	maturity	mountain meadow soil, Los Alamos, NM	grown in potted (1.5 kg) soil spiked with 18 nCi to 0.5 μCi/g of ^{241}Am; soil had 5% indigenous organic matter	10^{-2}	Adams et al. (1975)
bahiagrass (*Paspalum notatum*)	whole, several cuttings	maturity	troup sandy loam (pH 5.5) and Dothan sandy clay loam (pH 4.5), Savannah River Plant	grown in potted (2 kg) soil spiked with 500 pCi/g of ^{241}Am; soil amended with lime and organic matter	10^{-4}–10^{-1}	Hoyt and Adriano (1979)
barley (*Hordeum vulgare*)	leaves or upper shoots	3 weeks	Yolo loam (pH 6) and Hacienda loam (pH 7.5), CA	grown in potted (500 g) soil spiked with 1 nCi/g of ^{241}Am; soil amended with DTPA	10^{-1}–10^{0}	Wallace et al. (1976)
barley	grain	maturity	surface soil from the Nevada Test Site; pH 8	grown in potted (3 kg) soil containing 1–11 nCi/g of ^{241}Am from high explosive (nonnuclear) detonations > 20 years ago	10^{-6}	Schulz et al. (1976)
	foliage	maturity	same soil as above	same as above	10^{-4}	Schulz et al. (1976)
barley	grain	dough stage	surface soil from the Nevada Test Site; pH 8	same comments as for the alfalfa (Nevada Test Site) above	10^{-5}–10^{-3}	Adriano et al. (1979)
	foliage	maturity	same soil as above	same as above	10^{-5}–10^{-3}	Adriano et al. (1979)
barley	whole, several cuttings	maturity	mountain meadow soil; Los Alamos, NM	same comments as for the alfalfa (Los Alamos) above	10^{-4}	Adams et al. (1975)
barley	whole	18 days	Cinebar (pH 4.5) and Ephrata (pH 7.5), Richland, WA	used Neubauer technique with soils spiked with 1.8 μCi/g of ^{241}Am	10^{-3}	Cline (1968)
beans, bush (*Phaseolus vulgaris*)	leaves or upper shoots	3 weeks	Yolo loam (pH 6) and Hacienda loam (pH 7.5), CA	grown in potted (500 g) soil spiked with 1 nCi/g of ^{241}Am; soils amended with DTPA	10^{-1}–10^{1}	Wallace et al. (1976)
beans, bush	whole shoots	2 weeks	Troup sandy loam (pH 5.5) and Dothan sandy clay loam (pH 4.5); Savannah River Plant	grown in potted (500 g) soil spiked with 2 nCi/g of ^{241}Am; soils amended with lime and DTPA	10^{-1}–10^{1}	Adriano et al. (1977)
beans	whole	4 days	Hoagland nutrient solution	nutrient solution spiked with 0.9 μCi/mL of ^{241}Am	10^{-3}	Cline (1968)
corn (*Zea mays*)	leaves or upper shoots	3 weeks	Yolo loam (pH 6) and Hacienda loam (pH 7.5), CA	same comments as for bush beans (California) above	10^{-2}–10^{0}	Wallace et al. (1976)
corn	whole shoots	2 weeks	Troup sandy loam (pH 5.5) and Dothan sandy clay loam (pH 4.5); Savannah River Plant	same comments as for bush beans (South Carolina)	10^{-2}–10^{0}	Adriano et al. (1977)
(field) corn	ears (+ cobs)	maturity	vegetable garden sandy loam soil, MA	grown in garden plots in Cape Cod; soil had 1 fCi/g from global fallout; plants shielded from direct deposition or resuspension contamination	10^{-3}	Hardy et al. (1977)
orange, Valencia	leaves	4 months	Hacienda loam (pH 7.5) CA	grown in potted (500 g) soil spiked with 20 nCi/g of ^{241}Am; soils amended with chelating agents	10^{-3}	Wallace (1972)
(field) peas (*Pissum* spp.)	shelled grain	maturity	Vegetable garden sandy loam soil, MA	same comments as for field corn (MA)	10^{-3}	Hardy et al. (1977)
(field) potatoes (*Solanum tuberosum*)	skins removed	maturity	same soil as above	same as above	10^{-3}	Hardy et al. (1977)
rice[c] (*Oryza sativa*)	unshelled grain	maturity	Dothan sandy clay loam (pH 4.5), Savannah River Plant, SC	grown in potted (5 kg) soil under flooded condition; standing water	BG[b]–10^{-3}	this study

rice[c]	stem	maturity	same soil as above	spiked with 2 μCi of ^{241}Am at 3 stages of growth, with or without DTPA same as above	10^{-2}–10^{-1}	this study
	green blade	maturity	same soil as above	same as above	10^{-3}–10^{-2}	this study
	dead blade	maturity	same soil as above	same as above	10^{-2}–10^{0}	this study
	leaf sheath	maturity	same soil as above	same as above	10^{-1}–10^{1}	this study
	unshelled grain	maturity	same soil as above	grown in potted (5 kg) soil under flooded or nonflooded condition; spiked top 1 kg soil with 2 μCi of ^{241}Am; soils amended with DTPA or organic matter	BG–10^{-4}	this study
soybean (*Glycine max*)	stem	maturity	same soil as above	same as above	10^{-3}–10^{-2}	this study
	green blade	maturity	same soil as above	same as above	10^{-3}–10^{-2}	this study
	dead blade	maturity	same soil as above	same as above	10^{-2}	this study
	leaf sheath	maturity	same soil as above	same as above	10^{-2}	this study
	fruit	maturity	surface soils from the Nevada Test Site and Tonopah Test Range, NV; pH 8	grown in potted (3 kg) soil having 0.2–2 nCi/g of ^{241}Am from high explosvie (nonnuclear) detonations > 20 years ago	10^{-5}–10^{-2}	Adriano et al. (1979)
wheat (*Triticum aestivum*)	foliage	maturity	same soil as above	same as above	10^{-3}–10^{-1}	Adriano et al. (1979)
	grain	maturity	Aiken clay loam (pH 6.3), Panoche fine sandy loam (pH 7.9) and Yolo loam (pH 7), CA	grown in potted (3 kg) soil spiked with 45 nCi/g of ^{241}Am, either in chloride or nitrate form	10^{-6}–10^{-5}	Schulz et al. (1976a)

[a] Calculated as the ratio of radioactivity in plants to that in soil (or substrate), with equal units of mass. [b] Radioactivity was below the detection level. [c] In calculating CR values for rice, 400 pCi/g dry soil was used, based on the total soil mass in each pot. If based on only the top 1 kg soil, 2000 pCi/g should be used, and a conversion factor of 0.20 should be used.

readily available to the rice plant through direct interception of the adventitious roots or immersed portions of the plant.

Secondly, the chelating agent DTPA markedly enhanced the uptake of Am ions when Am was water applied (Table I). This is in accordance with significant increases in the uptake of ^{241}Am, as well as ^{239}Pu, by plants caused by chelating agents which were demonstrated recently (Adriano et al., 1979; Lipton and Goldin, 1976; Ballou et al., 1978). Increases in uptake by plants due to chelation were in the order of 10^3 for ^{239}Pu (Lipton and Goldin, 1976) and also 10^3 for ^{241}Am (Adriano et al., 1977). With water application (Table I), the plant Am ratios of +DTPA/−DTPA treatments ranged from 9 to 18 for the leaf sheath; 4 to 14 for the dead blade; 1 to 2 for the green blade; and 1 to 5 for the stem. No consistent pattern, however, was obtained with soil application (Table II). Chelating agents are important in studying the radionuclide uptake by agricultural crops for two reasons: (1) they are used in agriculture, at the rates of several kilograms per hectare, to supply micronutrients, such as Fe, Zn, Mn, and Cu, and (2) they are used in decontamination operations in nuclear facilities (Means et al., 1978). Therefore, they are expected to be present in complexed forms with metal radionuclides in nuclear waste burial areas. The basic roles of chelating agents on the availability of metals to plants have been reviewed by Wallace (1963). These compounds complex with metals and can keep the metals in the soluble form in the aqueous phase or soil solution in the soil–plant environment. Wallace (1963) contends that the metal–chelate complex may separate in the roots into metal ions and chelate ions or both may be transported to the leaves in a complexed form. Chelating agents alone, i.e., without the metal, often compete with the roots and soil particles for metals. This might partially explain the inconsistent effect of soil-mixed DTPA on the rice plant uptake (Table II). Recent significant findings indicate that rats, fed plants contaminated with DTPA-chelated ^{239}Pu, ^{241}Am, and ^{244}Cm, did not accumulate these elements in their tissues (Ballou et al., 1978).

Thirdly, the uptake of water-applied ^{241}Am appeared to have increased in later stages of growth of the rice plant (Table I). It was found in Japan (Kasai and Asada, 1964) that some elements, like Ca and Sr, when detected in the grain were absorbed mainly after the flowering stage. In Italy (Bourdeau et al., 1965), uptake by rice for ^{144}Ce, ^{90}Sr, ^{137}Cs, ^{106}Ru, and ^{54}Mn continued until harvest time but only trace quantities were translocated to the grain. Again in Japan (Kasai and Asada, 1964) trace amounts of ^{90}Sr and ^{45}Ca were also detected in the grain, with most of the accumulation, however, occurring in the leaf blades. Some of the minor elements (Fe, Mn, and Al) accumulated only in the old leaves and stems of the rice plant (Ishizuka, 1971). Thus, it appears that some elements could be more available to the rice plant at certain stages of growth. Of particular concern with regard to human health is when harmful elements are taken up during the fruiting stage when there is greater possibility of them being incorporated in the edible portions.

Fourthly, some soil amendments may affect Am uptake by the rice plant (Table II). When soil mixed (Table II), DTPA did not have consistent effects on uptake. Similarly, organic matter mixed with the soil did not have consistent effects although in some instances it decreased the uptake (Table II). In an earlier study, Hoyt and Adriano (1979) found that organic matter reduced ^{241}Am uptake by bahiagrass (*Paspalum notatum*) especially when applied at high rates (5% by dry weight). This was at-

tributed to increases in fixing capacity or complexation for metals by organic matter in soils. Kirkham (1977) recently reviewed the roles of soil organic matter on the availability of metals to plants and pointed out that, in general, organic matter decreased metal availability to plants. It should be pointed out that addition of high rates of fresh organic matter to anaerobic flooded soils can result in the production of organic acids (Rao and Mikkelsen, 1977) which, consequently, can cause toxic effects to the rice plant. The flooded rice plants in experiment 2 were apparently affected by these organic acids, as indicated by the lower dry matter production than in experiment 1 (Tables III and IV).

Water submergence (flooding) of soil induces reducing conditions and can affect nutrient availability to the rice plant. Some trace elements were taken up more by the rice plant under these conditions (Jugsujinda and Patrick, 1977; Cherian et al., 1968). Beneficial effects of flooding on rice growth frequently have been attributed to increased availability of some mineral nutrients. However, no consistent trend in uptake of ^{241}Am can be discerned between the flooded and nonflooded rice although this might have been caused by the inherent genetic and physiological characteristics of the two rice varieties.

The relative distribution of ^{241}Am in various rice plant parts is presented in Tables III and IV. These values were calculated from the concentration data and dry matter mass of each part. The results indicate that most of the Am tend to accumulate in the sheath with water application, which was probably caused by physical absorption primarily, rather than physiological translocation. The quantities of Am in the dead blades and sheaths differed according to the method of placement and whether Am was chelated or not. When water applied as ^{241}Am-DTPA, approximately 87% (average of three treatments) of the total Am in the plant occurred in the sheaths as compared to only 78% when not chelated (Table III). When soil applied, however, an average 45% accumulated in the dead blades vs. 34% in the sheaths of the flood variety; and correspondingly, 54 and 22% for the nonflood variety (Table IV). Thus, with water application, Am tended to accumulate in the sheath and, conversely, in the older blades with soil application.

The total accumulation of ^{241}Am by the rice plant from the substrate is summarized in Table V. Greater accumulation occurred when the radionuclide was applied in the water, ranging from 0.03 to 2.25% of the total applied compared with only 0.006 to 0.04% when applied to the soil. The highest total accumulation occurred with water application of ^{241}Am-DTPA and was generally higher than previously reported values for agricultural crops (Thomas and Healy, 1976).

The various treatments did not produce significant changes in the soil pH which could have caused the differences in uptake. Soil cores collected after harvest from pots which received water application of ^{241}Am had pH of 5.5 on the average, which was higher than the original soil pH of 4.5 (experiment 1). In experiment 2, the flooded soils had an average pH of 5.9; the nonflooded soils had an average pH of 5.2, with soils amended with the 5% organic matter, showing larger increases over the original pH. The organic matter treatment caused a twofold increase in the total carbon contents of the soils, from 1% C to 2.1% C for nonflooded soils and to 1.7% C for flooded soils. The rest of the soils had an average carbon contents of approximately 0.75% after the harvest.

The ^{241}Am uptake data, expressed as CR values, for various agricultural crops reported in the literature are summarized in Table VI. These crops were grown either in laboratories, greenhouses, or in the field. The CR ranged from 10^{-6} to 10^{1}, where availability to the plant increases as CR increases. Within a particular plant, the grain usually had lower CR, as was the case with rice (this study), barley (Schulz et al., 1976b), and soybeans (Adriano et al., 1979). On the average, the grain had 100 times lower concentration of ^{241}Am than the foliage. The use of synthetic chelating agent DTPA usually caused the highest CR, of up to 10^{1}. This was demonstrated in alfalfa (Adriano et al., 1979), barley (Adriano et al., 1979; Wallace et al., 1976), bush beans (Adriano et al., 1977; Wallace et al., 1976), corn (Adriano et al., 1977; Wallace et al., 1976), rice (this study), and soybeans (Adriano et al., 1979). The increase in CR caused by DTPA usually was about ten to one-hundred times over those treatments that did not receive this chelating agent. Other soil amendments, like organic matter, nitrogen, and sulfur, did not have marked effects and, in some cases, even decreased the CR (Adriano et al., 1979; Hoyt and Adriano, 1979). Of the various soil amendments tested by various investigators, lime appeared to be the most effective in lowering the CR, as demonstrated with bush beans and corn (Adriano et al., 1977). However, application of DTPA in limed soils potentially can result in increasing the CR, as shown by Adriano et al. (1977) and Wallace et al. (1976). Thus, while lime alone can immobilize ^{241}Am in the soil, application of DTPA in limed soils can cause the opposite effect. Undoubtedly, other factors affect the CR: plant species, soil type, soil pH, growth period, form of ^{241}Am applied, etc., which are quite complicated to assess. Therefore, CR values can be used to assess the effects of certain treatments on the availability of a given radionuclide and its translocation to various plant parts. As would be expected there is some question concerning the use of soil concentration for determining the CR for water application, but this CR could demonstrate the relative translocation or redistribution of ^{241}Am in the various rice parts.

It can be noted from Table VI that the amount of data from field experiments is limited. This is probably caused by the difficulties in conducting this type of experiment without contamination from aerosol and resuspension particulates. Thus, most of the studies have been conducted indoor, where radionuclide concentration in the soil can be elevated above that caused by global fallout. Nevertheless, whatever field data are available, they indicate that field-grown crops had CR values identical with the average CR (10^{-3}) of indoor-grown plants.

This paper demonstrates one possible entry of a transuranic element, ^{241}Am, to the food-chain pathway leading to man, i.e., via the root uptake. Marked uptake of ^{241}Am by important agricultural crops has been demonstrated depending on plant species and soil amendment. Therefore, lateral migration of this radionuclide from nuclear waste burial sites into vegetated areas can potentially cause the entry of ^{241}Am into the human food-chain pathway.

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Bacterial Conversion of Alkylphosphonates to Natural Products via Carbon–Phosphorus Bond Cleavage

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The phosphorus-containing breakdown products of *O*-alkyl alkylphosphonate toxicants, which are particularly resistant to cleavage at the C–P bond, were fully degraded to natural products by *Pseudomonas testosteroni*. When present as a sole and limiting phosphorus source, an *O*-alkyl alkylphosphonate was attacked aerobically via release of the alkoxy group as the alcohol, followed by cleavage of the alkyl–phosphorus bond (methyl, ethyl, or propyl) to produce the respective alkane and an inorganic phosphorus compound that was detected as inorganic orthophosphate. The bacterium could not cleave the bonds of other carbon–heteroatoms (e.g., arsonates, sulfonates, and mercurials). This is the first report of the metabolism of simple, aliphatic alkylphosphonates and the first pathway described for an organophosphorus toxicant to yield exclusively natural products (i.e., alcohols, alkanes, and phosphate).

The carbon–phosphorus bond is an exceedingly nonreactive constituent of unsubstituted alkyl- and arylphosphonate insecticides, herbicides, fungicides, nerve gases, flame retardants, and several other economically important categories of chemicals. Reviews of the metabolism of phosphonates have considered the terminal phosphorus-containing residues in animals and plants and emphasize that the existing evidence indicates that the C–P bond of methyl-, ethyl-, and phenylphosphonates resists cleavage by higher organisms (Menn, 1971; Menn and McBain, 1974). To our knowledge, the only reported exception to the inability of higher organisms to cleave this bond is the finding that rice plants cleave ionic *O*-ethyl phenylphosphonothioate, a hydrolytic product of Inezin, yielding ionic *O*-ethyl phosphorothioate (Endo et al., 1970); however, the authors did not eliminate the possibility of microbial attack.

Cook et al. (1978a) reported and reviewed the microbial utilization of representative classes of phosphorus-containing breakdown products of organophosphorus pesticides. They concluded that only one previous report gave definitive evidence of extensive utilization of a chemically stable organophosphorus compound (i.e., dimethyl hydrogen phosphate). The first direct evidence for the biological cleavage of the C–P bond of alkylphosphonates has recently been obtained (Daughton et al., 1979b), and the present study characterizes the metabolic pathway and its control. This is the first account of the complete metabolism to natural products of an organophosphorus xenobi-

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