THE ASSOCIATION OF TRANSURANIC ELEMENTS WITH LACUSTRINE SEDIMENTS

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Approximately 97% of the fallout ^{239,240}Pu and an even greater fraction of the ^{241}Am in Lake Michigan is in the sediments.[1] In addition, studies of gross sediment samples taken with a grab sampler have shown that the ^{239,240}Pu in the sediments is almost entirely associated with the hydrous oxides of iron and manganese.[2] Since these observations indicate that the vast majority of plutonium and americium introduced into the lake is associated with sedimentary phases, studies were continued to determine (1) if the americium is also associated with the hydrous oxides; (2) if the association of plutonium with the oxides remained unchanged with depth in the sediment; and (3) if the particle size of the sediment is important in the distribution of the plutonium.

Sediment samples were taken using a 3" "Benthos" gravity corer at a station (approximately 7 miles WSW of Grand Haven, Michigan). In addition, material was collected at the same location from a sediment trap placed approximately 5 m off the bottom at a water depth of 60 m. The core was sectioned into one centimeter subsections which were freeze-dried as were the sediment trap samples. The dried sediment trap material and material from the top four subsections of the core (depth to which significant ^{239,240}Pu concentrations were found) were sequentially extracted into the ion exchangeable, hydrous oxide and organic phases present in the sediment. This separation was carried out using solutions of $MgCl_2$, citrate-dithionite and NaOH. The exact procedure for the separations and subsequent analysis of the extracts has been discussed previously.[2]

The results of this investigation confirm earlier studies[2,3] in that the ^{239,240}Pu is primarily associated with the reducible hydrous oxides phase of the sediment and also show that there is no apparent change in the phase distribution with depth in the sediment core. The maximum in the ^{239,240}Pu activity at the 1- to 2-cm interval is normal for this station, being a function of the sedimentation rate.[3] The activity in the 0- to 1-cm interval is identical to that of material collected in a sediment trap set at 5 m above the bottom. The

Table 1. ^{239,240}Pu and ^{241}Am activities in several chemically defined phases of Lake Michigan sediment and sediment trap material (activities in fCi/g dry wt).

Station	$MgCl_2$		Citrate-Dithionite		NaOH	
	^{239,240}Pu	^{241}Am	^{239,240}Pu	^{241}Am	^{239,240}Pu	^{241}Am
0—1	<0.5	--	80.0 ± 4.5	--	<0.5	--
1—2	3.1 ± 0.5	ND	155.0 ± 5.2	35.7 ± 1.3	0.6	0.6
2—3	<0.5	--	27.3 ± 1.5	--	<0.5	--
3—4	<0.5	--	0.6	--	<0.5	--

	Total		$MgCl_2$		Citrate-Dithionite		NaOH	
	^{239,240}Pu	^{241}Am	^{239,240}Pu	^{241}Am	^{239,240}Pu	^{241}Am	^{239,240}Pu	^{241}Am
May—June 60 m Sediment Trap	80.2 ± 13.8	15 ± 2.9	1.3	<0.5	103 ± 10.0	15 ± 1.4	~6.0	<0.5

distribution of ^{239,240}Pu between the phases is also identical within the statistical accuracy of these experiments.

The results shown in Table 1 indicate that the ^{241}Am is also strongly associated with the hydrous oxides in sediment trap materials and the sediments. This similarity in behavior is not unreasonable if it is recognized that a majority of the ^{241}Am is formed in situ by the decay of ^{241}Pu, which is already absorbed on the hydrous oxides, and is probably fixed without migration to another phase.

The similarity in the concentration of ^{239,240}Pu in the upper sediment layer and in the sediment trap material strongly suggests that resuspension of the surface sediments occurs and that this mechanism may be responsible for the remobilization and translocation of plutonium with the sediments.[3,4] To further investigate the possible role of resuspension and mobilization of sediments in the translocation of plutonium, a study was undertaken to determine the distribution of plutonium with respect to the size of the sediment particles. A sample from a separate 0- to 2-cm sediment increment from the same location was subdivided by elutriation into particle sizes, greater than 45 μm, 45 to 20 μm, 20 to 4 μm, 4 to 2 μm, and less than 2 μm.[6] These fractions were dried and analyzed for plutonium. The results are shown in Table 2. Almost 50% of the ^{239,240}Pu occurs in the 20 to 4 μm, or silt, fraction of the sediment which comprises approximately 45% of the sediment by weight. In addition, 80% of the total activity is present in the sedimentary fractions with a size less than

Table 2. Distribution of ^{239,240}Pu activity in various particle size ranges of Lake Michigan sediments.

	Station 5 Core 0—2 cm				
	>45 μm	45—20 μm	20—4 μm	4—2 μm	<2 μm
% of Total Activity	7.9	10.2	50.5	10.9	20.5

20 μm, with almost 20% of the total activity in the clay fraction. This distribution indicates that a major portion of the plutonium in the sediments is associated with particle sizes that are easily resuspended by physical mixing. Analyses have not been completed for the ^{241}Am, so it is not possible to determine at this time if the particle size distribution for this isotope follows that of ^{239,240}Pu as does the chemical phase.

It appears that in Lake Michigan sediments the transuranic elements ^{239,240}Pu and ^{241}Am are associated with reducible hydrous oxides of iron and manganese and hence are inert to chemical mobilization as long as the sediments remain aerobic. However, a significant quantity of ^{239,240}Pu is associated with a highly transportable fraction of the sediment column. The consequences of this association are not fully understood, but it is possible to imagine circumstances in which clay-size particles could be transported with their attendant ^{239,240}Pu to areas where the necessary chemical changes required to remobilize the attached isotopes chemically can occur.

References

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