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Relative Mobilization of Zinc, Cerium, and Americium from Sediment in an Aquatic Microcosm

JOHN P. GIESY and RICHARD A. GEIGER
Savannah River Ecology Laboratory, Aiken, South Carolina

ABSTRACT

The effect of rooted aquatic macrophytes on the relative mobility of ^{65}Zn, ^{144}Ce, and ^{241}Am from sediment was investigated in sediment—water microcosms. The presence of macrophytes resulted in a greater total above-sediment biomass and reduced seston and aufwuchs biomass. Macrophytes also increased the total mass of all three elements in the above-sediment components. This increase was not caused solely by increased biomass into which the isotopes could partition since concentrations of all three isotopes were increased in the water, seston, and aufwuchs as a result of the presence of macrophytes. The ratio of ^{241}Am to ^{144}Ce was the same in seston, aufwuchs, and macrophytes when macrophytes were present. The presence of macrophytes caused a reduction of ^{241}Am relative to ^{144}Ce in the water, seston, and aufwuchs. The dynamics of ^{65}Zn were different from those of ^{144}Ce and ^{241}Am. The ratio of ^{241}Am to ^{65}Zn was approximately sevenfold greater in seston, aufwuchs, and macrophytes than in sediments or water, with macrophytes causing a significant increase in this ratio only in water. Even though the sediments contained a large quantity of stable zinc, the observed behavior of ^{65}Zn could not be attributed to isotopic dilution. Rather ^{65}Zn seems to be restricted from uptake by biota, relative to ^{241}Am and ^{144}Ce. The microcosms studied here were sufficient to study the relative mobility of trace elements from sediments, as well as specific pathways of mobilization, but were not useful for measuring the rate and absolute magnitude of elemental fluxes from sediments, which could be extrapolated to larger systems.

When added to surface waters, many trace elements rapidly become associated with the sediments (Noshkin, 1972; Fowler and Heyraud, 1974; Wahlgren et al., 1976; Livingston and Bowen, 1977). Thus aquatic sediments can act as either sinks or potential reservoirs of toxic trace elements (LeLand, Shukla, and Shimp, 1973). Many of

the trace contaminants released or mobilized into the environment end up in surface waters (Buhler, 1972) directly from effluents and runoff.

There are many mechanisms [e.g., resuspension (Edgington et al., 1976), bioturbation (Renfro, 1973; Beasley and Fowler, 1976), diffusion (Duursma and Hoede, 1967), and active mobilization by plants (Emery, Klopfer, and Garland, 1976)] which may be responsible for mobilization of trace elements from sediments to sites where they can interact with organisms in the water column and potentially re-enter food webs with pathways to man. Even some sediment-dwelling organisms absorb trace elements not from the sediment directly but from the water (Beasley and Fowler, 1976); thus mechanisms that can mobilize trace elements from sediments are important in determining availability to these organisms.

This study was conducted to examine the relative mobility of three types of metals—^{65}Zn (transition), ^{144}Ce (lanthanide inner transition), and ^{241}Am (actinide inner transition)—and to determine the importance of rooted aquatic macrophytes as vectors of mobilization of these metals from sediments. All three of these isotopes are potential environmental contaminants from nuclear fuel cycles. This type of study may be useful in elucidating mechanisms and determining if metallic contaminants behave similarly. Finally, this study was used to evaluate sediment—water microcosms for determining mechanisms and relative magnitudes of elemental fluxes from sediments.

METHODS AND MATERIALS

This experiment was conducted in cylindrical acrylic microcosms (Fig. 1). To facilitate the addition of microcosm components, we constructed each microcosm so that it could be assembled and disassembled by separating the two sections, which were bolted together and sealed with a rubber gasket. Rubber diaphragm sampling ports were spaced along the side so that the water column could be sampled at several depths. Two 40-μm sintered-glass sampling ports were positioned in the sediment region extending through the side of the microcosm and were capped by rubber diaphragm tops. When assembled, the microcosms were placed in a secondary containment vessel filled with water to the same depth as that in the microcosms to minimize the effect of unnatural light penetration of the water column and sediments and to buffer the microcosms from rapid temperature fluctuations. The microcosms

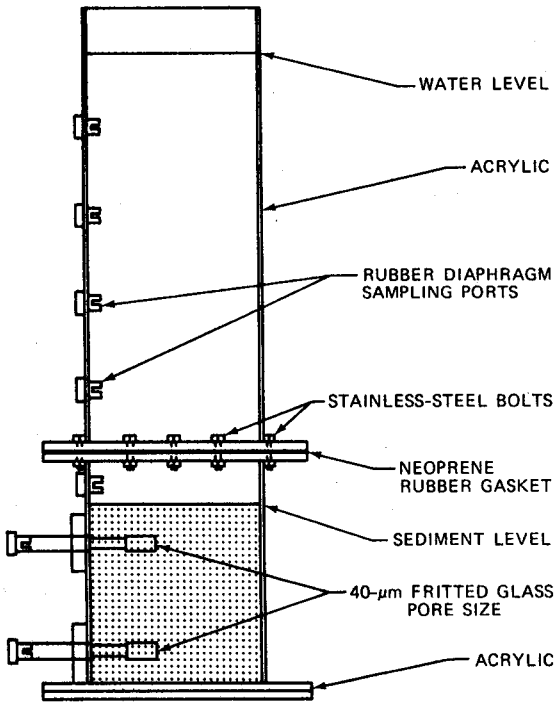

Fig. 1 Schematic drawing of microcosm. Sediment depth, 15.25 cm; volume, 2600 ml; water depth, 38.1 cm; and water volume, 6375 ml.

were assembled and put in place on Oct. 19, 1977, and were disassembled on Apr. 26, 1978, after operating for 207 days.

Organic sediments were collected from Upper Three Runs Creek at the Savannah River Plant (Table 1). There was no detectable ^{241}Am or ^{144}Ce in the sediment, and the stable zinc concentration, which could be extracted from dried sediment by hot concentrated HNO_3, was 0.46 mg Zn/g dry weight (4.2×10^{18} Zn atoms/g). The density of the wet soil was 1.042 g/ml. Zinc-65, ^{144}Ce, and ^{241}Am were added to the sediment in a polyethylene carboy and tumbled for 4 days, at which time all three isotopes were homogeneously mixed with the sediments (Table 2).

Water for the experiment was collected from Skinface Pond, a soft-water, acid pond (Wiener and Giesy, 1979) adjacent to the Savannah River Plant (Table 3). Water levels in the microcosms were maintained by adding distilled water at weekly intervals. The water

TABLE 1

SEDIMENT CHARACTERISTICS

Organic carbon (as C), % by weight	28.4
Sand, %	37.2
Silt, %	30.7
Clay, %	3.6
Cation exchange capacity, meq/100 g	67
Hot HNO_3 extractable Zn, mg/g dry weight	0.46
Wet sediment density, g/ml	1.042

TABLE 2

INITIAL AND FINAL ISOTOPE CONCENTRATIONS IN SEDIMENTS*

Isotope	Initial concentration,† atoms/g dry weight	Final concentration, atoms/g dry weight	
		Macrophytes	No macrophytes
^{65}Zn	$1.5 \pm 0.15 \times 10^{10}$	$8.0 \pm 0.22 \times 10^{9}$	$7.4 \pm 0.8 \times 10^{9}$
^{144}Ce	$9.3 \pm 0.4 \times 10^{10}$	$3.8 \pm 0.12 \times 10^{10}$	$3.6 \pm 0.32 \times 10^{10}$
^{241}Am	$1.5 \pm 0.5 \times 10^{14}$	$7.0 \pm 0.24 \times 10^{13}$	$6.6 \pm 0.57 \times 10^{13}$

*$\bar{X}$ ± 95% confidence interval; n = 4.

†Corrected for radioactive decay to termination of experiment to facilitate comparison.

TABLE 3

MEAN WATER COLUMN CHEMISTRY*

Time	pH	Redox, mv
Macrophytes Present		
Apr. 28, 1978	4.2 ± 0.17	519 ± 4
Apr. 7, 1978	4.9 ± 0.48	423 ± 41
Feb. 17, 1978	5.7 ± 0.16	464 ± 21
Nov. 22, 1977	4.8 ± 0.22	463 ± 18
Macrophytes Absent		
Apr. 28, 1978	4.4 ± 0.05	535 ± 6.2
Apr. 7, 1978	4.7 ± 0.33	431 ± 11
Feb. 17, 1978	5.4 ± 0.42	459 ± 17
Nov. 22, 1977	5.1 ± 0.14	441 ± 12

*$\bar{X}$ ± 1 standard error; n = 4.

columns were aerated by passing compressed air through a hypodermic needle inserted through the third rubber diaphragm from the top of the microcosm.

Approximately equal quantities of the rooted aquatic macrophytes *Dulichium arundinaceum* (L.) Britt. (Cyperaceae) and *Juncus pelocarpus* forma submersys E. Meyer (Juncaceae) were planted in the sediments of four of the microcosms, and four other microcosms received no macrophytes. Colonization of the microcosms with micro- and macroinvertebrates and algae occurred naturally. The sestonic fauna of individual microcosms varied because of fluctuations in the sizes of the populations of ostracods and the cladoceran *Simocephalus serrulatus.*

Water samples were collected from overlying water and from interstitial water several times during the experiment, including just before dismantling the microcosms. Samples were taken into Vac-u-tainers to avoid exposure to the atmosphere. Redox potential and pH were measured with platinum and reference electrodes and a combination glass electrode, respectively. Dissolved oxygen was measured potentiometrically by a modification of the azide modified micro-Winkler method. After 207 days the microcosms were dismantled and separated into their component parts. Water was siphoned from the water column and filtered through 11-cm Whatman No. 41 filters to remove all seston. Periphyton was scraped from the walls and macrophytes were harvested at the sediment—water interface. Macrophyte samples included associated epiphytes. Sediments and biomass were homogenized, dried, and ashed at 400°C before radioassay. Two liters of filtered water from the water column were evaporated to 5 ml by heating before radioassay.

Activity was measured with a 3- by 3-in., well-type NaI detector interfaced to a Northern Scientific model 720 multichannel pulse-height analyzer. All samples were counted until the number of counts accumulated over background fell within 95% confidence limits. Samples were corrected for background and decay to the last day of the experiment and corrected to atoms to facilitate comparisons. Statistical differences between treatment groups were examined by analysis of variance, and multiple comparisons were made with Tukey's W procedure ($\alpha = 0.05$).

RESULTS AND DISCUSSION

The presence of macrophytes increased the total final biomass in the microcosms but reduced the biomass of both seston and periphyton present in the microcosms (Table 4). This result may

TABLE 4

MEAN FINAL STANDING BIOMASS IN MICROCOSMS*

Biomass	Dry weight, g	%
Macrophytes Present		
Macrophytes	6.6 ± 0.60	88.0
Periphyton	0.26 ± 0.042	3.5
Seston	0.61 ± 0.023	8.5
Total	7.5 ± 2.2	100.0
Macrophytes Absent		
Periphyton	0.68 ± 0.058	42.5
Seston	0.93 ± 0.15	57.5
Total	1.6 ± 0.12	100.0

*95% confidence interval; n = 4.

have been caused by competition for nutrients or some other limiting resource; this study was not designed to examine that result, however. In microcosms that did not contain macrophytes, the biomass was equally divided between seston and periphyton. The greater biomass in the microcosms is important in interpreting isotope flux results and will be considered later.

Samples of interstitial water indicated that the sediments at the level of the lower sampling port were anaerobic (Table 5). Samples from the upper sampling port were highly variable but never anaerobic. Water from above the sediments may have been pulled into the sample, causing increased and more variable measurements of O_2 and redox; thus only data for the lower sampling ports are reported. There were no significant differences in the parameters measured in interstitial water between microcosms with macrophytes and those without at the first two samplings. However, the pH was lower and redox potential higher at the last two samplings in microcosms containing macrophytes. The more positive redox and lower pH observed in the sediments of microcosms containing macrophytes may be caused by O_2 transport into the sediments by macrophyte roots.

All the ^{144}Ce and ^{241}Am could be accounted for in the sampled microcosm components, but approximately 40% of the ^{65}Zn was unaccounted for in the final mass balance. Sediment samples taken at the termination of the experiment were from the top 4 cm of sediment. Mass balances for each isotope were calculated assuming a

TABLE 5

MEAN INTERSTITIAL* WATER CHEMISTRY

Time	pH	Redox, mv	Dissolved oxygen, mg/liter
		Macrophytes Present	
Apr. 28, 1978	4.8 ± 0.7	259 ± 82	0
Apr. 7, 1978	5.9 ± 0.08	306 ± 17	0
Feb. 17, 1978	5.7 ± 0.04	200 ± 74	0
Nov. 22, 1977	6.4 ± 0.2	87 ± 19	0
		Macrophytes Absent	
Apr. 28, 1978	5.8 ± 0.2	180 ± 63	0
Apr. 7, 1978	6.3 ± 0.07	101 ± 42	0
Feb. 17, 1978	5.7 ± 0.05	196 ± 76	0
Nov. 22, 1977	6.1 ± 0.03	73 ± 17	0

*Samples from lower interstitial water sampling ports. 95% confidence interval; n = 4.

uniform distribution throughout the sediment. Thus transport into the lower sediments or lack of replacement of ^{65}Zn lost from the upper sediments by ^{65}Zn from the lower sediments may have biased the data. Plant roots were not included in the mass balance, and sediment samples were collected to exclude roots; this may account for some of the discrepancy.

The presence of macrophytes resulted in a significantly greater proportion of all three isotopes in the above-sediment components (Table 6). From these results alone it is not possible to determine whether macrophytes were responsible for mobilizing the isotopes into the above-sediment portion of the microcosm or whether the increased mass of isotopes was caused by a greater biomass for the isotopes to partition into. The concentrations of all three isotopes were increased in the aufwuchs and water (Table 7), however; this indicates that macrophytes actively increased the mass of isotope available in the above-sediment components of the microcosm, as well as providing additional biomass into which the isotopes can partition. Both of these mechanisms are important in mobilizing elements from the sediments and maintaining them in the above-sediment biota, thus increasing their potential contact with some components of the food web.

The presence of macrophytes had no significant effect on the ratio of ^{241}Am to ^{144}Ce in the sediments (Fig. 2). Within

TABLE 6

TOTAL NUMBER OF ATOMS IN ABOVE-SEDIMENT COMPONENTS*

Component	Macrophytes	No macrophytes
	^{65}Zn	
Water	$2.9 \pm 0.9 \times 10^7$ (1.8)	$0.93 \pm 0.03 \times 10^7$ (3)
Seston	$2.2 \pm 1.0 \times 10^8$ (14)	$2.1 \pm 0.54 \times 10^8$ (70)
Aufwuchs	$1.2 \pm 0.1 \times 10^8$ (7.6)	$0.8 \pm 0.2 \times 10^8$ (26)
Macrophytes	$1.2 \pm 0.04 \times 10^9$ (76.5)	
Total	1.6×10^9	3.0×10^8
	^{144}Ce	
Water	$3.4 \pm 1.9 \times 10^8$ (0.56)	$5.0 \pm 3.4 \times 10^7$ (0.4)
Seston	$8.7 \pm 3.6 \times 10^9$ (15)	$8.1 \pm 2.1 \times 10^9$ (67)
Aufwuchs	$5.0 \pm 0.6 \times 10^9$ (8.6)	$3.8 \pm 1.2 \times 10^9$ (32)
Macrophytes	$4.4 \pm 0.78 \times 10^{10}$ (75.8)	
Total	5.8×10^{10}	1.2×10^{10}
	^{241}Am	
Water	$7.0 \pm 4.2 \times 10^{11}$ (0.6)	$7.9 \pm 0.4 \times 10^{11}$ (3.6)
Seston	$1.6 \pm 0.66 \times 10^{13}$ (14)	$1.4 \pm 0.9 \times 10^{13}$ (64)
Aufwuchs	$9.0 \pm 1.2 \times 10^{12}$ (8)	$7.1 \pm 3.0 \times 10^{12}$ (32.4)
Macrophytes	$8.5 \pm 0.27 \times 10^{13}$ (77)	
Total	1.1×10^{14}	2.2×10^{13}

*$\bar{X}$ ± 95% confidence interval; n = 4; % of total in parentheses.

macrophyte treatments the ^{241}Am-to-^{144}Ce ratio was greater in the water than in any other microcosm component. The ^{241}Am-to-^{144}Ce ratio was significantly lower in water, seston, and aufwuchs because of the presence of macrophytes. This response was not caused by selective uptake by or restriction of ^{144}Ce or ^{241}Am from macrophytes because the ratio of these two isotopes in macrophytes was the same as in the sediments. Alternative possibilities are that macrophytes selectively mobilized ^{241}Am from the sediments relative to ^{144}Ce or that the increased redox potential and decreased pH caused by macrophytes facilitated ^{241}Am release from the sediment relative to ^{144}Ce. This increase of ^{241}Am relative to ^{144}Ce may have been enough to cause a significant increase in the water, seston, and aufwuchs without causing a measurable difference between the sediments which contained macrophytes and those which did not.

TABLE 7

ISOTOPE CONCENTRATIONS IN MICROCOSM COMPONENTS AT END OF 207 DAYS*

Component	Macrophytes	No macrophytes
Interstitial water, atoms/liter		
^{65}Zn	$8.1 \pm 0.22 \times 10^{9}$	$7.4 \pm 0.82 \times 10^{9}$
^{144}Ce	$3.8 \pm 0.12 \times 10^{10}$	$3.6 \pm 0.32 \times 10^{10}$
^{241}Am	$7.0 \pm 0.23 \times 10^{13}$	$6.6 \pm 0.57 \times 10^{13}$
Water, atoms/liter		
^{65}Zn†	$4.3 \pm 1.2 \times 10^{6}$	$1.5 \pm 0.06 \times 10^{6}$
^{144}Ce†	$5.4 \pm 3.1 \times 10^{7}$	$7.8 \pm 5.3 \times 10^{6}$
^{241}Am†	$1.1 \pm 0.66 \times 10^{11}$	$1.5 \pm 0.67 \times 10^{10}$
Seston, atoms/g dry weight		
^{65}Zn	$3.5 \pm 1.6 \times 10^{8}$	$2.3 \pm 0.6 \times 10^{8}$
^{144}Ce	$1.4 \pm 0.6 \times 10^{10}$	$8.7 \pm 2.3 \times 10^{9}$
^{241}Am	$2.7 \pm 1.1 \times 10^{13}$	$1.8 \pm 0.4 \times 10^{13}$
Aufwuchs, atoms/g dry weight		
^{65}Zn†	$4.5 \pm 0.48 \times 10^{8}$	$1.2 \pm 0.28 \times 10^{8}$
^{144}Ce†	$1.9 \pm 0.24 \times 10^{10}$	$5.5 \pm 1.7 \times 10^{9}$
^{241}Am†	$3.5 \pm 0.46 \times 10^{13}$	$1.1 \pm 0.4 \times 10^{13}$
Macrophytes, atoms/g dry weight		
^{65}Zn	$1.7 \pm 0.32 \times 10^{8}$	
^{144}Ce	$6.6 \pm 1.2 \times 10^{9}$	
^{241}Am	$1.8 \pm 1.0 \times 10^{13}$	

*$\bar{X} \pm$ 95% confidence interval; n = 4.

†There is a significant difference due to macrophytes in the concentration of this isotope in this component ($\alpha = 0.05$).

Zinc-65 behaved rather differently from ^{241}Am (Fig. 3). The ^{241}Am-to-^{65}Zn ratio was the same in sediments with and without macrophytes and was similar in seston, periphyton, and macrophyte tissues, which had much greater ratios than did the water and sediment.

On the basis of the amount of zinc extractable by hot, concentrated HNO_3, there was a factor of 10^8 more stable zinc in the sediments than the ^{65}Zn added. The observed results were not caused by isotopic dilution because there was no increase in the ^{241}Am-to-^{65}Zn ratio in the water relative to that in the sediments. This indicates that little of the stable zinc extracted by hot HNO_3 was available for isotopic dilution. Wagemann, Brunskill, and Graham (1977) found that between 72 and 98% of the zinc in bottom sediment could not be easily removed. Similarly, Benes and Steinnes (1976) found that isotopic exchange between introduced ^{65}Zn and stable zinc present in an aquatic system was very slow, and

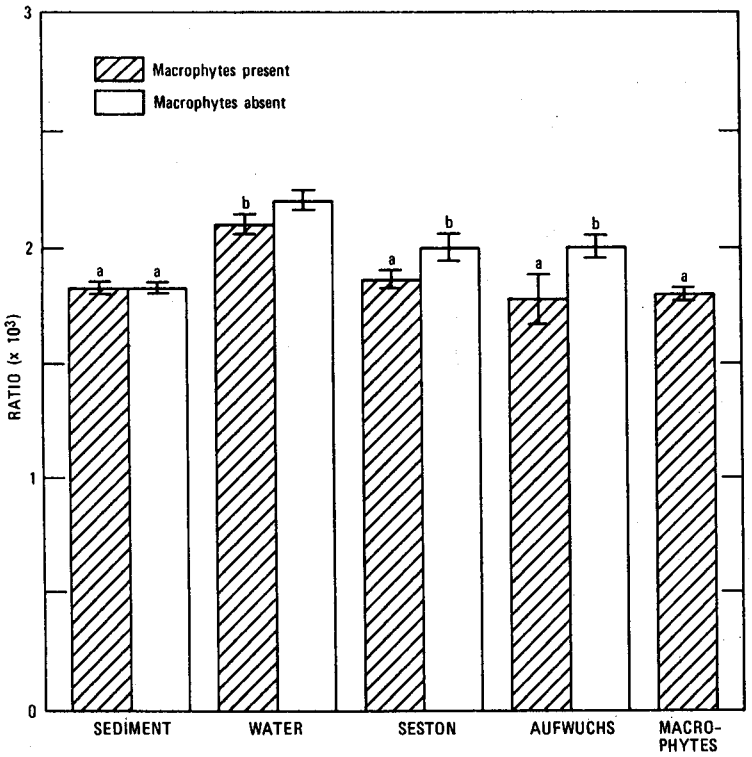

Fig. 2 Ratio of ^{241}Am to ^{144}Ce (atom basis) in microcosm components at the termination of the experiment ($\bar{X}$ ± 95% confidence interval). Means denoted by the same letter are not significantly different from one another (Tukey's W procedure, $\alpha = 0.05$, n = 4).

substantial differences between stable and tracer zinc remained after 35 days. This also indicates that the ^{65}Zn added cannot be used to model the behavior of zinc already present in the sediment and that the process by which zinc is bound into the sediments is rather slow. Benes and Steinnes (1976) also concluded that radiotracer studies have serious limitations in the field because of this disequilibrium unless all the tracer and stable elements exist as free ions at the beginning of the experiment. We wish to reiterate these caveats, which complicate the interpretation of results from tracer studies if this disequilibrium situation exists for inputs of elements or compounds previously in the study system. Because of isotopic dilution, specific activities should be measured in all components

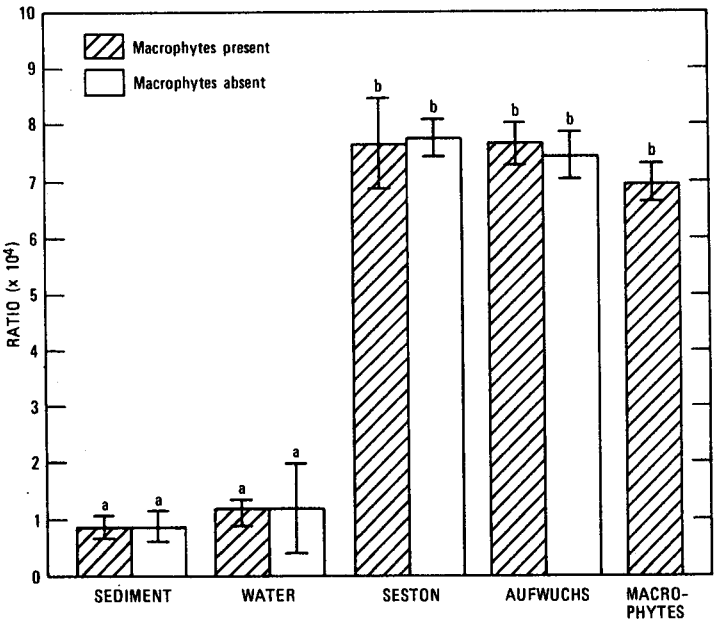

Fig. 3 Ratio of ^{241}Am to ^{65}Zn (atom basis) in microcosm components at the termination of the experiment ($\bar{X}$ ± 95% confidence interval). Means denoted by the same letter are not significantly different from one another (Tukey's W procedure, $\alpha = 0.05$, n = 4).

when a tracer is added to a system containing the stable element or compound (Atkins, 1969).

The presence of macrophytes reduced the concentration factors of ^{144}Ce and ^{241}Am into seston and periphyton (Fig. 4). This was a result of increased water concentrations of the isotopes in the presence of macrophytes, whereas the seston and periphyton were apparently at a saturated steady state.

The concentration ratios between water and seston, aufwuchs, or macrophytes were lower for ^{65}Zn than for ^{144}Ce or ^{241}Am (Fig. 4). Since this result was probably not caused by isotopic dilution, uptake of ^{65}Zn by biota seems to be restricted relative to ^{144}Ce and ^{241}Am. We suggest that this may be a result of homeostatic regulation of zinc, which is a required element.

Livingston and Bowen (1977) found that ^{241}Am concentration factors of planktonic algae were one or two orders of magnitude below those of attached algae. No such difference was observed in this study. Livingston and Bowen also reported that ^{241}Am

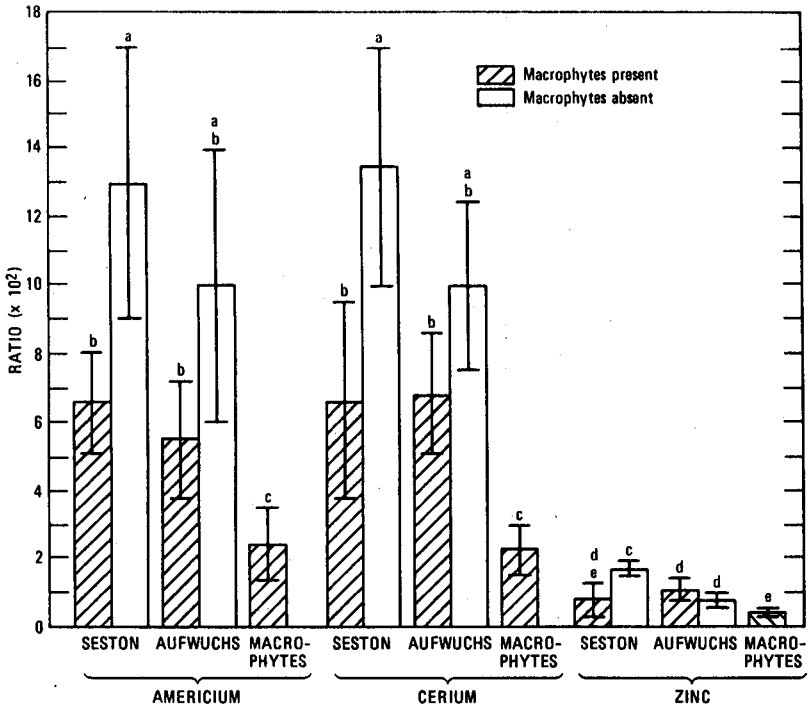

Fig. 4 Concentration ratios (atoms in tissue/atoms in water) of ^{144}Ce, ^{241}Am, and ^{65}Zn in seston, aufwuchs, and macrophytes ($\bar{X} \pm 95\%$ confidence interval). Means designated by the same letter are not significantly different from one another (Tukey's W procedure, $\alpha = 0.05$, n = 4).

concentration factors into plants were similar to those of other trace metals; this indicates no discrimination against ^{241}Am relative to other elements. The hydrated ionic radii of ^{65}Zn and ^{144}Ce are similar, and Dawson and Duursma (1974) found that concentration factors in soils and plankton were the same for the two elements.

We would expect the behavior of ^{144}Ce and ^{241}Am, both inner-transition metals, to be more similar to each other than to ^{65}Zn, a transition metal (Garder and Skulberg, 1964). Because the lanthanide elements all have a similar outside 5p electron shell, they exhibit similar chemical behavior and would be expected to exhibit similar environmental chemistries. In fact, ^{144}Ce and ^{241}Am did behave similarly relative to ^{65}Zn. The presence of macrophytes caused differences in their behavior, however. These differences may have been a result of redox and chelation effects since cerium exists

in the +3 and +4 oxidation states and americium exists in the +5, as well as +3 and +4. Approximately 30% of the ^{241}Am in the sediments studied by Emery, Klopfer, and Garland (1976) was removed by oxalate extraction; this suggests that ^{241}Am was associated with hydrous oxides (Means et al., 1978). Reducing conditions will dissolve iron and manganese oxides, perhaps releasing other trace elements associated with the oxide layers. Ceric hydroxide precipitation and complexing are important in determining the form and availability of cerium (Carpenter and Grant, 1967; Hirano and Koyanagi, 1978). Thus the flux of cerium from sediments is a pH-dependent process (Patel, Patel, and Pawar, 1978) and would be expected to be similar to that of americium. We would expect to see an increase in ^{241}Am relative to ^{65}Zn in the anaerobic interstitial water since ZnS has a low solubility (Holmes, Slade, and McLerran, 1974) and americium release is favored by low pH and low Eh. Conditions favoring zinc release should be the opposite of those favoring americium release (Lu and Chen, 1977).

Microcosms of the type described here are appropriate and sufficient to study the relative mobility of compounds or elements into or out of sediments and to examine the relative accumulation of the element or compound of interest in sediments, water, aufwuchs, seston, and macrophytes. Because of its design, this type of microcosm is especially suited to the study of cycling processes in the littoral zone, but it is not adequate to study profundal processes because of differences in light and pressure. The microcosm described here could be modified for use in field situations by removing the bottom and pushing the acrylic cylinder into the sediments. The microcosms can also be filled with the sediment of interest and placed in the field. The water quality overlying the sediments can be manipulated to study effects of varying pH, redox, O_2 concentration, ionic strength, or concentration of particular ions or molecules. The system can also be used to determine how different sediments will react under similar water qualities.

Because the microcosm is inexpensive, contains a small volume, and does not release elements or compounds to the environment, it is easily replicable and amenable to tracer studies. It is particularly useful because it allows calculation of mass balances and recovery of transformation products in the study of organics.

We feel that microcosms of this type are not sufficient to measure rates or absolute magnitudes of flux into or from sediments because of wall effects, reduced turbulence, and small overlying volume. In areas of dense macrophyte growth, the turbulence difference between the natural area and the microcosm may be small.

These limitations make this type of microcosm inappropriate for verification of global transport models; however, it is appropriate and sufficient to test some mechanisms. Levins (1974) studied the extrapolation of microcosm-determined material transfers to ecosystem transfers and concluded that appropriate scale-up and extrapolation procedures can be devised.

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