

FURTHER INVESTIGATIONS OF PLUTONIUM IN AQUATIC BIOTA OF THE GREAT MIAMI RIVER WATERSHED INCLUDING THE CANAL AND PONDS IN MIAMISBURG, OHIO

C. W. Wayman, G. E. Bartelt, and S. E. Groves

A preliminary investigation of plutonium in aquatic organisms of the Great Miami River Watershed, Ohio, has been conducted.¹ This report is a continuation of that study to include new species and additional samples of old species. For the first time data are available on the plutonium content of aquatic biota from the canal and ponds located adjacent to the Mound Laboratory. These areas have elevated levels of ²³⁸Pu in the water and sediments as the result of a past incident. It is the purpose of this report not only to record plutonium activities in aquatic biota but to study trophic level relationships and investigate the mode of plutonium uptake by organisms. Radiochemical analysis of biota follows the methods described by Nelson et al.²

Aquatic plants from the watershed (Table 1) were sampled both upstream and downstream from the effluent pipe of Mound Laboratory. Plants collected downstream of the laboratory concentrate more ²³⁸Pu than the plants located upstream by two to three orders of magnitude. Activities of ²³⁸Pu in background samples are unexpectedly high and may be attributed to contamination. Activities of ^{239,240}Pu in the plants from upstream and downstream are approximately the same.

Aquatic macrophytes, Potamogeton and Myriophyllum, sampled from Franklin contain approximately the same concentrations of plutonium. The green alga, Cladophora, sampled downstream from Mound Laboratory, appears to concentrate ²³⁸Pu more than the macrophytes by an order of magnitude.

Cladophora sampled at the Franklin site was transplanted with its substrata intact from Bear Creek, a tributary of the Great Miami River situated upstream from Mound Laboratory. The Cladophora shows an immediate uptake of ²³⁸Pu of at least one order of magnitude within 10 hours after transplanting. Another slight increase occurs after an additional 5 hours. It is probable that the Cladophora had reached an equilibrium level and that the second increase represents sample variability. The rapid accumulation of plutonium by

Table 1. ^{238}Pu and $^{239,240}\text{Pu}$ activities (pCi/kg wet wt)^(a) in aquatic plants from the Great Miami River watershed, Ohio.

Sample Type and Location	Date	^{238}Pu	$^{239,240}\text{Pu}$	$^{238}\text{Pu}/^{239,240}\text{Pu}$
<u>Cladophora</u>				
Rip Rap Bridge	5 Nov 74	0.15 ± 0.04	0.22 ± 0.05	0.7 ± 0.2
Rip Rap Bridge	9 June 75	1.2 ± 0.4	2.0 ± 0.4	0.6 ± 0.2
Mad River	10 Sept 74	0.15 ± 0.08	0.09 ± 0.07	2.0 ± 2.0
Stillwater River	11 Sept 74	0.2 ± 0.2	0.7 ± 0.2	0.3 ± 0.3
Bear Creek	24 June 75	<1	N.D.	
Bear Creek	5 Nov 75	<0.5	N.D.	
Franklin	24 June 75	15 ± 1	0.6 ± 0.3	25 ± 10
Bonham	16 Sept 75	4.4 ± 0.4	0.2 ± 0.1	22 ± 10
Canal	12 Sept 75	910 ± 20	2 ± 1	450 ± 200
<u>Potamogeton</u>				
Mad River	10 Sept 74	0.05 ± 0.01	0.13 ± 0.02	0.4 ± 0.1
Chautauqua	23 July 74	± 0.4	0.22 ± 0.04	150 ± 20
Franklin	24 July 74	1.9 ± 0.1	<0.03	>60
Franklin	14 Aug 74	8.6 ± 0.4	0.24 ± 0.07	36 ± 10
Franklin	24 June 75	5.1 ± 0.4	0.07 ± 0.06	72 ± 60
Franklin	16 Sept 75	1.4 ± 0.1	0.18 ± 0.05	8 ± 2
<u>Myriophyllum</u>				
Franklin	10 June 75	4.1 ± 0.2	0.09 ± 0.04	44 ± 20
Franklin	24 June 75	4.8 ± 0.3	0.05 ± 0.04	95 ± 70
Franklin	16 Sept 75	4.2 ± 0.4	0.5 ± 0.2	9 ± 3
<u>Duckweed</u>				
Canal	7 Nov 74	800 ± 20	5 ± 1	160 ± 40
Canal	13 May 75	$2,900 \pm 30$	8 ± 2	340 ± 70
South Pond	10 June 75	380 ± 20	0.7 ± 0.7	500 ± 500
Canal	12 Sept 75	$1,000 \pm 20$	2.1 ± 0.7	500 ± 200
Canal	15 Oct 75	840 ± 20	8 ± 2	100 ± 30
Canal	17 Dec 75	640 ± 9	2.7 ± 0.6	240 ± 50
<u>Cattails</u> ^(b)				
North Pond	13 May 75	1.9 ± 0.1	<0.05	>40
Canal	13 May 75	7.5 ± 0.4	<0.09	>80
Canal	10 June 75	2.8 ± 0.2	0.42 ± 0.07	7 ± 1

(a) The \pm value is 1σ counting error. When the counting error is $>100\%$, the concentration is recorded as $<2\sigma$.

(b) Whole plants minus the roots.

N.D. = not detectable.

Cladophora suggests that uptake is probably more a function of adsorption than absorption. A study of the distribution of plutonium in giant brown algae showed that most of the activity was associated with the outer surfaces.³

The accumulation of plutonium in crayfish and fish species from the river was also studied (Table 2). Activities of ^{239, 240}Pu are approximately the same in all samples from both above and below the Mound Laboratory effluent pipe. The activities of ²³⁸Pu in all upstream samples are one to three orders of magnitude less than the levels in specimens from downstream. Downstream, crayfish contain twice as much ²³⁸Pu as goldfish and minnows. Also, the crayfish contain two to three orders of magnitude more ²³⁸Pu than carp minus gastrointestinal tracts; however, their activity is almost equal to that of shad collected downstream.

In a separate study of crayfish from the Great Miami River, it was found that most of the plutonium was concentrated in soft tissues rather than in the sclerotized shell. Similar results have been found for the Tridacna clam and lobster from Eniwetok Atoll.⁴ In other studies, however, shell or skeletal portions of animals contain higher levels of plutonium than the soft tissue.⁵⁻⁷

A comparison among four species of river fish shows that shad (only one sample) had the greatest ²³⁸Pu activities. Minnows and goldfish contain the same level of ²³⁸Pu, which is about half the value exhibited in shad. The ²³⁸Pu activity in carp minus gastrointestinal tracts and in their GI tracts combined is lower than the activities in the other fish species, with the exception of one GI sample. The higher level of ²³⁸Pu in this sample is attributed to sediment present in the gastrointestinal tract.

Biological samples have also been analyzed from the canal and ponds which are known to contain elevated levels of ²³⁸Pu. Aquatic plants collected from these sites are duckweed, cattail, and Cladophora (Table 1). Duckweed contains the highest levels of plutonium of all the biota sampled from the watershed. Duckweed from the canal is two to eight times higher in ²³⁸Pu activity than duckweed from the ponds, reflecting the slight difference that exists in ²³⁸Pu activity levels of the two bodies of water. ²³⁸Pu activities in the canal average about 1.0 pCi/1 as compared to 0.7 pCi/1 in the ponds.

Table 2. ^{238}Pu and $^{239,240}\text{Pu}$ activities (pCi/kg wet wt)^(a) in crayfish and fish from the Great Miami River, 1974.

Sample Type and Location	Date	Number of Individuals	Average Total Length (cm)	^{238}Pu	$^{239,240}\text{Pu}$	$^{238}\text{Pu}/^{239,240}\text{Pu}$
<u>Crayfish (whole)</u>						
Rip Rap Bridge	11 Sept	1	<0.1	0.14 \pm 0.08	<0.7	
Chautauqua	23 July	0.6 (b)	6.2 \pm 0.4	0.11 \pm 0.06	56 \pm 30	
Chautauqua	13 Aug	0.8 (b)	8.6 \pm 0.4	0.09 \pm 0.05	96 \pm 50	
Franklin	14 Aug	0.5	8.7 \pm 0.4	0.03 \pm 0.03	300 \pm 300	
<u>Carp (minus GI)</u>						
Dayton	17 Nov	3	0.009 \pm 0.001	0.022 \pm 0.005	4 \pm 1	
Chautauqua (c)	15 Nov	4	44.8	0.25 \pm 0.09	0.04 \pm 0.02	6 \pm 4
Chautauqua (c)	13 Aug	1	54.0	0.17 \pm 0.02	0.024 \pm 0.008	7 \pm 2
Chautauqua (c)	15 Nov	1	48.0	0.031 \pm 0.004	<0.003	>10
Chautauqua	15 Nov	1	54.0	0.054 \pm 0.006	0.029 \pm 0.004	1.9 \pm 0.4
<u>Carp GI (for above)</u>						
Dayton	17 Nov		0.05 \pm 0.01	0.010 \pm 0.005	5 \pm 3	
Chautauqua	15 Nov		0.05 \pm 0.02	N.D.		
Chautauqua	13 Aug		6.5 \pm 0.3	N.D.		
Chautauqua	15 Nov		0.15 \pm 0.04	N.D.		
Chautauqua	15 Nov		0.09 \pm 0.02	0.16 \pm 0.03	0.6 \pm 0.2	
<u>Goldfish (whole)</u>						
Chautauqua	23 July	17	15.1	3.12 \pm 0.09	0.03 \pm 0.01	100 \pm 30
Chautauqua	13 Aug	15	15.2	5.5 \pm 0.2	0.47 \pm 0.06	12 \pm 2
Chautauqua	14-15 Aug	5	18.1	3.2 \pm 0.1	0.07 \pm 0.02	46 \pm 10
Franklin	14 Aug	13	13.0	1.35 \pm 0.07	0.011 \pm 0.006	120 \pm 70
<u>Minnows (whole)</u>						
Rip Rap Bridge	10-11 Sept	1 (b)		0.11 \pm 0.02	0.04 \pm 0.01	2.4 \pm 0.6
Chautauqua	25 July	1 (b)		0.73 \pm 0.04	<0.01	>70
Chautauqua	13 Aug	1 (b)		5.7 \pm 0.2	0.19 \pm 0.03	30 \pm 5
<u>Shad (whole)</u>						
Rip Rap Bridge	10-11 Sept.			0.011 \pm 0.004	0.15 \pm 0.01	0.08 \pm 0.03
Chautauqua	25 July					
	13 Aug			- 10.7 \pm 0.2	0.13 \pm 0.02	82 \pm 10
	6 Nov					

(a) The \pm value is 1σ counting error. When the counting error is $>100\%$, the concentration is recorded as $<2\sigma$.

(b) kg wet weight.

(c) Samples collected by DePauw University, Greencastle, Indiana.

N.D. = not detectable.

The study of plutonium activities in aquatic organisms presented in this report does not mean to imply that complete food chains have been examined. The plants and animals investigated represent different trophic levels, which may or may not be interrelated. Generally, aquatic plants accumulate more plutonium than either crayfish or fish. Crayfish appear to concentrate ^{238}Pu to a greater extent than fish with the possible exception of shad. This report reiterates the finding of previous studies that plutonium is discriminated against at higher trophic levels.

References

1. C. W. Wayman, G. E. Bartelt, and D. N. Edgington, Plutonium in aquatic biota of the Great Miami River Watershed, Ohio, Radiological and Environmental Research Division Annual Report, January-December 1974, ANL-75-3, Part III, pp. 78-86.
2. D. M. Nelson, E. M. Yaguchi, B. J. Waller, and M. A. Wahlgren, Radiochemical methods, Radiological and Environmental Research Division Annual Report, January-December 1973, ANL-8060, Part III, pp. 6-17.
3. K. M. Wong, V. F. Hodge, and T. R. Folson, Plutonium and polonium inside giant brown algae, *Nature* 237, 460-462 (1972).
4. V. Nelson and V. E. Noshkin, Eniwetok Radiological Survey, U.S. Atomic Energy Commission, Nevada Operations Office, NOV-140 (1973).
5. A. Aardrog, Radioecological investigations of plutonium in an arctic marine environment, *Health Phys.* 20, 31-47 (1971).
6. V. E. Noshkin, Ecological aspects of plutonium dissemination in aquatic movements, *Health Phys.* 22, 537-549 (1972).
7. E. E. Ward, Uptake of plutonium by the lobster, *Homarus vulgaris*, *Nature* 209 625-626 (1966).