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SPATIAL DISTRIBUTION OF RADIONUCLIDES IN LAKE MICHIGAN BIOTA NEAR THE BIG ROCK POINT NUCLEAR PLANT

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SPATIAL DISTRIBUTION OF RADIONUCLIDES IN LAKE MICHIGAN BIOTA NEAR THE BIG ROCK POINT NUCLEAR PLANT*

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ABSTRACT

A survey was made of four groups of biota in the vicinity of the Big Rock Point Nuclear Plant near Charlevoix, Michigan, to determine their usefulness in locating possible sources of plutonium and other radionuclides to Lake Michigan. This 70 MW boiling-water reactor, located on the Lake Michigan shoreline, was chosen because its fuel contains recycled plutonium, and because it routinely discharges very low-level radioactive wastes into the lake. Samples of crayfish (Orconectes sp.), green algae (Chara sp. and Cladophora sp.), and an aquatic macrophyte (Potamogeton sp.) were collected in August, 1973, at varying distances from the discharge and analyzed for $^{239,240}Pu$, ^{90}Sr, and five gamma-emitting radionuclides. Comparison samples of reactor waste solution have also been analyzed for these radionuclides.

Comparisons of the spatial distributions of the extremely low radionuclide concentrations in biota clearly indicated that ^{137}Cs, ^{134}Cs, ^{65}Zn, and ^{60}Co were released from the reactor; their concentrations decreased exponentially with increasing distance from the discharge. Conversely, concentrations of $^{239,240}Pu$, ^{95}Zr, and ^{90}Sr showed no correlation with distance, suggesting any input from Big Rock was insignificant with respect to the atmospheric origin of these isotopes.

The significance of these results is discussed, particularly with respect to current public debate over the possibility of local environmental hazards associated with the use of plutonium as a nuclear fuel.

INTRODUCTION

Since the beginning of the "nuclear age" some thirty years ago, the number of sources of controlled release of radioactive material to the environment has been gradually increasing. With the exception of very localized waste disposal areas, concentrations of radionuclides are, in general, orders of magnitude below those levels which would pose a health hazard because of alpha, beta, or gamma radiations. Nevertheless, the sources and fate of these materials are of great interest, particularly in relatively undisturbed natural ecosystems. Lake Michigan, with the second largest volume of the Laurentian Great Lakes, is such an aquatic ecosystem. It also is one which is experiencing increased utilization as a natural resource by the developing industrial area surrounding it. The siting of six operational nuclear power plants along the shore has focused increased attention upon the lake's radioecology. Sources of radioactive material to be considered include atmospheric fallout (both from natural processes and weapons testing), routine releases of low-level radioactive wastes from nuclear reactors, and non-routine releases from such potential sources as reactors and the transportation of radioactive material.

With sophisticated instrumentation, very low concentrations of several radionuclides may be accurately measured in the biota of Lake Michigan. This study was designed to consider attached or sedentary (i.e., not migratory) organisms as spatial biological monitors. Varying

concentrations of radioisotopes in spatially separated groups of organisms could reflect different sources of radioactivity and/or differences in local environments affecting the assimilation of any given element.

Major sampling locations were located near the Big Rock Point Nuclear Plant, Charlevoix, Michigan. This 70 MW boiling-water reactor, which has been in operation since 1962, routinely discharges very dilute, low-level radioactive wastes into Lake Michigan. Earlier studies (Nelson et al., 1971; and Nelson et al., 1972) have surveyed the distribution of ^{137}Cs, ^{134}Cs, ^{65}Zn, and ^{60}Co in biota near this reactor. Also, the recent increase in use of recycled plutonium as a portion of the Big Rock reactor's fuel (USAEC, 1972) has prompted local debate over possible regional environmental hazards associated with the handling of larger amounts of mixed Pu-U oxide fuels. The present study compares the concentrations of $^{239,240}Pu$, ^{90}Sr and gamma-emitting radionuclides in littoral-zone organisms near this reactor and those at control sites elsewhere in Lake Michigan.

METHODS

Samples of crayfish (Orconectes sp.), green algae (Chara sp. and Cladophora sp.), and an aquatic macrophyte (Potamogeton sp.) were collected by hand in shallow water and frozen for storage. Collections at various distances up to 4 km from the Big Rock reactor were made 22 August 1973, 21 days after the most recent radioactive release (Axtell, personal

communication). Control samples (subject only to atmospheric fallout and natural redistribution processes within the lake) were collected as follows: Orconectes, 73 km NNE of Big Rock, Chara and Potamogeton, 84 km NW of Big Rock on 30 August 1973; and Cladophora at nine locations around the lake from July through September, 1973. The choice of these locations was based primarily upon availability of the organisms being studied.

All biological samples were dried at 105°C; typical percent dry weights were 23% for Orconectes, 12% for Cladophora, 21% for Chara, and 13% for Potamogeton. The samples were then ashed to constant weight at 500°C, and portions of the ash were analyzed for plutonium by the method of Nelson et al. (1974). This procedure involves dissolution of the ash in acid, separation of plutonium by anion exchange and electrodeposition, and resolution of ^{238}Pu and $^{239,240}Pu$ by alpha spectrometry. (The isotopes ^{239}Pu and ^{240}Pu have nearly identical alpha energies and cannot be distinguished by this method.) Strontium-90 was determined by chemical separation and measurement of the beta activity of the ^{90}Y daughter. Gamma-ray emitting radionuclides were detected with a 10 x 10 cm NaI(Tl) crystal coupled to a multichannel analyzer, and the resulting spectra resolved by computer least-squares analysis. Sample sizes for gamma-ray spectrometric analyses ranged from 70 to 1700 g fresh weight, whereas plutonium and strontium were separated from 1 to 10 g of the ash. In addition, samples of the liquid waste released from the Big Rock Plant to Lake Michigan in 1970, 1971, and 1974 were

RESULTS AND DISCUSSION

Table 1 includes the concentrations of selected radionuclides in the biological samples. Two isotopes which are common to atmospheric fallout are ^{95}Zr and ^{90}Sr (HASL, 1974); as expected, their concentrations are independent of distance from the reactor. Of major significance is the observation that $^{239,240}Pu$ shows a similar behavior. There are no consistent trends in $^{239,240}Pu$ levels among the four groups of biota. Chara and Potamogeton have lower $^{239,240}Pu$ levels near Big Rock than at control sites, while concentrations in Orconectes are not significantly different between the locations. The mean concentration of plutonium-239,240 in Cladophora near Big Rock was significantly higher than the mean concentration in control samples (Student t test, $\alpha = 0.01$ level), but was within the range of the control concentrations. The observed variations may reflect differences in physiological state among the samples or some environmental parameter. These four genera are not ubiquitous along the lake shore, and are sensitive to local nutrient sources, wave action, substrate composition, etc.

As in the earlier study by Nelson et al. (1971), the concentrations in biota of reactor-produced isotopes (Sinderman and Axtell, 1972) show somewhat differing rates of decrease with increasing distance from the discharge. These results are illustrated in Figure 1, which also demonstrates the apparent independence of $^{239,240}Pu$, ^{95}Zr, and ^{90}Sr concentrations with respect to distance as discussed above. In the graphs all concentrations have been normalized to those for crayfish for purposes of comparison with the 1971 study. (An exception is the case of ^{95}Zr

which is not detectable in crayfish.) The differences among the reactor-produced isotopes may result from their existence in varying physicochemical states (Robertson, 1971), and/or from differences in their respective biological half-lives. The two isotopes of cesium, which would be expected to exist in the same chemical form, have different spatial relationships; this reflects the substantial background levels of ^{137}Cs from atmospheric input as is shown by the ^{137}Cs concentrations in the "control" samples.

While ^{238}Pu is present in such low concentrations in Lake Michigan as to severely limit its study, certain additional information can be obtained from consideration of ^{238}Pu:$^{239,240}Pu$ activity ratios. The ^{238}Pu:$^{239,240}Pu$ activity ratio in the samples of reactor radioactive waste averaged 45% reflecting the isotopic composition of the PuO_2 contained in the Big Rock reactor (Smith et al., 1973). In contrast, the observed ratio in biological samples from control locations and near Big Rock averaged about 7%.

Plutonium-239,240 are known components of weapons-test fallout (HASL, 1974) and are present in Lake Michigan water with a mean combined concentration of 0.7 fCi/l (Wahlgren and Nelson, 1974). As a further check on the possibility of reactor waste as a significant source term, analyses of the waste release samples are reported in Table 2. While no discharge samples were available at the time the biota was sampled, there was no known procedural change in the handling of wastes at Big Rock between 1970 and 1974, and the 1974 $^{239,240}Pu$ concentrations are shown to be comparable to those which existed in 1970 and 1971. The concentration of $^{239,240}Pu$ in this waste was found to be approximately

70,000 times that in the ambient lake water, but with the dilution of the typical release volume of only 19,000 liters (Axtell, personal communication) with the total volume (at a rate of 200,000 liters/min) of lake water used for the plant's once-through cooling (Sinderman and Axtell, 1972), one would not expect to see a substantial effect of this source of plutonium.

In order to estimate average concentrations of reactor-produced radionuclides to which attached and local organisms may be exposed in the vicinity of the Big Rock Plant, a comparison is made in Table 2 of the background levels in Lake Michigan water and in the cooling water discharge. It can be seen that the biota serve as a useful integrating and concentrating indicator of the radionuclides that are discharged at elevated concentrations, and that at the present time the discharge of plutonium is so low as to be undetectable in the presence of background (fallout) plutonium levels.

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TABLE 1. Radionuclide concentrations in Lake Michigan biota near the Big Rock Point Nuclear Plant.

Sample	Distance, direction from plant	Concentrations, pCi/g wet weight ± S.E.[a]							
		^{239}Pu	^{238}Pu	^{137}Cs	^{134}Cs	^{95}Zr	^{90}Sr	^{65}Zn	^{60}Co
Oncorhynchus	.2 km E	0.32 ± .04	.02 ± .01	311 ± 6	61 ± 4	< 14	492 ± 5	556 ± 11	288 ± 3
Oncorhynchus	2 km E	0.22 ± .03	.02 ± .01	170 ± 5	26 ± 3	< 13	439 ± 4	95 ± 6	13 ± 2
Oncorhynchus	4 km E	0.20 ± .03	.03 ± .01	121 ± 5	19 ± 3	< 11	557 ± 5	44 ± 6	< 6
Oncorhynchus	73 km NNE	0.33 ± .04	< .03	78 ± 1	< 50	< 30	471 ± 9	< 100	< 30
Cladophora	.3 km E	4.7 ± .2	.45 ± .05	204 ± 4	81 ± 21	194 ± 12	56 ± 5	< 120	51 ± 14
Cladophora	2 km E	4.3 ± .2	.21 ± .04	209 ± 4	23 ± 4	177 ± 6	53 ± 3	< 17	55 ± 3
Cladophora	4 km E	5.2 ± .2	.30 ± .03	57 ± 3	< 10	259 ± 9	36 ± 1	< 5	< 6
Cladophora	(b)	2.4 ± .2	.16 ± .03	27 ± 3	< 9	142 ± 31	25 ± 7	< 10	< 6
Chara	.3 km E	2.7 ± .2	.21 ± .07	591 ± 6	117 ± 6	16 ± 1	324 ± 6	186 ± 11	738 ± 7
Chara	2 km E	3.0 ± .2	.23 ± .06	186 ± 4	17 ± 3	9 ± 1	316 ± 6	17 ± 5	39 ± 2
Chara	4 km E	3.5 ± .3	.25 ± .05	159 ± 5	< 7	15 ± 1	306 ± 6	11 ± 9	16 ± 5
Chara	84 km NW	5.2 ± .1	.28 ± .03	238 ± 3	7 ± 2	12 ± 1	lost	< 15	< 5
Potamogeton	.3 km E	1.3 ± .1	.11 ± .04	272 ± 5	42 ± 5	15 ± 1	51 ± 2	121 ± 10	152 ± 3
Potamogeton	.8 km E	.98 ± .04	.06 ± .01	124 ± 8	44 ± 9	5 ± 3	56 ± 1	88 ± 17	78 ± 7
Potamogeton	2 km E	.71 ± .06	.06 ± .02	48 ± 1	15 ± 6	15 ± 2	46 ± 2	29 ± 10	7 ± 4
Potamogeton	4 km E	1.5 ± .1	.10 ± .02	35 ± 1	< 12	14 ± 1	63 ± 1	< 22	5 ± 3
Potamogeton	84 km NW	2.2 ± .1	.11 ± .01	90 ± 2	< 7	14 ± 5	62 ± 1	< 20	< 5

(a) When the one standard deviation counting error was > 100%, the concentration is recorded as < 3σ.

(b) Mean values of 10 samples (except ^{90}Sr, which is mean of 2 samples) ..cated from 73 to 400 km from the plant, in all directions.

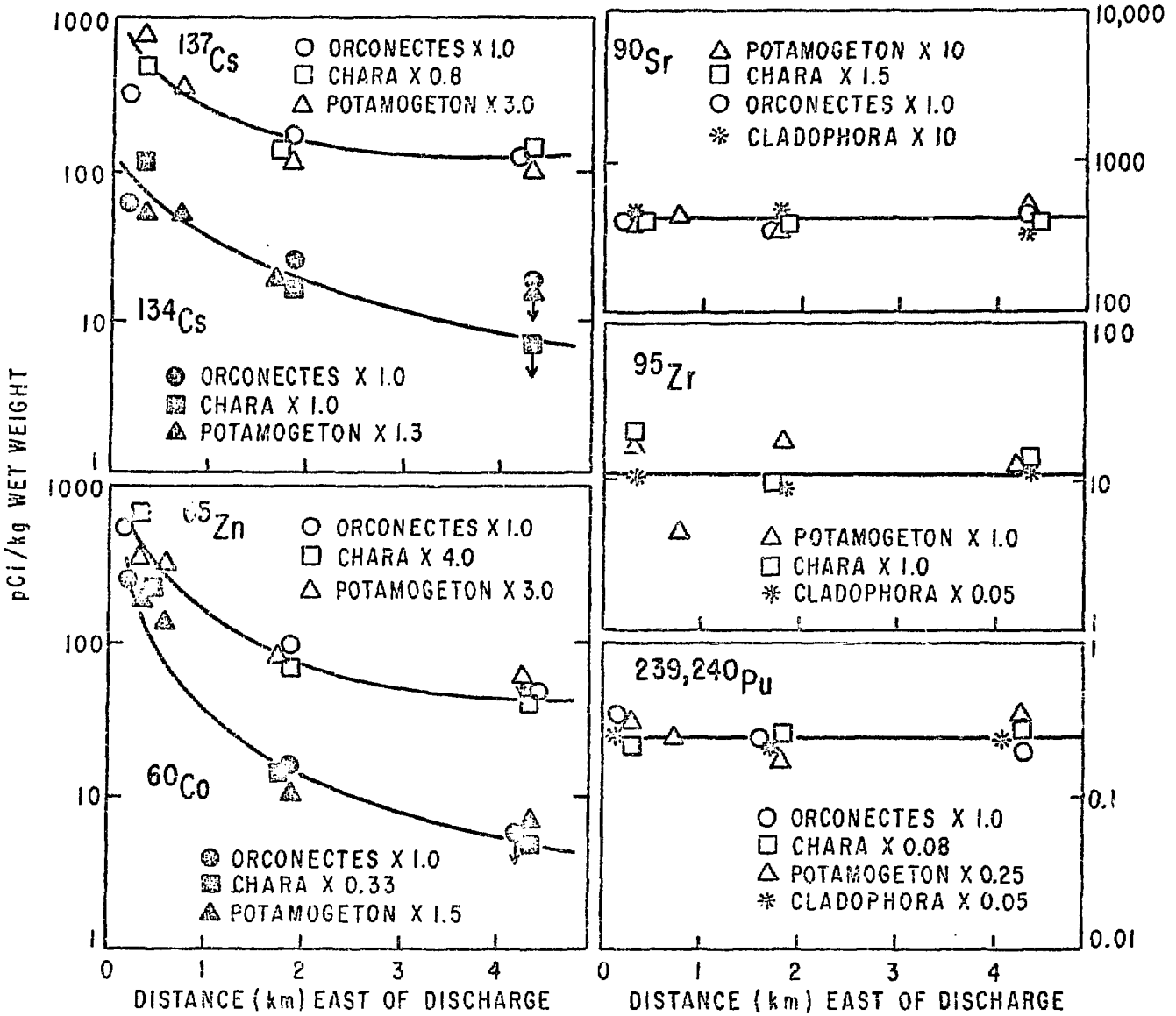
TABLE 2. Radionuclide concentrations in Big Rock waste releases

Sample No.	Date of Release	Concentration, pCi/ℓ [a]						
		$^{240,239}Pu$	^{238}Pu	^{137}Cs	^{134}Cs	^{90}Sr	^{65}Zn	^{60}Co
48 - 70	19 Oct 70	17.4	7.2	730 000	273 000	--	260 000	43 000
20 - 71	14 Jun 71	83.3	36.6	556 000	255 000	--	1 010 000	357 000
22 - 74	26 Nov 74	20.5	9.4	512 000	139 000	--	18 800	73 000
24 - 74	28 Dec 74	70.5	34.3	477 000	144 000	--	165 000	519 000
Mean concentration in waste solutions		46	22	570 000	200 000	~10 000 [b]	365 000	250 000
Mean concentration (annual basis) diluted in plant cooling water		0.0002	0.00010	2.7	1	~0.05	1.8	1.2
Mean lake concentration (fallout background)		.0007	0.00004	.050	<.001	0.8	<0.010	<0.010

(a) The 1 σ counting error is ~5%.

(b) Axtell, 1974

FIGURE 1. Normalized concentrations of radionuclides in inshore biota at various distances from the Big Rock Nuclear Plant discharge. (An arrow ↓ indicates that the value is at the limit of detectability for that sample.)


pCi/kg WET WEIGHT
1000
100
10
1
137Cs
ORCONECTES X 1.0
CHARA X 0.8
POTAMOGETON X 3.0
134Cs
ORCONECTES X 1.0
CHARA X 1.0
POTAMOGETON X 1.3
65Zn
ORCONECTES X 1.0
CHARA X 4.0
POTAMOGETON X 3.0
60Co
ORCONECTES X 1.0
CHARA X 0.33
POTAMOGETON X 1.5
0
1
2
3
4
DISTANCE (km) EAST OF DISCHARGE
90Sr
POTAMOGETON X 10
CHARA X 1.5
ORCONECTES X 1.0
CLADOPHORA X 10
10,000
1000
100
95Zr
POTAMOGETON X 1.0
CHARA X 1.0
CLADOPHORA X 0.05
100
10
1
239,240Pu
ORCONECTES X 1.0
CHARA X 0.08
POTAMOGETON X 0.25
CLADOPHORA X 0.05
1
0.1
0.01
0
1
2
3
4
DISTANCE (km) EAST OF DISCHARGE