

MASTER

Report COO-3568-18

PLUTONIUM AND AMERICIUM CONCENTRATION ALONG FRESH-WATER
FOOD CHAINS OF THE GREAT LAKES, U. S. A.

Progress Report
for Period July, 1976—September 30, 1977

Vaughan T. Bowen

Woods Hole Oceanographic Institution
Woods Hole, Massachusetts 02543

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

Prepared for

THE U. S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
UNDER CONTRACT NO. E(11-1)-3568

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Abstract

The primary purpose of studying the biogeochemical behavior of transuranic radionuclides in large freshwater lakes began with studies of the distribution of these radionuclides, essentially introduced from the fallout of atmospheric nuclear weapons tests, in Lake Ontario. The recognition that an additional source of supply of these nuclides to Lake Ontario (subsequent to their release into Lake Erie) existed in the form of leakage from the Nuclear Fuel Services reprocessing plant at Springville, New York, led to expansion of the program to include sampling at the eastern end of Lake Erie. Much of the program is devoted to studies of the distribution of these nuclides in the lake sediments as they appear to be a major repository for transuranics in freshwater lakes. The extent to which this is a temporary or permanent repository is illuminated by studies of transuranic distributions in the lake waters and biota.

Bibliographic Summary

1. The following report has been published but no reprints have been provided. Six Xerox copies are enclosed.

C00-3568-16: Heit, M. and J. C. Burke, 1976. Sediment sampling in six western lakes. U. S. ERDA, HASL Environmental Quarterly, HASL-315, pp. I-83 - I-91.

2. Six copies of the following reports are enclosed. These were published since our last Progress Report but no reprints have been provided.

C00-3568-12; C00-3563-33: Livingston, H. D. and V. T. Bowen, 1977. Contrasts between the marine and freshwater biological interaction of plutonium and americium. U. S. ERDA, HASL Environmental Quarterly, HASL-315, pp. I-157 - I-174.

C00-3568-17: Volchok, H., 1976. Plutonium-238 in sediment cores from the Mound Laboratory area. U. S. ERDA, HASL Environmental Quarterly, HASL-308, pp. I-21 - I-53.

3. The following report, of which six preprints are enclosed, was presented at a meeting and will be published.

C00-3568-15; C00-3563-51: Bowen, V. T. Natural matrix standards. Presented to Working Group on Low-Level Measurement Techniques, of the International Committee for Radionuclide Metrology, Paris, October 1976.

4. Still in preparation or in process of publication are the following:

COO-3568-6: Farmer, F. G., V. T. Bowen and V. E. Noshkin. Long-lived artificial radionuclides in Lake Ontario. I. Supply from fallout, and concentrations in lake water, of plutonium, americium, strontium 90 and cesium 137. This has been submitted to Limn. & Oceanogr. and was included with our 1975-76 Progress Report.

Farmer, J. G., V. T. Bowen and V. E. Noshkin. Long-lived artificial radionuclides in Lake Ontario. II. Plutonium, americium, strontium 90 and cesium 137 in sediment cores.

Farmer, J. G. Pb-210 dating of Lake Ontario sediments.

Farmer, J. G. Distributions of Pb-210 and stable lead in lake Ontario sediments.

Farmer, J. G. Distributions of trace elements in Lake Ontario sediments.

Farmer, J. G., V. T. Bowen and V. E. Noshkin. Pb-210 dating and fallout radionuclide profiles in Lake Ontario sediments.

Collecting Program

Two cruises were made this year to collect samples for this program.

A. In June we sampled in Lake Erie on the R/V C.A. DAMBACH of New York State University College at Buffalo. In Table 1 are listed the various station locations and the type of sample collected. The sampling pattern was chosen to provide samples which would reflect the input to Lake Erie of effluent radio-nuclides coming out of Cattaraugus Creek from the Nuclear Fuel Services reprocessing plant at West Valley, New York. The bottom in the immediate area close to the input point from Cattaraugus Creek is largely sandy and difficult to penetrate with our coring device. However, we were successful in taking two good cores at places where pockets of fine grained sediments were found.

B. We returned to Buffalo in August and, again on R/V C. A. DAMBACH, sampled on Lake Ontario. The station locations and type of sample collected are listed in Table 2. At the stations where water samples were collected, we also tried out the MnO_2 impregnated filter cartridge method of extracting transuranics from the water. One hundred gallons of water were pumped through each cartridge to concentrate the transuranics. The samples collected using near to the outflows of the Ginna and Nine Mile Point nuclear power complexes, will be of especial interest.

Performance of Modified 21-cm Corer

In the past year we have modified our 21-cm corer to improve its coring characteristics. By adding a tripod assembly and increasing the weight capacity, its penetration ability and resistance to falling over have been improved considerably. We tried the modified device in both Lake Erie and Lake Ontario. In Lake Erie some problems were encountered due partly to the hardness of the sediments encountered and partly to the frequent parting of the closure valve wire in these coarse sediments. In Lake Ontario, with a stronger wire, the performance of the device was truly superb and, combined with some very careful coring techniques by our sampling team, we obtained a suite of cores which are the finest we have ever collected in the lake. The quality of the core tops was excellent; flat, undisturbed, and under clear water in the core barrell (containing Amphipods and Euphasids). Some modifications and improvements made to the techniques used for core extrusion worked very well, and we have every expectation that the data from this set of samples should be most valuable in elucidating the behavior of the transuranics in the lake sediments.

Analytical Program

Our analytical methods are in general unchanged from those described in last year's Progress Report. New or modified developments have been made in respect of ^{134}Cs , ^{55}Fe analysis, and of direct measurement of γ -emitting radionuclides using our new germanium diode spectrometer.

We continue to place a strong emphasis on regular and careful quality control of our analytical data. In addition to our program

of blanks and quality control samples, we participate wherever possible in intercalibration exercises organized between laboratories, by national and international agencies. We recently summarized our performance in recent years in analysis of biological and sediment matrices and have included these results in Tables 3A and 3B. The general good comparison between our results and the "best", "preferred", or "expected" values confirms our belief that we are continuing to turn out reliable data of very high quality.

Our new germanium diode has been put to good use in several ways in various of our programs. In relation to this program, two applications have been made with very good results. Firstly, the high resolution of the germanium diode combined with the much higher detection efficiency of our new detector as opposed to our old model, has meant that we have the capability of measuring ^{134}Cs in radiochemically purified Cs fractions when it is present in detectable quantities. In samples from Great Lakes, this has been the case so far in Lake Erie sediments containing radionuclides from the Nuclear Fuel Services reprocessing operation in West Valley, New York. We expect to detect ^{134}Cs in locations influenced by local releases from nuclear power plants situated along the lake shores, but have not as yet looked at such samples with this in mind.

Secondly, we have modified the Marinelli beaker used by HASL to hold samples for direct germanium detector γ -spectrometry. The purpose of the modifications was to reduce the beaker capacity to permit high efficiency counting of dry sediment samples prior

to radiochemical analysis. It would be unusual for us to have sufficient sample to fill the 600 ml capacity of these beakers. However, using a specially designed aluminum plug, the internal diameter of the beaker was reduced by filling the outside annulus with epoxy resin. The reduced volume of about 300 ml was sufficient to contain 220 g of dry sediment in optimal counting geometry. Preliminary measurement of γ -emitting radionuclides, principally ^{137}Cs , has permitted us to reduce the number of samples analyzed radiochemically by use of this "pre-scan" technique. We have found that data for ^{137}Cs measured in this way has been in excellent agreement with the measurement of ^{137}Cs by β -detection following radiochemical separation.

We have finally eliminated the problems which persistently plagued our ^{55}Fe detectors. We discovered a gating defect in our analyzer had been causing pulse loss and a consequent underestimate of sample radioactivity at count rates approaching detector background. After many headaches and much frustration this problem was finally identified and solved, leaving us presently with two detectors operating very reliably. This means that previously reported data for ^{55}Fe in our core Ontario 73-6 is erroneously low by about a factor of two. Some new ^{55}Fe data obtained with this system are included in this report and we plan to make additional measurements next year to examine the comparative geochemistry of ^{55}Fe and $^{239,240}\text{Pu}$ in greater detail.

Data

In Tables 4-7 are shown new data for cores collected on the R/V OCEANUS cruise in November 1975. They include OCEANUS 1 cores #2, #4, #6, #9. Tables 8-10 contain the data on hand for

some of the sediment samples collected in Lake Erie in 1976 -- E-76-3, E-76-7, E-76-14. Tables 11-15 have new data for Lake Ontario sediments in 1976 -- 0-76-1, 0-76-3, 0-76-6, 0-76-15. In table 16 are shown data for the 1976 plankton samples for Lake Ontario. Table 17 contains ⁵⁵Fe data for sediments in Lake Erie, Lake Ontario and the Gulf of St. Lawrence -- subsequent to our ⁵⁵Fe detection system being stabilized and finally calibrated.

Discussion

In Tables 8-15, are presented the radionuclide data measured in various grab samples and cores collected in Lake Erie and Lake Ontario in 1976. E-76-3, E-76-7 and E-76-14 are relevant to the questions of the extent to which leakage from the Nuclear Fuels Service reprocessing plant at West Valley, N. Y. is detectable in Lake Erie. 0-76-3 and 0-76-6 from the Niagara Delta region may be relevant to this question and also add to our suite of samples from this area which give a useful sedimentary record of the delivery of radionuclides to this location from both direct delivery and that from the upper lakes.

Of the Lake Erie samples, the E-76-7 grab sample seems to show the greatest influence of radionuclides from the West Valley source. This is especially indicated by the very high ²³⁸Pu/^{239,240}Pu ratios and the detection of ²⁴⁴Cm and ¹³⁴Cs -- both not to be found in lake sediments containing radionuclides solely from fallout. It is unfortunate that we did not have a long core at this location as the data suggest a rather rapid sedimentation rate. Both Pu and Cs radionuclide concentrations

are still increasing in the 8-10 cm section and it is tempting to speculate that we are seeing the record of delivery from Cattaraugus Creek -- the point of entry to Lake Erie of the West Valley releases -- and that much of the fallout record lies deeper with the sediment at this location. Hopefully we can examine this question in more detail with the 1977 samples from this area. E-76-3 shows much less evidence of contamination from the West Valley source. The concentrations of $^{239,240}\text{Pu}$ and ^{137}Cs are lower than E-76-3 and ^{134}Cs was undetectable. Yet the $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios of 0.09-0.012 in the top 5 cm suggest that some material from West Valley must be mixed in with that arising from fallout. The nuclide concentration profiles decreasing from the surface are suggestive of an area of low sedimentation reworked by organisms. The location (on the westward side of a sandbar which separates it from the area which is fed directly by Cattaraugus Creek) is essentially "upstream" from the point of entry of the West Valley releases and hence the reduced evidence of their presence is not surprising. E-76-14 likewise shows little evidence of West Valley radionuclides. Although ^{134}Cs and Am/Cm analyses are not yet completed, the nuclide profiles and $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio record seem more consistent with that to be found in sediments containing mostly radionuclides from fallout. The location of this grab, being relatively far to the north and slightly west of the Cattaraugus Creek input would be unlikely to be much influenced by this source if the releases are swept to the northeast along the lakeshore rather soon after entering the lake.

In the samples from the Niagara River delta region, only data for Pu and ^{137}Cs are presently at hand. The low concentrations of both nuclides in the 0-76-3 grab are consistent with the coarse texture of this sample. Although this sample was closer to the point of entry of the Niagara to the Lake, the rate of flow must still be sufficiently high as to prevent accumulation of fine grained sediments. 0-76-6 being further to the north was clearly in the area of fine grained sediment accumulation and the radio-nuclide profile here was similar to that obtained in this area in previous years with a sub-surface maximum of each nuclide concentration profile and increased $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios in the sections near to the surface. The position of these maxima, 4-5 cm for Pu and 2-3 cm indicate either that the sedimentation rate at this point is not as great as in 0-73-6 reported in previous years' Progress Reports (where the maxima were deeper buried in the sediment column) or that both Pu and Cs are being rather rapidly returned toward the water sediment interface by upward diffusion mechanisms.

The 0-76-1 grab sediment was collected to see whether any effluents could be detected in this sediment which originated from the Ginna nuclear reactor nearby. Neither the concentrations of Pu and ^{137}Cs detected, nor the ratio $^{238}\text{Pu}/^{239,240}\text{Pu}$, were indicative of significant contributions from low level discharges from the reactor.

The data from 0-76-10 and 0-76-15 will be useful additions to our expanding body of data for sediments collected at the eastern end of Lake Ontario. The data from the core (0-76-10) in the deepest part of the lake continues to show the deficiency

of ^{137}Cs relative to $^{239,240}\text{Pu}$ which was discussed last year in respect of cores 71-5, 73-13A and 73-13B.

As mentioned in the Analytical Program section, we have made some ^{55}Fe measurements in some Lake Erie, Lake Ontario, and Gulf of Lt. Lawrence sediments since the problems remaining with our detector systems were finally straightened out. Both samples of bulked lake sediment analyzed yielded data (Table 17) which showed the sediments to be characterized by $^{55}\text{Fe}/^{239,240}\text{Pu}$ ratios which are not significantly different from the value of 12 (as of 1/1/1975) which we believe is representative of cumulative fallout by that date. We would take this to mean that little fractionation between ^{55}Fe and Pu takes place during delivery to the lake sediments or post-depositionally. We hope to examine this proposition in greater detail through analyses of ^{55}Fe and Pu in a larger number of lake cores and their ratio variation with depth. In the Gulf of St. Lawrence sediments analyzed for ^{55}Fe , relatively higher amounts of ^{55}Fe relative to $^{239,240}\text{Pu}$ were found, ratios ranging from 24-43. The explanation we would at present favor would be one of a deficiency of $^{239,240}\text{Pu}$ rather than an excess of ^{55}Fe . The inventories of Pu at both locations (from sediment core analyses) reveals considerably less Pu than would be expected in shallow estuarine sediments. The excess ^{55}Fe is too great, and the $^{55}\text{Fe}/^{239,240}\text{Pu}$ ratio in the lake sediments too similar to fallout, to permit an explanation based on differential input of ^{55}Fe from the St. Lawrence River.

This year our concentration on sediment analyses has resulted in few additional analyses of biota from the lakes. However, the two plankton samples analyzed (Table 16) are worthy of comment. The high $^{239,240}\text{Pu}$ concentration in the sample taken

at the 0-76-7 station just north of the Niagara River input and the high $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio would seem to reflect the Lake Erie input containing some nuclides from the Nuclear Fuel Services source. On the other hand, the even higher ratio observed in the sample from the East end of the Lake lacks a good explanation, and we are presently re-analyzing this sample to confirm that the value obtained was not spurious.

TABLE 1

Lake Erie -- 1977 Stations

<u>Station</u>	<u>Location</u>	<u>Type of Sample</u>			
		<u>Core</u>	<u>Grab</u>	<u>Water</u>	<u>Plankton</u>
1	42°36.9'N; 79°08.6'W		✓		
2	42°35.8'N; 79°09.2'W	✓		✓	
3	42°36'N; 79°09'W				✓
4	42°34.3'N; 79°09.3'W			✓	
5	42°33.5'N; 79°10.3'W			✓	
6	42°33.5'N; 79°10.3'W	--	--	--	--
7	42°37.8'N; 79°17'W	Tripod & Old Style		✓	
8	42°39.8'N; 79°12.7'W	✓		✓	
9	42°38'N; 79°10.8'W			✓	
10	42°39'N; 79°10'W				✓

TABLE 2
Lake Ontario -- 1977 Stations

<u>Station Number</u>	<u>Location</u>	<u>Core Sample</u>	<u>Water Sample</u>	<u>Plankton Number</u>
1	43° 21.4'N; 79° 37.5'W	1		
2	43° 25'N; 79° 25'W		1	
2-A				1
3	43° 20'N; 79° 08.5'W	2		
4	43° 21.3'N; 79° 06'W	3		
5	43° 22.5'N; 79° 04'W	4		
6	43° 33'N; 78° 00'W	5	2	
6-A				2
7	43° 17.5'N; 77° 18.6'W		3	
8	43° 23.6'N; 77° 00'W	6		
9	43° 30.2'N; 76° 57.5'W	7	4	
10	43° 31.6'N; 76° 41'W	8		
10-A				3
11	43° 37.2'N; 76° 32'W	9		3
12	43° 32'N; 76° 23'W		5	

	pCi/g					pCi/Kg				
	<u>55</u> _{Fe}	<u>90</u> _{Sr}	<u>134</u> _{Cs}	<u>137</u> _{Cs}	<u>238</u> _{Pu}	<u>239,240</u> _{Pu}	<u>241</u> _{Pu}	<u>241</u> _{Am}	<u>242</u> _{Cm}	<u>244</u> _{Cm}
IAEA SD-B-1										
Reference date: 1/1/73										
WHOI values		14.7±0.5	10.1±1.4	424±12	60±5	890±30	4890±490	231±18	0.11±0.08	0.09±0.05
IAEA values		15.0±1.4	9.6±0.3	361±10	42±4	960±30	---	---	---	---
NBS River Sediment SRM 4350										
Reference date: 1/1/75										
WHOI values	29.2±0.1(1)			2.36±0.08	1.6 ±0.06	39.2±0.3		8.6 ±0.7	<0.05	0.41±0.05
NBS values				2.70±0.12	2.0 ±0.1	37.8±3.2		---	---	---
HASL values				2.42±0.06	---	37.8±1.3				
HSL values				2.54±0.05	---			8.4±0.7		<0.2
Miettinen	43 ± ?				2.1 ±0.2	35.1±0.7				
NBS River Sediment RM 45										
Aliquot 33-WHOI	28.9±0.3			2.31±0.01	1.02±0.09	29.0±0.8		8.06±0.3		0.28±0.05
	<u>55</u> _{Fe}	<u>60</u> _{Co}	<u>134</u> _{Cs}	<u>137</u> _{Cs}	<u>238</u> _{Pu}	<u>239,240</u> _{Pu}	<u>241</u> _{Pu}	<u>241</u> _{Am}	<u>242</u> _{Cm}	<u>244</u> _{Cm}
HUDSON RIVER SEDIMENT	SLOSH III	#1000 Q-1								
Reference date	11 June 1975									
WHOI values	1 }	0.462± 0.022	0.483±0.038 0.445±0.062	2.72±0.03 2.72±0.05	1.08±0.23 0.79±0.14	25.3±1.4 24.5±1.0				
Lamont values for MP43 -from Simpson, et al. Science 194, 179 (1976)		0.400±0.07	0.345±0.042	2.70±0.72	---	25.0±1.6				

TABLE 3 B

16

BIOLOGICAL MATERIALS -- INTERCOMPARISON ANALYSES

19 May 1977

	^{90}Sr	^{134}Cs	^{137}Cs	^{238}Pu	$^{239,240}\text{Pu}$	^{241}Pu	^{241}Am	^{242}Cm	^{244}Cm									
	pCi/g									pCi/Kg								
<u>IAEA AG-I-1 (<i>Fucus serratus</i>)</u>																		
Reference date: 1/1/72																		
WHOI values	10.0 ± 0.3	10.5 ± 0.2	78.2 ± 0.3	4.8×10^3 $\pm 0.2 \times 10^3$	30.0×10^3 $\pm 0.6 \times 10^3$	800×10^3 $\pm 30 \times 10^3$	5.2×10^3 $\pm 0.2 \times 10^3$	0.08×10^3 $\pm 0.01 \times 10^3$										
IAEA values	10.0 ± 0.2	10.0 ± 0.2	$75. \pm 1$	3.8×10^3 $\pm 0.1 \times 10^3$	27.0×10^3 $\pm 0.5 \times 10^3$													
Other laboratory values								4.4×10^3 $\pm 0.1 \times 10^3$	0.03×10^3 $\pm 0.1 \times 10^3$									

IAEA MA-B-1 (*Anadara granosa*) -- clam

Reference date: 1/1/75

WHOI values	8.0 ± 0.4	17.7 ± 0.1	1.41 ± 0.25	38.3 ± 1.4	18.6 ± 2.5	
IAEA values	7.4 ± 0.8	16.2 ± 0.4	1.5 ± 0.1	$44. \pm 4$		
Other laboratory values					$8-35$	

IAEA MA-B-2 (*Aplysia benedicti*) sea hare

Reference date: 1/1/75

WHOI values		5.56 ± 0.02	0.76 ± 0.12	1.05 ± 0.17	0.9 ± 0.4	
IAEA values		4.2 ± 0.2	0.86 ± 0.07	1.1 ± 0.5	---	

TABLE 4

 RADIOCHEMICAL ANALYSES OF LAKE ONTARIO SEDIMENT CORE
 RADIONUCLIDES IN DISINTEGRATIONS PER MINUTE PER KG DRY SEDIMENT

Oceanus I, Station #2 Core #2

Core Collection Date: 10/30/75

Location: 42°30.5'N, 79°53.3'W

Water Depth: 63.5m

Core Cross-Sectional Area: 206.6 cm²

Depth in Sediment (cm)	Wet/Dry Weight	¹³⁷ Cs	²³⁹ Pu	²³⁸ Pu/ ²³⁹ Pu	Pu/Cs	²⁴¹ Am
0-1	3.41	8264±24	107.7±3.0	0.07±.009	0.013	
1-2						
2-3	3.37	11056±27	156.6±8.2	0.074±.008	0.014	
3-4						
4-5	3.23	15112±33	180.0±8.6	0.056±.006	0.012	
5-6						
6-7	3.12	20064±37	284.6±11.3	0.029±.003	0.014	
7-8						
8-9	3.07	21924±36	307.9±18.8	0.036±.005	0.014	
9-10						
10-12	2.79	11980±23	227±5.6		0.019	
12-14	3.04	11655±26	242.1±11.0	0.046±.005	0.021	
14-16						
16-18	2.93	7545±22	173.1±4.4	0.033±.002	0.023	
18-20						
20-22	2.67	1668±8.3	36.6±1.1	0.029±.004	0.022	
22-24						
24-26	2.53	255±5	5.45±0.35	0.027±.018		
26-28						
28-30	2.53	69.6±3.5	0.79±0.12	0.04±.04		
30-32						
32-34						

TABLE 5

RADIOCHEMICAL ANALYSES OF LAKE ONTARIO SEDIMENT CORE

RADIONUCLIDES IN DISINTEGRATIONS PER MINUTE PER KG DRY SEDIMENT

OCEANUS I, Station #3 Core #4

Core Collection Date: 10/31/75

Location: $42^{\circ}43'N$, $79^{\circ}48.5'W$

Water Depth: 25m

Core Cross-Sectional Area:

Depth in Sediment (cm)	Wet/Dry Weight	^{137}Cs	^{239}Pu	$^{238}Pu/^{239}Pu$	Pu/Cs	^{241}Am
0-1	3.32	4934 ± 10	67.8 ± 1.9	$0.055 \pm .004$	0.0137	
1-2						
2-3	2.55	4842 ± 9	82.5 ± 2.2	$0.054 \pm .004$	0.0170	
3-4						
4-5	2.40	2422 ± 7	42.9 ± 1.3	$0.042 \pm .004$	0.0177	
5-6						
6-7	2.38		$8.62 \pm .45$	$0.032 \pm .008$		
7-8						
8-10						
10-12	2.09	14.15 ± 3.00	$0.53 \pm .07$			
12-14						
14-16						
16-18	1.86	-4.4 ± 3.5	$0.27 \pm .05$			

TABLE 6
 RADIOCHEMICAL ANALYSES OF LAKE ONTARIO SEDIMENT CORE
 RADIONUCLIDES IN DISINTEGRATIONS PER MINUTE PER KG DRY SEDIMENT

OCEANUS I, Station #4 Core #6

Core Collection Date: 10/31/75

Location: $43^{\circ}20.5'N$, $79^{\circ}04'W$

Water Depth: 98m

Core Cross-Sectional Area: 263 cm^2

Depth in Sediment (cm)	Wet/Dry Weight	^{137}Cs	^{239}Pu	$^{238}\text{Pu}/^{239}\text{Pu}$	Pu/Cs	^{241}Am
0-1	4.40	20373 ± 42	398.1 ± 14.0	$0.034 \pm .004$	0.0195	
1-2						
2-3	4.35	15399 ± 32	315.7 ± 10.6	$0.032 \pm .003$	0.0205	
3-4						
4-5	4.02	11777 ± 26.5	224.4 ± 7.8	$0.030 \pm .003$	0.019	
5-6						
6-7	3.97	7161 ± 23	145.8 ± 5.2	$0.032 \pm .004$	0.0204	
7-8						
8-9	3.75	3338 ± 11	53.4 ± 2.9 54.3 ± 1.4	$0.019 \pm .006$	0.016	
9-10						
10-12	3.49	1908 ± 7.3	51.2 ± 2.3	$0.03 \pm .0055$	0.027	
12-14						
14-16	2.91	24.2 ± 3.5	0.82 ± 0.25		0.034	
16-18						
18-20	2.93	1.8 ± 6.1				

Total ^{137}Cs - 130.03 mCi/km^2

Total ^{239}Pu - 2.62 mCi/km^2

TABLE 7

 RADIOCHEMICAL ANALYSES OF LAKE ONTARIO SEDIMENT CORE
 RADIONUCLIDES IN DISINTEGRATIONS PER MINUTE PER KG DRY SEDIMENT

OCENUS I, Station #7 Core #9

Core Collection Date: 11/1/75

Location: $43^{\circ}29'N$, $76^{\circ}58.5'W$

Water Depth: 241m

Core Cross-Sectional Area: 206.6 cm^2

Depth in Sediment (cm)	Wet/Dry Weight	^{137}Cs	^{239}Pu	$^{238}Pu/^{239}Pu$	Pu/Cs	^{241}Am
0-1	6.62	25515 ± 62	779.9 ± 16.4	$0.032 \pm .001$	0.0306	
1-2						
2-3	6.00	9275 ± 31	361.5 ± 7.9	$0.023 \pm .001$	0.0390	
3-4						
4-5	5.28	3869 ± 19	134.4 ± 4.3	$0.029 \pm .003$	0.0347	
5-6						
6-7	5.03	2495 ± 15	66.0 ± 2.5	$0.030 \pm .005$	0.0265	
7-8						
8-9	4.58	1273 ± 8	34.2 ± 1.8	$0.025 \pm .007$	0.0269	
9-10						
10-12						
12-14	3.77	493 ± 5.6	$11.8 \pm .51$	$0.059 \pm .002$	0.024	
14-16						
16-18						
18-20	4.36	147 ± 5.5	2.8 ± 0.4	$0.32 \pm .025$		
20-22						
22-24						
24-26						
26-28						
28-30						
30-32						

Total ^{239}Pu $1.918 \text{ mCi}/\text{km}^2$ Total ^{137}Cs $64 \text{ mCi}/\text{km}^2$

TABLE 8
RADIOCHEMICAL ANALYSES OF LAKE ERIE SEDIMENT GRABS
RADIONUCLIDES IN DISINTEGRATIONS PER MINUTE PER KG DRY SEDIMENT

Station: E-76-3 Grab
 Core Collection Date: 8/24/76
 Location: 42°33'30"N; 79°10'20"W; 3/4 mi. off-shore, to mi. east of
 Water Depth: 11 m Silver Creek
 Core Cross-Sectional Area:

Depth in Sediment (cm)	Wet/Dry Weight	<u>137</u> _{Cs} *	<u>239</u> _{Pu}	<u>238</u> _{Pu} / <u>239</u> _{Pu}	<u>239</u> _{Pu} / <u>137</u> _{Cs}	<u>241</u> _{Am}
0-1	1.65	1200±25	11.3±0.4	0.09±0.01	0.0095	
1-3	1.58	453±23	3.8±0.2	0.11±0.02	0.0094	
3-5	1.54	159±18	1.6±0.1	0.12±0.03	0.0110	
5-7	1.49	128±17	1.1±0.1	0.20±0.04	0.0100	

TABLE 9

Station: E-76-14
 Core Collection Date: 8/25/76
 Location: 42°38'50"N; 79°12'30"W; 7 mi. from Silver Creek, 5½ mi. from
 Water Depth: 23 m Lotus Point
 Core Cross-Sectional Area:

Depth in Sediment (cm)	Wet/Dry Weight	<u>137</u> _{Cs} *	<u>239</u> _{Pu}	<u>238</u> _{Pu} / <u>239</u> _{Pu}	<u>239</u> _{Pu} / <u>137</u> _{Cs}	<u>241</u> _{Am}
0-2	2.34	4202±16	64±2	0.070±0.006	0.015	
2-4	2.13	7246±20	148±4	0.046±0.003	0.020	
4-6	1.76	4100±13	104±3	0.031±0.003	0.025	
6-8	1.52	755±3	27±1	0.022±0.004	0.036	
8-10	1.44	140±2	5±0.3	0.017±0.046	0.037	

* ¹³⁴Cs values all less than 20.

TABLE 10
RADIOCHEMICAL ANALYSES OF LAKE ERIE SEDIMENT GRABS
RADIONUCLIDES IN DISINTEGRATIONS PER MINUTE PER KG DRY SEDIMENT

Station: E-76-7

Core Collection Date: 24 Aug 76

Location: $42^{\circ}35'25''N$; $79^{\circ}09'00''W$; 3 miles off Silver Creek, 1 mile off Lotus Point

Water Depth: 15 m

Core Cross-Sectional Area:

Depth in Sediment (cm)	Wet/Dry Weight	$^{239}_{\text{Pu}}$	$^{241}_{\text{Am}}$ ($^{244}_{\text{Cm}}\right)^*$	$^{238}_{\text{Pu}}/^{239}_{\text{Pu}}$	$^{239}_{\text{Pu}}/^{137}_{\text{Cs}}$	$^{241}_{\text{Am}}/^{239}_{\text{Pu}}$	$^{137}_{\text{Cs}}$	$^{134}_{\text{Cs}}$	$^{134}_{\text{Cs}}/^{137}_{\text{Cs}}$
0-2	1.58	10.6 ± 0.3	3.4 ± 0.3 (0.18 ± 0.08)	0.17 ± 0.01	0.0057	0.32 ± 0.03	1890 ± 40	114 ± 29	0.061 ± 0.016
2-4	1.90	29.9 ± 0.7	7.9 ± 0.5 (0.46 ± 0.12)	0.16 ± 0.01	0.0071	0.27 ± 0.02	4390 ± 50	118 ± 37	0.027 ± 0.008
4-6	1.59	26.6 ± 0.7	11.1 ± 0.6 (0.96 ± 0.14)	0.32 ± 0.01	0.0043	0.42 ± 0.03	6200 ± 40	203 ± 38	0.033 ± 0.006
6-8	1.62	25.3 ± 0.6	11.8 ± 0.6 (1.10 ± 0.15)	0.40 ± 0.02	0.0035	0.47 ± 0.03	7200 ± 36	257 ± 28	0.036 ± 0.004
8-10	1.54	32.9 ± 0.8		0.38 ± 0.02	0.0033		9976 ± 19	-	-

* $^{244}_{\text{Cm}}$ values are given in parenthesis.

TABLE 11
 RADIOCHEMICAL ANALYSES OF LAKE ONTARIO GRABS AND CORES
 RADIONUCLIDES IN DISINTEGRATIONS PER MINUTE PER KG DRY SEDIMENT

Station: 0-76-1 Grab
 Core Collection Date: 9 August 76
 Location: $43^{\circ}17'40"N$; $77^{\circ}18'40"W$; 1 mile off shore, 1/4 mile east of
 Ginna Nuclear Power Plant
 Water Depth: 20 m

Core Cross-Sectional Area:

Depth in Sediment (cm)	Wet/Dry Weight	^{137}Cs	^{239}Pu	^{241}Am	$^{238}\text{Pu}/^{239}\text{Pu}$	$^{239}\text{Pu}/^{137}\text{Cs}$
0-4	1.49	500 ± 2	7.5 ± 0.3		0.06 ± 0.01	0.015

TABLE 12

Station: 0-76-3 Grab
 Core Collection Date: 11 August 76
 Location: $43^{\circ}16'40"N$; $79^{\circ}04'20"W$; 1 mile from Fort Niagara
 Water Depth: 9 m
 Core Cross-Sectional Area:

Depth in Sediment (cm)	Wet/Dry Weight	^{137}Cs	^{239}Pu	^{241}Am	$^{238}\text{Pu}/^{239}\text{Pu}$	$^{239}\text{Pu}/^{137}\text{Cs}$
0-2	1.30	183 ± 2	3.7 ± 0.2		0.04 ± 0.01	0.020
2-4	1.22	203 ± 3	2.4 ± 0.2		0.08 ± 0.02	0.012
4-6	1.21	129 ± 5	3.1 ± 0.2		0.05 ± 0.01	0.024

TABLE 13

Station: 0-76-15 Grab
 Core Collection Date: 14 August 76
 Location: $44^{\circ}05'10"N$; $76^{\circ}23'00"W$; 1 mile off shore; 1/2 mile from
 (B245) Buoy.
 Water Depth: 21 m
 Core Cross-Sectional Area:

Depth in Sediment (cm)	Wet/Dry Weight	^{137}Cs	^{239}Pu	^{241}Am	$^{238}\text{Pu}/^{239}\text{Pu}$	$^{239}\text{Pu}/^{137}\text{Cs}$
0-1	2.85	10206 ± 14	157 ± 4		0.044 ± 0.002	0.015
1-2	2.29	2598 ± 7	83 ± 2		0.041 ± 0.003	0.032

TABLE 14

RADIOCHEMICAL ANALYSES OF LAKE ONTARIO GRABS AND CORES
RADIONUCLIDES IN DISINTEGRATIONS PER MINUTE PER KG DRY SEDIMENT

Station: 0-76-6 Core
 Core Collection Date: 12 August 1976
 Location: $43^{\circ}20'50''N$: $79^{\circ}03'45''W$; 2 miles out from R₂ Buoy
 Water Depth: 75 m
 Core Cross-Sectional Area:

Depth in Sediment (cm)	Wet/Dry Weight	^{137}Cs	^{239}Pu	^{241}Am	$^{238}Pu/^{239}Pu$	$^{239}Pu/^{137}Cs$
Jerry Jug	-	8712 ± 20	117 ± 4		0.059 ± 0.005	0.0134
0-1	3.43	10602 ± 21	133 ± 3		0.080 ± 0.004	0.0125
2-3	3.34	18057 ± 20	242 ± 4		0.050 ± 0.002	0.0134
4-5	3.69	15672 ± 20	339 ± 5		0.032 ± 0.001	0.0216
6-7	3.27	9095 ± 16	219 ± 4		0.027 ± 0.001	0.0240
8-9	3.12	4927 ± 13	111 ± 2		0.025 ± 0.002	0.0225
10-11	2.50	943 ± 6	20 ± 1		0.030 ± 0.006	0.0213
12-13	2.40	91 ± 3	1.6 ± 0.2		0.010 ± 0.020	0.0180
14-15	2.20	9 ± 2	0.4 ± 0.1		b.d.l.*	0.04
16-17	2.20	13 ± 2	0.03 ± 0.02		b.d.l.	0.002

Inventory: $^{239}Pu = 3.55 \text{ mCi/Km}^2$; $^{137}Cs = 204 \text{ mCi/Km}^2$

* below detection level

TABLE 15

RADIOCHEMICAL ANALYSES OF LAKE ONTARIO GRABS AND CORES

RADIONUCLIDES IN DISINTEGRATIONS PER MINUTE PER KG DRY SEDIMENT

Station: 0-76-10 core
 Core Collection Date: 13 August 1976
 Location: $43^{\circ}30'00''N$; $76^{\circ}54'00''W$
 Water Depth: 220-225 m
 Core Cross-Sectional Area:

Depth in Sediment (cm)	Wet/Dry Weight	^{137}Cs	^{239}Pu	^{241}Am	$^{238}Pu/^{239}Pu$	$^{239}Pu/^{137}Cs$
Jerry Jug	-	20019 ± 31	378 ± 13		0.048 ± 0.004	0.019
0-1	6.28	21432 ± 34	622 ± 17		0.034 ± 0.002	0.029
2-3	4.57	1730 ± 14	55 ± 3		0.040 ± 0.006	0.030
4-5	4.30	957 ± 8	34 ± 1.3		0.029 ± 0.005	0.035
6-7	4.00	254 ± 6	6 ± 0.4		0.026 ± 0.008	0.024
8-9	4.06	37 ± 5	--		--	--
10-11	3.96	27 ± 10	--		--	--
12-13	3.81	6 ± 5	--		--	--

TABLE 16
RADIOCHEMICAL ANALYSES OF LAKE ONTARIO PLANKTON - 1976

	d.p.m./Kg wet weight			
	^{137}Cs	^{238}Pu	$^{239,240}\text{Pu}$	$^{238}\text{Pu}/^{239,240}\text{Pu}$
Location: 0-76-7, $43^{\circ}18'20''\text{N}$, $79^{\circ}03'00''\text{W}$				
Niagara delta area	57.5 ± 0.5	0.17 ± 0.02	1.04 ± 0.05	0.16 ± 0.02
Date Collected: 12 August 1976				
Location: 0-76-12, $43^{\circ}56'N$, $76^{\circ}30'W$				
East end of lake, between Galloo and Main Duck Islands	19.8 ± 0.3	0.11 ± 0.01	0.25 ± 0.02	0.44 ± 0.05

TABLE 17

 ^{55}Fe IN LAKE ERIE, LAKE ONTARIO AND GULF OF ST. LAWRENCE SEDIMENTS

<u>Sample</u>	^{55}Fe dpm/Kg (dry wt)	$^{55}\text{Fe}/^{239,240}\text{Pu}$
Oceanus-I, Station 1, Lake Erie $41^{\circ}52'N$, $81^{\circ}58'W$, 30 Oct. 1975 Bulk sediment from dredge	3400 ± 130	10.9
Oceanus-I, Station 4, Lake Ontario $43^{\circ}20.5'N$, $79^{\circ}04'W$, 31 Oct. 1975 Bulk sediment from dredge	4770 ± 140	13.4
Oceanus-I, Station 14, Gulf of St. Lawrence $48^{\circ}25'N$, $63^{\circ}45'W$, 17 Nov. 1975, ~132m Dredge sediment, 0-4 cm	3060 ± 70	24.1
Oceanus-I, Station 13, Gulf of St. Lawrence $49^{\circ}14.5'N$, $64^{\circ}57.8'W$, 16 Nov. 1975, 390 m 8" diameter core		
0-1 cm	2480 ± 110	31
2-3 cm	1120 ± 80	36
4-5 cm	470 ± 80	43
6-7 cm	350 ± 70	36