THE BEHAVIOR OF PLUTONIUM AND OTHER LONG-LIVED RADIONUCLIDES IN
LAKE MICHIGAN: II. PATTERNS OF DEPOSITION IN THE SEDIMENTS

by

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THE BEHAVIOR OF PLUTONIUM AND OTHER LONG-LIVED RADIONUCLIDES IN
LAKE MICHIGAN: II. PATTERNS OF DEPOSITION IN THE SEDIMENTS *

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At present eight operating nuclear power reactors, and one more which
will be in operation by 1980, are situated along the shores of Lake Michigan.
Since their releases of radioactivity have been much less than that entering
the lake from stratospheric fallout, $^{239,240}$Pu and $^{137}$Cs from the latter source
have been measured in order to attempt to predict the depositional patterns of
long-lived radionuclides in the lake.

Ninety-seven percent of the $^{239,240}$Pu and ninety-five percent of the
$^{137}$Cs that has entered the lake by wet and dry deposition of atmospheric
fallout now reside in the sediments. The vertical distribution of $^{137}$Cs has
been measured in 60 sediment cores, and $^{239,240}$Pu in 14 cores taken from all
over the lake. The sampling locations were chosen to be representative of the
various types of sedimentation viz., areas with high sedimentation associated
with river inputs, offshore locations where organic-rich, very fine-grained
material is settling out, and areas of low sedimentation where erosion may be
occurring.

*This work was performed under the auspices of the U.S. ERDA.
The results of these studies indicate that the scavenging of $^{239,240}\text{Pu}$ and $^{137}\text{Cs}$ appears to occur at the same overall rate but controlled by different processes in the water column. Analysis of the profiles shows that the sedimentation rate varies by more than an order of magnitude and that the total content of these two radionuclides is strongly correlated with the mass-sedimentation rate. As a result, there are large areas of the lake where there is virtually no deposition, and other areas where the total content of $^{239,240}\text{Pu}$ and $^{137}\text{Cs}$ per unit area is several times greater than would be expected. This depositional pattern is probably related to the hydrodynamic properties of the lake. The shape of the profiles in areas of high and low sedimentation also suggest that there is a redistribution of the radioactivity which has already been deposited in the sediments. In areas of high sedimentation, the concentration in the upper few centimeters of the core is much higher than can be explained in terms of new deposition or erosion from the watershed.

Therefore, any monitoring scheme for the long-term effects of releases from nuclear power plants must take account of the massive redistribution of sediment that may occur and must recognize that the final point of deposition may be far removed from the site of release.

1. INTRODUCTION

As one of the requirements for the granting of a construction permit or an operating license for a nuclear powered generating station in the United States, the applicant must plan and carry out environmental studies in relation to the operation of the plant. Since the sediments will be the ultimate sink for most of the long-lived radionuclides which may be discharged into the environment as a result of the operation of nuclear reactors, an effective monitoring scheme for this part of the ecosystem is of prime importance. This is particularly true for those plants that will be built on the shores of lakes or coastal enbayments where little or no loss of sediment will be associated with water exchange.

The Laurentian Great Lakes are one of the largest freshwater resources in the world, and a significant fraction of the total population of Canada and the United States live within their basins. As of May 1975, there were a total of seventeen operating nuclear power plants located in the Great Lakes Basin, which means that more than 30% of the total nuclear power capacity of North America was located in this region [1]. Of these plants, eight are located along the shores of Lake Michigan (Figure 1), which is the only one of the lakes totally contained within the continental United States. Lake Michigan is particularly vulnerable to the effects of the addition of undesirable pollutants because of the long residence time (> 100 years) for conservative materials in the water column [2]. There is an almost infinite residence time for pollutants that are readily transported to the sediments. This arises because Lake Michigan is virtually disconnected from the main flow pattern of the other lakes and there are no possible flushing mechanisms.

Since the releases of radioactivity from these operating nuclear plants, which have to be as low as feasible in order to meet the federal guidelines [3],
have been much lower than the amount entering the lake from stratospheric fallout, $^{239,240}\text{Pu}$ and $^{137}\text{Cs}$ from the latter source have been measured in order to attempt to predict the patterns of deposition of long-lived radionuclides in Lake Michigan. This information will be important in evaluating the suitability of additional sites for power plants, for the prediction of the effects of nonroutine releases, and for the implementation of meaningful routine surveys.

The intent of this paper is to summarize the results obtained from the analysis of a large number of sediment cores for $^{137}\text{Cs}$, and a lesser number for $^{239,240}\text{Pu}$, taken from Lake Michigan in 1972, 1973 and 1974. An attempt will be made to relate the observed distributions to limnological factors and to possible releases of radioactivity from nuclear power plants.

2. METHODS

2a. Sample Collection

Multiple 3"-diameter cores were taken from mainly the soft clays and muds at the stations shown in Figure 1. Coring operations were carried out on the University of Michigan research vessels, Inland Seas, Laurentian and Mysis, during several cruises between 1972 and 1974. The cores were characterized in nearly all instances by the presence of an extremely fluid brown flocculent material up to 1 cm thick at the sediment/water interface overlying either more consolidated black sediment or, particularly on the western side of the lake, pink glacial till. The gravity coring unit was lowered as slowly as possible into the sediments to prevent lateral motion of this flocculent material. The plastic core liners, containing 2—3 ft of sediment with overlying water, were removed from the corer barrel and stored in an upright position. Cores that showed the minimum of disturbance were carefully sectioned, using a hydraulic extruder, after the floc had been allowed to settle. Sections were taken at 0.5-, 1.0-, or 2.0-cm intervals, depending upon the depth in the core, by extruding the sediment into a scribed collar (the liquid floc was generally removed by use of a syringe). The sectioned material was stored in preweighed plastic bottles and frozen for return to the laboratory. The bulk density was measured before air drying at 90°C for 48 hr before analysis so that the porosity of each section could be determined.

2b. Radioactivity Measurements

The $^{137}\text{Cs}$ activity in each section was determined by gamma counting, in standard glass scintillation vials, ≤ 20 g of the oven-dried samples for up to 800 min by use of a 10 x 10 cm NaI detector multichannel-analyzer system. Standards of $^{137}\text{Cs}$, uranium ore, thorium ore, and potassium having the same density and shape as the unknowns were used and the $^{137}\text{Cs}$ concentrations were determined by least-squares analysis of the resulting gamma spectra [4].
These samples were analyzed for plutonium by use of techniques similar to those described by Golchert [5], Talavitie [6], and Wong [7]. Up to 5-g aliquots of the ground and well-mixed air-dried material, with 1—2 pCi of $^{242}$Pu as an isotopic diluent, were digested in concentrated nitric acid to destroy organic material and then in aqua regia. If the sample is to be analyzed for $^{210}$Pb [8], the sample is treated with concentrated hydrochloric acid until all the nitric acid has been destroyed. The sample is then centrifuged and filtered to remove all solids that are resistant to this digestion procedure and the supernatant taken down almost to dryness. This residue is taken up in 20 ml of 8 M nitric acid which is then passed through a column of Dovex 1 x 8 (100—200 mesh) which has been preconditioned with nitric acid. The column is washed with 75 ml of 8 M nitric acid to remove all traces of uranium and almost all other heavy elements. In order to remove thorium, the column is then washed with 100 ml of 12 M hydrochloric acid. Plutonium is then eluted with 50 ml of a mixture of 0.1 M hydrochloric acid and 0.01 M hydrofluoric acid.

The eluent is taken to dryness, redissolved in 5 ml of concentrated HCl, taken to dryness again. This step is repeated twice. Finally, the residue is taken up in a few drops of 6 M HCl and then a 5 ml of a mixture of 1 M NH$_4$Cl and 0.01 M oxalic acid with warming. The plutonium is plated out of this solution onto pre-electropolished stainless steel discs as described by Talavitie [6].

The plutonium is finally determined by alpha spectroscopy by use of silicon barrier detectors whose background in the region of interest 4.5—5.5 MeV is $\leq 0.001$ cpm. Activities of $^{238}$Pu and $^{239,240}$Pu are determined by comparing the integrated count in their respective peaks with that in the $^{242}$Pu peak.

3. RESULTS AND DISCUSSION

The vertical distribution of $^{137}$Cs has been measured in sixty sediment cores, and $^{239,240}$Pu in fourteen. A large proportion of these cores were taken from the southern basin of the lake (i.e., south of latitude 43° N, Figure 1). Since there have been only minor releases of radioactivity from nuclear power plants around the lake, effectively all the $^{239,240}$Pu and $^{137}$Cs found in the lake results from nuclear testing in the 1950's and early 1960's. As only a minor contribution to the total activity in the lake has come from runoff into tributary rivers and streams [9], the activity is a result of wet and dry deposition of radioactive fallout over the lake.

In all the cores studied, the $^{239,240}$Pu and $^{137}$Cs activity was confined to the uppermost 6 cm of the sediment, and in many cores no activity was found below 3 cm. These findings indicate that surveys of radionuclides performed using less exact sampling techniques, such as taking grab samples, are probably quite unreliable because of the likelihood of losing the uppermost flocculent layer during sampling. Typical $^{239,240}$Pu and $^{137}$Cs profiles are shown in Figures 2—5. These profiles illustrate the different observed types of distribution of these two radionuclides with depth in sediment cores taken.
from Lake Michigan. The shapes of the profiles will depend upon the time variability of the input of $^{239,240}\text{Pu}$ or $^{137}\text{Cs}$ to the lake, their residence times in the water column, the sedimentation rate, the effects of vertical mixing due to biological or physical processes in the uppermost sediment layers, and the finite thickness of the sampling interval [8].

Wahlgren and Nelson [10] have shown from measurements of $^{239,240}\text{Pu}$ and $^{137}\text{Cs}$ in samples of water from the lake during the period 1966—1973, that the residence time of these radionuclides in the water column is of the order of 1 yr (in contrast to $\geq 100$ yr for conservative elements). The result is that less than 5% of the total activity which entered the lake over the last 20 yr remains in the water column, and also, since the annual flux to the lake has been extremely variable, two horizons should be preserved in the sediments. One corresponding to the onset of nuclear testing around 1950 and the other to the maximum in activity corresponding to the 1963 input. Present day inputs are more than an order of magnitude lower than this latter value [11].

The total concentration of $^{239,240}\text{Pu}$ or $^{137}\text{Cs}$ at any location in the lake will depend upon three factors: the flux $P_t$ (in pCi/cm$^2$ per yr) at the air/water interface, the sedimentation rate $R$ (in cm/yr) or $\omega$ (in mg/cm$^2$ per yr), and a factor $f$ by which the flux $P'_t$ (in pCi/cm$^2$ per yr) measured at the sediment/water interface is normalized to the flux $P_t$ at the air/water interface, where $P'_t = fP_t$. Here the time $t$, measured backward from the present, is in years. Therefore, the activity in sediment at a depth $z$ in a core corresponding to $t$ years ago is

$$A_t = (fP_t/\omega) e^{-\lambda t}$$

where $\lambda$ is the radioactive decay constant. As the sedimentation rate in Lake Michigan (between 0.01 and 0.3 cm/yr) results in an annual deposit that is thinner than the thinnest section that can be reliably produced, the activity in a layer between $t$ and $t+\delta t$ years, corresponding to a depth between $z$ and $z+\delta z$ is

$$A_{t+\delta t} = \int_t^{t+\delta t} (fP_t/\omega) e^{-\lambda t} dt$$

Here the relationship between $t$ and $z$ is given by

$$t = \left(\frac{\rho_s}{\omega}\right) \int_0^z (1 - \phi_z) dz$$

where $\rho_s$ is the density of the sediment particles and $\phi_z$ is the porosity at depth $z$. These equations provide a simple model for the distribution of radioactivity in sediments.

However, in several cores it has been observed that the two horizons appeared to be deeper in the core than would be expected on the basis of sedimentation rates from the $^{210}\text{Pb}$ measurements, and that there was a corresponding interval of constant activity of $^{210}\text{Pb}$ at the top of the sediment cores. These observations have been interpreted in terms of a homogenous layer at the surface of the sediment column due to either physical or
biological turbation, as has been postulated elsewhere [12]. The depositional model can be altered to take account of this process. It is assumed that this homogenous layer is of constant thickness, all new inputs are instantaneously mixed within it, and a constant fraction of the total mass of sediment contained within it (equivalent to the annual deposition) is permanently buried each year. Thus, if the first introduction of plutonium or cesium into the sediments occurred \( k \) years ago and the thickness of the homogenous layer is equivalent to \( N \) years of sedimentation, it can readily be shown that the activity measured at present in sediment taken from a depth corresponding to \( j \)th year of deposition is

\[
A_{j+N} = \frac{f}{N \omega} \left[ \sum_{n=j}^{k} \left( \frac{N-1}{N} \right)^{n-j} \cdot e^{(n-j) \lambda} \cdot p_n \right] e^{(1-j)\lambda}
\]

where \( j \leq n \leq k \). The best values for the sedimentation rate, mixing depth, and the normalization factor \( f \) are obtained from the distribution of each radionuclide in the core by means of an interactive least-squares method.

The dashed lines in Figures 2–5 represent the best fits to the data calculated from this model. Values of the sedimentation rate, mixing depth, normalization factor, and the total activity per unit area of the lake bottom are given in Tables I and II. In those cores for which the sedimentation rate has been determined radiometrically from the \( ^{210}\text{Pb} \) profiling [8,13] (Table I), the depths to which \( ^{239,240}\text{Pu} \) and \( ^{137}\text{Cs} \) activities are found are consistent with the known history of deposition from the atmosphere as a result of nuclear testing. The agreement between the values of the sedimentation rates calculated from the measured profiles of these three radionuclides indicates that this method of sampling resulted in no significant losses of material from the sediment/water interface. If significant losses of material had occurred during sampling, the sedimentation rates calculated from the \( ^{239,240}\text{Pu} \) and \( ^{137}\text{Cs} \) profiles would be lower than those calculated from the \( ^{210}\text{Pb} \) profiles. This arises because the flux of \( ^{210}\text{Pb} \) to the lake is constant \((P_t = \text{const} = 0.2 \text{ pCi/cm}^2 \text{ yr})[14] \), and the sedimentation rate is calculated solely from its rate of decay, while \( P_t \) is variable for the other two nuclides and the estimate of the sedimentation rate is almost entirely based on the position of two horizons as described earlier.

As a result of the seismic profiling experiments conducted in southern Lake Michigan, it is known that the distribution of sediments is extremely variable [15]. Large areas of the western side of the lake exhibit virtually zero sedimentation; but in other areas close to the eastern shore, where there are several large rivers (Figure 6a), the post-glacial sediment layer is up to 10 m thick. The \( ^{239,240}\text{Pu} \) and \( ^{137}\text{Cs} \) profiles shown in Figures 2 and 3 (station 73–5 and 73–6) are for areas where there is an intermediate rate of sedimentation, and the profiles shown in Figures 4 and 5 are for areas of low (72–54) and high (74–1) sedimentation, respectively.

The profile shown in Figure 4 is representative of many areas on the western side of the lake where there is a thin layer of brown floc (1–2 cm thick) overlying glacial till or clean sand. Since detectable concentrations of
239, 240Pu and 137Cs have been found within the sampling interval that includes the interface between the floc and the till, it is reasonable to suppose that this floc layer is of a transitory nature and that the sediments as represented by this core are undergoing episodic periods of deposition and erosion. As a result, there is a large probability that this flocculent material, which contains relatively high concentrations of 239, 240Pu or 137Cs, is undergoing resuspension and transport to other locations in the lake.

Such a remobilization of sedimentary material is required in interpreting the two profiles representative of regions of intermediate sedimentation (Figures 2 and 3). The profiles calculated on the basis of the least-squares fits indicate a much greater drop in the expected concentration in the upper few centimeters (as a result of the large decrease in atmospheric flux since 1963) than is reflected in either the 239, 240Pu or 137Cs data. This excess plutonium or cesium can originate from two possible sources: runoff from the watershed or remobilization of surficial sedimentary material. The present estimate of the contribution of cesium (or plutonium) from the watershed due to erosional losses from the soil is still less than the atmospheric inputs [9]. Therefore, it would appear that the excess radioactivity in these two cores is due to remobilization of surface sediments, probably from those areas where little permanent sedimentation occurs. The final profile (Figure 5) is representative of an area where there is high sedimentation and considerable mixing. At this station, the sedimentation rate is 0.5 cm/yr and the mixing depth is 4 cm. The effect of mixing is to produce a region of nearly constant activity at the top of the core and to broaden the maximum in concentration corresponding to the 1963 maximum in input.

The resuspension and transport of sediments will have far-reaching effects on the long-term distribution of long-lived radionuclides such as plutonium in Lake Michigan (and other lakes). They will be particularly important in terms of the fate of radionuclides discharged by nuclear power plants and in determining the validity of present and proposed monitoring schemes. From the data shown in Tables I and II, it is clear that there are significant variations in the distribution of 239, 240Pu and 137Cs in the sediments of the lake. The total deposition of plutonium varies between 0.02 and 0.427 pCi/cm² and that for cesium between 0.38 and 31.6 pCi/cm². The average deposition per unit area has been 0.2 pCi/cm² and 8.0 pCi/cm² for Pu and Cs, respectively, and therefore, the flux normalization factors f vary between 0.05 and 3.1. These results suggest that there are large differences in the fluxes of plutonium or cesium arriving at the sediment/water interface. These differences in concentration at the surface, which the data presented in Tables I and II show to be very local, could arise from either a large variability in the flux from the atmosphere to the lake surface or from differences associated with the hydrodynamic and scavenging processes removing plutonium and cesium from the water column. Since it is well known, from the monitoring of world-wide fallout for 90Sr, that there are only small variations in flux within any latitude band [11], the observed differences in deposition probably arise for the latter reason.

If radioactivity remained unassociated with bulk sedimentary material in the water column and arrived independently at the bottom, or if the
sedimentation rate and the flux normalization factor \( f \) were constant over the whole area of the lake, the total activity per unit area should have the constant value \( fT^t \), or a. However, since the sedimentation rate is extremely variable (Figure 6a) and the values of \( f \) vary from station to station as well as from nuclide to nuclide (Table I), the removal of radioactivity from the water column is more than likely controlled by the composition of the particulates in the water column. Therefore, in order to compare the nature and magnitude of this scavenging process, ratios of the flux normalization factors for the three radionuclides were calculated. The values are shown in Table I.

From these values it will be noted that, in general, \(^{210}\text{Pb}\) is scavenged more effectively at deeper water stations (≥ 300 ft) where the \(^{210}\text{Pb}/^{137}\text{Cs}\) and \(^{210}\text{Pb}/\text{Pu}\) ratios are ≥ 2, and cesium and plutonium are more effectively scavenged at the shallower water stations. The values of the \(\text{Pu}^{137}\text{Cs}\) ratios are, with one exception (station 73-103°), very close to unity and therefore appear to be removed to the sediments with the same efficiency regardless of sampling location.

These observations can be accounted for in terms of their known behavior in the environment. Shukla and Leland [16] have concluded that the scavenging of lead from the water column is governed primarily by its complexation with organic matter rather than by absorption on clays or hydrous oxides. Since the fraction of organic carbon in the sediments increases with increasing water depth [17], the preferential scavenging of \(^{210}\text{Pb}\) offshore would not be unexpected. On the other hand, \(^{137}\text{Cs}\) is very strongly adsorbed by the clay component of soils [18,19] and, consequently, a comparatively greater scavenging in the inshore regions where they may be a large proportion of fine-grained clays in the water column is not unexpected. The strong association between plutonium and \(^{137}\text{Cs}\) is rather unexpected because of their widely differing chemical properties. However, Alberts and Wahlgren [20], have shown that the plutonium is associated primarily with the hydrous oxide fractions of the sediment from Lake Michigan. Therefore, since Shimp et al. [17] have shown that there is a strong correlation between hydrous oxides (such as \(\text{Fe}_2\text{O}_3\)) and the < 2 \(\mu\)m clay fraction, the observed similarity between the behavior of plutonium and cesium in the lake can readily be understood.

The apparent similarity between the behavior of plutonium and cesium in Lake Michigan is fortunate because it means that measurements of \(^{137}\text{Cs}\), which are far less costly and time consuming to perform than those for \(^{239,240}\text{Pu}\), can provide the necessary information to predict the long-term behavior of plutonium in the sediments. The two maps shown in Figure 6 compare the deposition of \(^{137}\text{Cs}\) and, by inference, \(^{239,240}\text{Pu}\) (presented in terms of the flux normalization factor \( f \)) and the mass sedimentation rate \( \omega \). The triangles denote stations where plutonium has been measured. It will be noted that the distribution of radioactivity parallels that of the sediments. Particularly high depositions occur to the north and west of the major river inputs and the greater part of the radioactivity is concentrated within the sediments of the eastern half of the lake. The pattern of sedimentation in the
southern half of the lake is very interesting in that whereas the bottom topography indicates an almost continuous slope down to the maximum depth of 540 ft, the maximum depth of sediment occurs at a water depth of only 240 ft (80 m). In addition, no sedimentation occurs at water depths less than 150 ft (50 m) even on the east side of the lake near the rivers, and therefore very little radioactivity is being deposited in the nearshore zone. From this one concludes that the deposition of radioactivity in the lake is being controlled mainly by lake-wide processes such as current structure and sources of the sediment. Therefore, because of the very short residence times in the water column, radioactivity which is released to the lake along the western shore will be rapidly transported to areas where there is a significant deposition of sediment; and for Lake Michigan, these areas are the band of sedimentation parallel to the eastern shore. Discharges of radioactivity along the eastern shore would also be found predominantly in this band of high sedimentation.

These results are important in that they suggest that any monitoring schemes to determine the long-term fate of effluents from nuclear power reactors must take account of the limnological properties of the receiving body of water. For Lake Michigan, a fixed sampling grid for sediments in the immediate environs of the plant would provide no significant information because of the physical and current structure of the lake.

ACKNOWLEDGEMENTS

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REFERENCES


### Table 1: Summary of Measured and Calculated Parameters from the Distribution of \( { }^{239,240} \text{Pu}, { }^{137} \text{Cs}, \) and \( { }^{210} \text{Po} \) in Sediment Cores Taken from Lake Michigan in 1972, 1973, and 1974.

<table>
<thead>
<tr>
<th>Station</th>
<th>Sedimentation Rate</th>
<th>Measured Depth</th>
<th>Total Activity</th>
<th>Flux Normalization Factor</th>
<th>Flux Ratio</th>
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<td>( \text{cm yr} )</td>
<td></td>
<td></td>
<td></td>
</tr>
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<td>72-1</td>
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<td>---</td>
<td>0.30</td>
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*\( a \) cm/yr
*\( b \) mg/cm²/yr
TABLE II. Summary of measured and calculated parameters from the distribution of $^{137}$Cs in sediment cores taken from Lake Michigan in 1972.

<table>
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<tr>
<th>Station</th>
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Fig. 1. Stations occupied in Lake Michigan for the sampling of sediments in 1972, 1973 and 1974.
Fig. 2. Measured and calculated $^{137}$Cs and $^{239,240}$Pu profiles in the sedimentary column at station 73-5. The sedimentation rate is 34 mg/cm$^2$ yr or 0.17 cm/yr at the surface and there is no surficial mixing.

Fig. 3

Measured and calculated $^{137}$Cs and $^{239,240}$Pu profiles in the sedimentary column at station 73-6. The sedimentation rate is 32 mg/cm$^2$ yr or 0.17 cm/yr at the surface and there is no surficial mixing.
Fig. 4. Measured and calculated $^{137}$Cs and $^{239,240}$Pu profiles in the sedimentary column at station 72-54. The sedimentation rate is 4.8 mg/cm$^2$ yr or 0.015 cm/yr at the surface and there is mixing down to a depth equivalent to 82 years of deposition.

Fig. 5. Measured and calculated $^{137}$Cs and $^{239,240}$Pu profiles in the sedimentary column at station 74-1. The sedimentation rate is 96.3 mg/cm$^2$ yr or 0.50 cm/yr at the surface and there is mixing to a depth equivalent to 12 years of deposition.
Fig. 6. Comparison of the mass sedimentation rate and total deposition of $^{137}$Cs or $^{239,240}$Pu in southern Lake Michigan. (a) The variation in mass sedimentation rate (mg/cm$^2$ yr). (b) The variation in the flux normalization factor for $^{137}$Cs. Sampling locations where $^{239,240}$Pu have been measured are identified by a triangle.