

Determination of recent sedimentation rates in Lake Michigan using Pb-210 and Cs-137

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Abstract—This paper describes the use of ^{210}Pb and ^{137}Cs radioactivity measurements to determine the rates of sedimentation in the Great Lakes. Cores from eight locations in Lake Michigan were chosen for examination to cover as wide as possible a range of sedimentation rates and representative sedimentary environments. The surficial ^{210}Pb activity in the sediments varies between 7 and 23 pCi/g dry wt and its profile in each core shows the expected exponential decrease with depth consistent with the assumption of uniform sedimentation rate over the last hundred years and secular equilibrium between supported ^{210}Pb and ^{226}Ra (0.5–1.0 pCi/g dry wt). Companion measurements of ^{137}Cs indicate that the coring technique satisfactorily recovered the uppermost levels of the deposit and that the mobility of both radionuclides within the sediment is probably small.

Based on the limited number of cores analyzed to date, it appears that modern sedimentation rates are not very different from average rates for the last 7000 yr. The excess ^{210}Pb appears to originate primarily from atmospheric fallout, but a further inventory of the ^{210}Pb distribution over the lake bottom must be made to properly assess the significance of other sources. The spatial distributions of both ^{137}Cs and ^{210}Pb at certain stations suggest that the mode of transport of these radionuclides are comparable and involve attachment to settling particles. A mathematical model is developed which accounts for the observed limited mobility of both ^{210}Pb and ^{137}Cs in several of the cores in terms of post-depositional redistribution by physical or biological mixing processes.

INTRODUCTION

A KNOWLEDGE of the rate at which sediments have been accumulating in the Great Lakes during the past several hundred years is clearly of fundamental importance in understanding aquatic and sedimentary geochemical processes which may have been measurably altered in post-settlement times. In general, the interpretation of chemical profiles in fresh-water surficial sediments has been significantly limited by the virtual absence of a knowledge of sedimentation rates. When rates have been available, they have often been generalized rather than specific for a core under examination. Furthermore, sedimentation rates based on palynological (KEMP *et al.*, 1972; BORTELSON and LEE, 1972), radiocarbon (LIBBEY, 1967; GROSS *et al.*, 1970) or stratigraphic (LINEBACK *et al.*, 1970) methods often provide only historical averages involving many meters of sediment. Such measures are not only lacking in necessary accuracy and detail, but may not adequately reflect rates within the upper 20 cm or so of sediment where significant sediment–water exchanges are occurring at the present time.

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In circumstances where sedimentary diagenesis is controlled by chemical reactions and diffusion, measurement of the sedimentation rate in the vicinity of the sediment-water interface is essential for the development of relevant quantitative molecular diffusion models describing the transport of substances within pore water and their transfer across the sediment-water boundary (TZUR, 1971; BERNER, 1964). In cases where significant culturally-derived components are present in addition to natural backgrounds, such as in the case of accumulations of heavy metals observed in the uppermost few centimeters of fine-grained lacustrine sediments in southern Lake Michigan (LINEBACK and GROSS, 1972; SHIMP *et al.*, 1971), measurements of the recent sedimentation rate may play a decisive role in determining the relative importance of man's effect versus naturally-occurring diagenesis. In special instances where there is no significant natural background and the time-dependence of the input of a pollutant to the aquatic system is comparatively well-known, such as in the case of ^{137}Cs and ^{90}Sr (LERMAN and TANIGUCHI, 1972), or possibly pesticides (LELAND *et al.*, 1973), a detailed measurement of sedimentation rate may allow the effects of the aquatic system on the input to be separated from subsequent diagenesis through diffusion or physical mixing processes (CULLÉN, 1973; BERGER and HEATH, 1968).

Application of radiometric methods to sedimentary geochronology has enjoyed considerable success. Recently, KRISHNASWAMI *et al.* (1971) evaluated the use of ^{210}Pb as well as three other radionuclides (^{32}Si , ^{55}Fe , and ^{137}Cs) for dating recent freshwater lake sediments. These authors concluded that ^{210}Pb is ideal for dating lake sediments as far back as a century or so. Subsequently, KOIDE *et al.* (1972, 1973) further validated the utility of the method and determined sedimentation rates in a series of small lakes.

In this paper we report on the measurement of ^{210}Pb in sediment cores and its utility for the determination of sedimentation rates in Lake Michigan. We shall also briefly examine the feasibility of using ^{137}Cs fallout from nuclear testing for the same purpose.

METHODS

Sample collection and handling

Multiple 3-in. diameter cores were taken from the soft clays and muds at the stations in Lake Michigan shown in Fig. 1. Coring operations were carried out aboard the University of Michigan Research Vessel *Inland Seas* during May and June of 1972. Care was taken during coring to ensure minimal disturbance of the sediment-water interface, which is characterized in nearly all instances by an extremely fluid, brown flocculent material of up to 1 cm thickness overlying more consolidated sediment. The gravity coring unit was lowered as slowly as possible into the sediments to prevent lateral motion of the overlying floc in response to the pressure wave created by the descent of the corer. The plastic core liners, usually containing several feet of sediment with overlying water, were removed from the corer barrel, held in a vertical position and placed vertically in a stand where they were further inspected for integrity. Carefully chosen cores were sectioned, using a hydraulic extruder, after allowing the floc to resettle for up to an hour during which time the cores were kept at approximately their *in situ* temperature to prevent expansion due to gas formation. The suspension of brown floc was removed by a syringe, while, for the more consolidated sediments, intervals of well-defined thickness were isolated by means of a centimeter-scribed collar, placed over the end of the core liner, into which an appropriate amount of material was extruded. Core sections were stored frozen for return to the laboratory. Measurements were made of the bulk density before air drying the samples in an oven at 90°C for 48 hr prior to analysis.

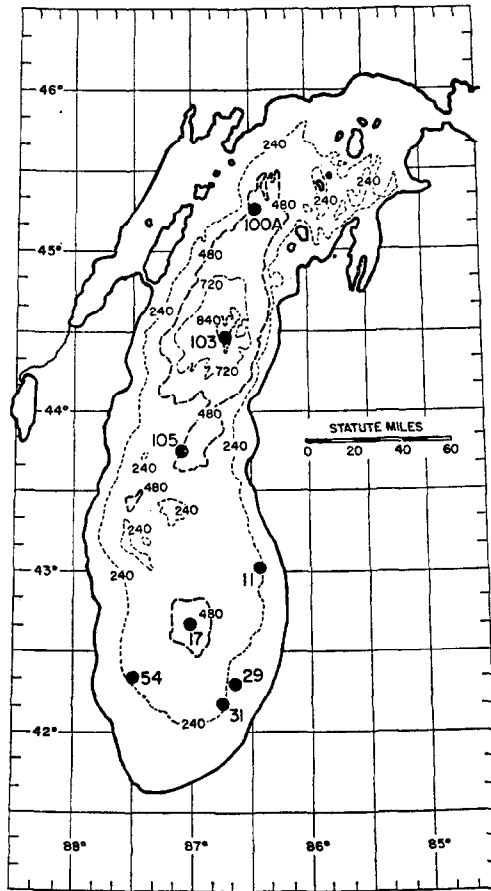


Fig. 1. Sampling stations occupied in Lake Michigan on the University of Michigan-Argonne cruise, June 1972. Depth contours given in feet.

Radioactivity measurements

The determination of the ^{210}Pb content is based upon the measurement of the daughter ^{210}Po activity which is assumed to be in secular equilibrium with its parent. Up to 5 g aliquots of ground air-dried samples were digested first in concentrated nitric acid, then in several aliquots of concentrated hydrochloric acid. The HCl treatments continued until no further fumes of NO_2 were evolved. The solution was then diluted and the insoluble matter filtered off. The filtrate was evaporated to small volume and further heated with concentrated hydrochloric acid to eliminate any remaining nitric acid. Elimination of the nitric acid in the filtrate is essential for the proper self-plating of ^{210}Po on to the silver planchettes. The ^{210}Po alpha-activity was determined using argon-purged, low background, 2π proportional counters connected to a simple scaler (HOLTZMAN, 1969). The counting error was generally less than 3 per cent, and in the upper sections of the core less than 1 per cent.

The ^{137}Cs activity in each section was determined by gamma counting 20 g of the over-dried samples for up to 800 min using a 4 in. \times 4 in. NaI detector-multichannel analyzer system. A ^{137}Cs standard, having essentially the same geometry and density was used. This was prepared from a standard solution supplied by Amersham-Searle. The limit of detection for ^{137}Cs by this method is 0.1 pCi/g and the counting error is $\leq \pm 10$ per cent in the upper layers of the core. Measurements were also made for this nuclide using a 30 ml lithium-drifted germanium detector

coupled to a 4096 channel multichannel analyzer. This method permits a clear separation of the 661 KeV ^{137}Cs gamma rays from possible contributions from ^{208}Tl (583 KeV) and ^{214}Bi (609 KeV). The samples were counted for at least 24 hr to obtain good statistical accuracy.

The concentration of ^{226}Ra in several of the samples was determined by the radon emanation technique (LUCAS, 1957) using aliquots of the same solution as were used for determination of ^{210}Pb .

RESULTS AND DISCUSSION

Model for distribution of ^{210}Pb in sediment cores

KRISHNASWAMI *et al.* (1971) presented a simple model which successfully described their observed ^{210}Pb profiles in sediments. In their model it is assumed that (1) the flux of excess ^{210}Pb to the sediment-water interface is constant, (2) the sedimentation rate is constant at all times, (3) there is no post-depositional migration of the radionuclide within the sediments, and (4) the activity of ^{210}Pb supported by ^{226}Ra in the sediments is independent of depth. Under these conditions the expected activity in pCi/g dry weight of a sediment section of age, t , is simply

$$A(t) = (P/\omega)e^{-\lambda t} + A', \quad (1)$$

where P is the flux of ^{210}Pb at the sediment-water interface in pCi/cm² yr, ω is the sedimentation rate in g/cm² yr, A' is the activity of supported ^{210}Pb in pCi/g dry weight, and λ is the radioactive decay constant for ^{210}Pb ($= 0.693/22.26 \text{ yr}^{-1}$) LEDERER *et al.*, 1967).

Equation (1) can be rewritten in terms of the depth z , cm, below the sediment-water interface,

$$A(z) = \frac{P}{R\bar{\rho}_s(1-\phi)} e^{-\lambda z/R} + A', \quad (2)$$

since

$$\omega = R(1-\phi)\bar{\rho}_s, \quad (3)$$

and $t = z/R$, where R is the sedimentation rate in cm yr⁻¹, ϕ is the corresponding sediment porosity and $\bar{\rho}_s$ is the effective density of the solid phase (found to be $2.45 \pm 0.05 \text{ g/cm}^3$ for all sediment cores examined). In the upper 20 or so cm, the porosity exhibits a strong decrease with depth below the sediment-water interface. In homogeneous sediments the porosity $\phi(z)$ at any depth z may be described by the equation (ATHY, 1930)

$$\phi(z) = (\phi_0 - \phi')e^{-\beta z} + \phi', \quad (4)$$

where ϕ_0 is the porosity at the sediment-water interface and ϕ' is the estimate of the porosity at final compaction, $\phi(z = \infty)$. While the effects of compaction can, of course, be taken into account without the use of such an analytic representation, it is useful for developing an explicit function relating depth to age in homogeneous compacting sediments.

In a column of unit cross-section, the total solids occupy a volume given by

$$V_s(\text{tot}) = \int_0^z (1-\phi) dz', \quad (5)$$

which must be equal to the volume transferred in time T

$$V_s(\text{tot}) = \int_0^T \frac{\omega}{\bar{\rho}_s} dt' = \frac{\omega T}{\bar{\rho}_s}. \quad (6)$$

Hence the correspondence between the age of the section and its depth is

$$T = \frac{\bar{\rho}_s}{\omega} \int_0^z (1 - \phi) dz', \quad (7)$$

and then substituting equation (7) in equation (2) we obtain

$$A(z) = \left\{ \frac{P}{\bar{\rho}_s R_0 (1 - \phi_0)} \right\} \exp [-(\lambda/R_0) f(z)] + A', \quad (8)$$

where

$$f(z) = \frac{1}{(1 - \phi_0)} \int_0^z (1 - \phi) dz', \quad (9)$$

and R_0 is the sedimentation rate at the sediment-water interface. In the absence of compaction $f(z) = z$, since $\phi(z) = \phi_0$ and T will simply be equal to z/R_0 as in equation (2). Equation (9) may be explicitly evaluated by substituting in equation (4)

$$f(z) = \frac{1}{(1 - \phi_0)} \left[z(1 - \phi') + \frac{[\phi(z) - \phi_0]}{\beta} \right]. \quad (10)$$

To complete a proper formalism for the analysis of ^{210}Pb activities in compacting but otherwise undisturbed sediments we must include the effect of a finite sampling interval. If the j th sampling interval is from z_j to z_{j+1} with $\Delta z_j = z_{j+1} - z_j$, the expected activity per gram of homogenized sample in the interval is

$$A_{z_j/z_{j+1}} = A \Delta z_j = \frac{1}{\Delta z} \int_{z_j}^{z_{j+1}} A(z') dz'. \quad (11)$$

Equations (8), (10) and (11) together constitute a model for the prediction of the measured profiles of the ^{210}Pb activities.

Application of the model to the ^{210}Pb data.

The porosity profiles at Stations 72-31 and 72-17 are shown in Fig. 2. From the data for Station 72-31 values of $\phi_0 = 0.882$, $\phi' = 0.763$ and $\beta = 0.019$ while for Station 72-17, $\phi_0 = 0.925$, $\phi' = 0.820$, and $\beta = 0.028$ were obtained. Cores from Stations 72-11 and 29 have a porosity profile similar to that from Station 72-31, while cores from Stations 72-100A, 103' and 105' have a porosity profile not differing significantly from that of Station 72-17.

The Pb^{210} profiles at the eight stations are shown in Fig. 3 and the data given in Table 1, together with a few values at each station for the ^{226}Ra concentration. The continuous lines shown in Fig. 3 are derived from an evaluation of equations (8), (10) and (11) for values of R_0 and P/ω which give a minimum variance in a two parameter least squares fit to the data. In most cases A' was held fixed at the value of the minimum ^{210}Pb activity in the core, or the average ^{226}Ra activities. The lines are merely an aid to visualization since equation (11) predicts values for

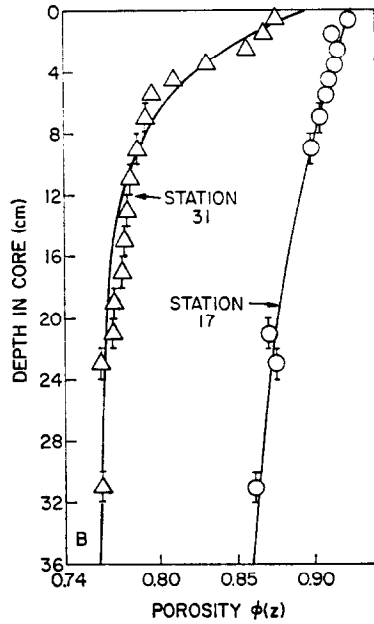


Fig. 2. Porosity profile $\phi(z)$ showing the effects of compaction at Stations 72-17 and 72-31.

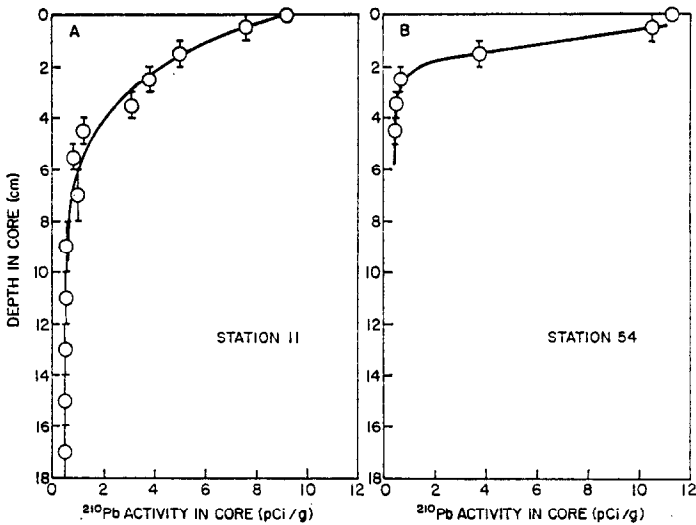


Fig. 3. Measured and calculated ^{210}Pb profiles at each station. Solid line is the theoretical fit obtained using equations (8), (10) and (11) excluding vertical mixing ($S = 0$), and the dashed lines the theoretical fit obtained with vertical mixing included in the model [equations (14) and (11)].

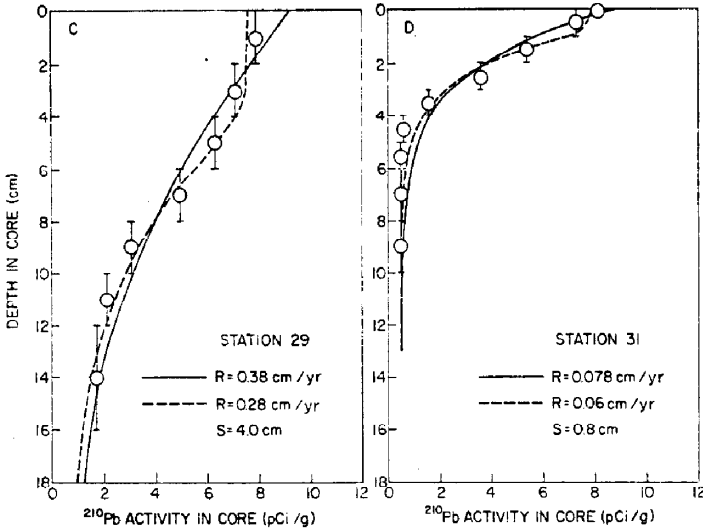


Fig. 3 (c, d)

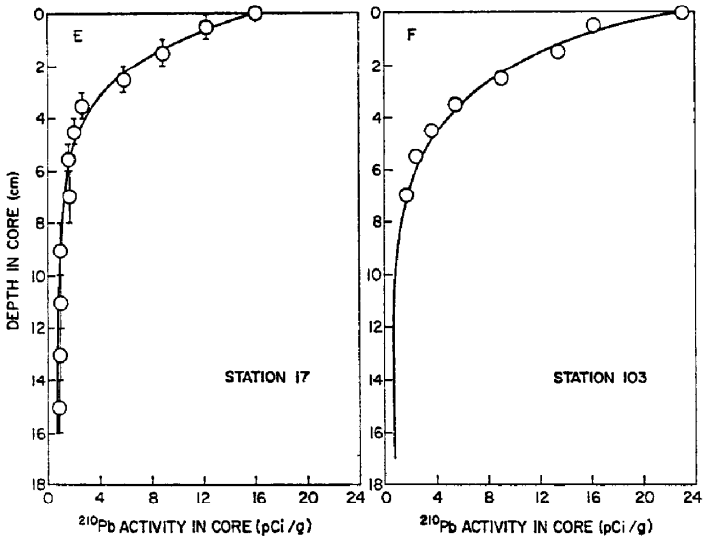


Fig. 3 (e, f)

finite sediment intervals. In calculating the least squares fit to the observed ^{210}Pb profiles it has been assumed that the floc occupies a layer at the surface which is thin with respect to the normal sampling thickness of layers in the core.

Values of the parameters derived from the least squares fit for all stations are summarized in Table 2. In addition to R_0 , P and ω , values are given for P/ω which is the excess activity of ^{210}Pb at the sediment-water interface (pCi/g) and R' (cm/yr) which is the rate of sedimentation at final compaction, $R' = R_0(1 - \phi_0)/(1 - \phi')$. If $\phi' = \phi_\infty$ the value of R' is the rate of increase in thickness of the whole sediment column with the respect to the basement (i.e. glacial till or bedrock).

The values obtained for the activity of ^{210}Pb in sections taken from sufficiently deep layers in cores where unsupported ^{210}Pb has totally decayed away (e.g. Station 72-17, Table 1) are very close to values measured for ^{226}Ra in selected sections for the same cores, indicating that the supported ^{210}Pb is almost in secular equilibrium with its parent ^{226}Ra . Therefore it can be assumed without significant error that the supported ^{210}Pb activity is constant and almost in secular equilibrium with ^{226}Ra in the entire core. The slight departure from secular equilibrium at depth in the core presumably indicates some mobility of the ^{226}Ra decay products, especially

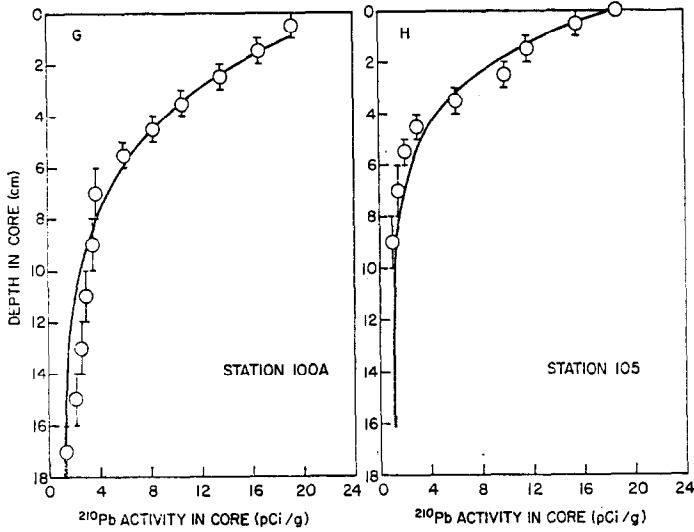


Fig. 3 (g, h)

Table 1. Summary of ^{210}Pb analyses and associated experimental data

Depth in core (cm)	Bulk density ρ_B	Fraction dry wt.	^{210}Pb (pCi/g)	^{226}Ra (pCi/g)	
Station no. 11 43°1.1'N, 86°24.3'W					
Floc	0.0	0.15	9.22		
0.0	1.00	1.14	0.23	7.60	
1.00	2.00	1.19	0.28	5.01	
2.00	3.00	1.21	0.30	3.83	0.60
3.00	4.00	1.22	0.31	3.07	
4.00	5.00	1.23	0.32	1.20	
5.00	6.00	1.24	0.33	0.84	0.69
6.00	8.00	1.24	0.33	0.98	
8.00	10.00			0.56	
10.00	12.00			0.54	
12.00	14.00			0.50	
14.00	16.00			0.51	
16.00	18.00			0.50	
30.00	32.00	1.48	0.54		

Table 1 (contd.)

Depth in core (cm)		Bulk density ρ_B	Fraction dry wt.	^{210}Pb (pCi/g)	^{262}Ra (pCi/g)
Station no. 17 42°40.0'N, 87°0.0'W					
	0-0		0.09	16.18	
0-0	1-00	1.08	0.19	12.32	
1-00	2-00	1.12	0.21	8.89	
2-00	3-00	1.10	0.20	5.93	0.90
3-00	4-00	1.10	0.21	2.74	
4-00	5-00	1.11	0.22	1.97	
5-00	6-00	1.12	0.22	1.57	
6-00	8-00	1.14	0.23	1.65	
8-00	10-00			0.96	
10-00	12-00	1.16	0.25	0.99	
12-00	14-00	1.16	0.25	0.88	
14-00	16-00	1.16	0.25	0.83	1.02
16-00	18-00	1.17	0.26		
20-00	22-00	1.21	0.32		
22-00	24-00	1.23	0.30		
50-00	52-00	1.33	0.34		
60-00	62-00	1.39	0.44		
Station no. 29 42°17.0'N, 86°38.0'W					
0-0	2-00	1.21	0.30	7.92	0.59
2-00	4-00	1.25	0.34	7.12	0.72
4-00	6-00	1.27	0.35	6.32	0.54
6-00	8-00			4.94	0.59
8-00	10-00			3.04	0.60
10-00	12-00			2.12	0.45
12-00	16-00			1.72	0.52
16-00	20-00			1.17	0.54
Station no. 31 42°10.0'N, 86°42.0'W					
	0-0	1.13	0.19	8.20	
0-0	1-00	1.19	0.30	7.28	
1-00	2-00	1.22	0.32	5.38	
2-00	3-00	1.26	0.35	3.60	
3-00	4-00	1.34	0.42	1.63	
4-00	5-00	1.41	0.47	0.60	
5-00	6-00	1.42	0.50	0.52	
6-00	8-00	1.45	0.51	0.56	
8-00	1-00	1.48	0.52	0.56	
10-00	12-00	1.50	0.54		
18-00	20-00	1.54	0.56		
30-00	32-00	1.57	0.59		
40-00	42-00	1.55	0.58		
50-00	52-00	1.55	0.59		

Table 1 (contd.)

Depth of core (cm)		Bulk density ρ_B	Fraction dry wt.	^{210}Pb (pCi/g)	^{226}Ra (pCi/g)
Station no. 54 42°20.0'N, 37°32.0'W					
	0-0		0.12	11.29	
0-0	1-00	1.15	0.25	10.52	
1-00	2-00	1.28	0.40	3.76	
2-00	3-00	1.45	0.53	0.65	
3-00	4-00	1.38	0.47	0.48	0.49
4-00	5-00	1.41	0.49	0.44	
8-00	10-00	1.36	0.44		
18-00	20-00	1.32	0.40		
30-00	32-00	1.31	0.40		
70-00	72-00	1.38	0.48		
Station no. 100A45°15.0'N, 86°24.5'W					
	0-0			19.34	
1-00	2-00	1.10	0.20	16.59	
2-00	3-00	1.11	0.15	13.58	
3-00	4-00	1.17	0.26	10.56	
4-00	5-00	1.10	0.19	8.28	
5-00	6-00	1.17	0.26	5.96	
6-00	8-00	1.13	0.20	3.78	
8-00	10-00	0.14	0.23	3.50	
10-00	12-00	1.14	0.22	2.94	1.28
12-00	14-00			2.60	
14-00	16-00			2.07	
16-00	18-00			1.29	
18-00	20-00			1.24	
20-00	22-00			1.18	
22-00	24-00			1.07	
Station no. 103 44°28.5'N, 86°42.3'W					
	0-0	1.04	0.07	23.13	
0-0	1-00	1.04	0.18	16.25	
1-00	2-00	1.13	0.18	13.56	1.09
2-00	3-00	1.13	0.19	9.01	
3-00	4-00	1.13	1.19	5.46	
4-00	5-00	1.14	0.20	3.62	1.26
5-00	6-00	1.14	0.21	2.48	
6-00	8-00	1.13	0.21	1.76	1.35
8-00	10-00	1.14	0.22	1.18	
10-00	12-00			1.31	
12-00	14-00			1.20	
14-00	16-00			1.16	
16-00	18-00			1.20	
Station no. 105 43°43.3'N, 87°2.6'W					
	0-0	1.01	0.03	18.74	
0-0	1-00	1.13	0.18	15.46	
1-00	2-00	1.10	0.19	11.70	
2-00	3-00	1.12	0.20	9.92	1.35
3-00	4-00	1.15	0.21	5.05	
4-00	5-00	1.14	0.23	2.96	
5-00	6-00	1.09	0.15	2.05	
6-00	8-00			1.46	1.00
8-00	10-00	1.15	0.24	0.95	

Table 2. Summary of measured and calculated parameters for ^{210}Pb dating of Lake Michigan sediments

Station	Sedimentation rate		Mass, ω (g/cm ² yr)	Flux of ^{210}Pb sediment-water interface, P (pCi/cm ² yr)†	Specific activity at sediment-water interface, P/ω (pCi/g)		Supported ^{210}Pb , A' (pCi/g)	Average ^{226}Ra (pCi/g)‡
	At surface R_0 (cm/yr)	At depth, R' (cm/yr)			(obs)	(calc)		
11	0.088	0.036	0.0198	0.17	9.22	8.64	0.50	0.65
17	0.066	0.028	0.0121	0.18	16.18	15.20	0.90	0.96
29	0.416	0.172	0.0938	0.65	6.93	6.90	0.57§	0.57
31	0.078	0.032	0.0176	0.14	8.20	8.07	0.56	
54*	0.01				11.29		0.44	0.49
100A	0.116	0.048	0.0213	0.38	19.34		0.80	1.28
103	0.080	0.033	0.0147	0.32	23.13	21.48	0.70	1.23
105	0.083	0.035	0.0153	0.27	18.74	17.86	1.17	1.17

* No calculation of compaction made because of very short length of apparent recent sedimentation. (Mixing depth = 1.0 cm). (See text.)

† The atmospheric flux of ^{210}Pb over Lake Michigan ≈ 0.2 pCi/cm² yr (See text and JAWOROWSKI, 1966).

‡ If there is no migration of daughter products of ^{226}Ra , supported ^{210}Pb should be in equilibrium with ^{226}Ra .

§ ^{210}Pb not measured deep enough in core, $A_{\text{Pb}^{210}} = A_{\text{Ra}^{226}}$.

^{222}Rn , within the sediment. Loss of ^{222}Rn from the surface of the sediments followed by decay to ^{210}Pb in the water column may alter the profile of supported ^{210}Pb within the upper few centimeters of the deposit. We assume that the extent of this redistribution is small in comparison to the flux from the atmosphere.

The inferred sedimentation rates over the past hundred years or so appear to be constant within the experimental uncertainties in the data. The validity of this method of dating was checked by examination of the distribution of pollen in a companion core taken at Station 72-11. The *Ambrosia* (ragweed) pollen horizon at approximately 10 cm depth in this core (HATCHER, unpublished data) reflects the effect of forest clearance around 1840 (DAVIS *et al.*, 1971; HATCHER, unpublished data). After correction for compaction a value of $R_0 = 0.11$ cm/yr was obtained (based on the assumption of a constant rate of sedimentation above the *Ambrosia* horizon). This value is in satisfactory agreement with that obtained from the ^{210}Pb method (0.088 cm/yr). This is an important result because it suggests that, like pollen, ^{210}Pb tends to remain in fixed association with the solid phase after deposition. If the nuclide possessed a significant post-depositional mobility, the inferred sedimentation rate would tend to be high relative to that obtained from pollen analysis.

The inferred sedimentation rates may also be compared with those calculated from the thickness of the Waukegan Member (assuming uniform sedimentation rates), which is the uppermost stratigraphic unit measured by seismic profiling (LINEBACK *et al.*, 1972). This member has been dated by the radiocarbon method at 7000 yr B.P. (GROSS *et al.*, 1973).

The results are summarized in Table 3. Since there are large unassignable uncertainties in the estimate of the post-glacial sedimentation rate, the agreement with our estimates from ^{210}Pb data and porosity calculations must be considered remarkably good. The results suggest that modern rates of sediment accumulation in Lake Michigan may not be very different from average rates over the last 7000 yr. In general, the data support the assumption of a uniform sedimentation rate over

Table 3. Comparison of sedimentation rates measured using the ^{210}Pb method (corrected for compaction) with those estimated from the thickness of the Waukegan Member

Station	Thickness of* member, L (cm)	Sedimentation rate (cm/yr)	
		Seismic, (R_w)†	This study (R')
11	300–600	0.042–0.084	0.039
17	150–300	0.021–0.042	0.028
29	900–1200	0.13–0.17	0.12
31	300–600	0.042–0.084	0.016
54	0–30	0.0–0.0004	0.01

* Data taken from LINEBACK *et al.*, 1972.

$$\dagger R_w = \frac{\text{Thickness of Waukegan Member}}{\text{age}} = \frac{L}{7000} \text{ cm yr}^{-1}.$$

the last hundred years or so. It is highly desirable to study post-glacial stratigraphic features together with ^{210}Pb in cores taken at the location to properly examine the question of modern versus historical rates.

Integrity of the cores

It is important to realize that the determination of sedimentation rates by the ^{210}Pb method is not critically dependent on retrieval of the top portions of the deposits during coring. However, evaluation of the unsupported flux of ^{210}Pb to the surface, P , requires locating the position of the sediment–water interface. KOIDE *et al.* (1973) have speculated that in normal coring devices there is a possibility of losing the top portions of the deposit and have therefore suggested that this loss may be checked by the presence of ^{137}Cs .

In most of the cores, measurements of the ^{137}Cs activity were made down to 10 cm, while in core 72–100A sections were analyzed down to 80 cm for possible ^{137}Cs activity. The profiles are shown in Fig. 4. In no case was ^{137}Cs found below 8 cm. This result may be contrasted to the recent work in Lake Ontario reported by BOWEN and NOSHKIN (1973) in which small concentrations of ^{137}Cs were detectable as deep as 50 cm in several of their cores.

The observed ^{137}Cs profiles (Fig. 4A–H) are generally consistent with the time scale established by the ^{210}Pb measurements and the known onset of testing of nuclear devices. This results indicates that, if there is post-depositional mobility of ^{137}Cs , the extent of loss of surficial material must be comparable to the extent of downward movement of this nuclide. Since the thin layer of floc was retrieved from each of the cores, the sediment–water interface is probably defined to better than one centimeter and the range of mobility of the radionuclide to a similar extent. These results indicate that the uppermost layers have probably been recovered.

Post-depositional mobility

The low surficial concentration of ^{210}Pb at several of the stations can possibly be interpreted in terms of diagenesis or vertical mixing. KOIDE *et al.* (1973) have attributed low surficial concentrations of ^{210}Pb in several of the cores they have

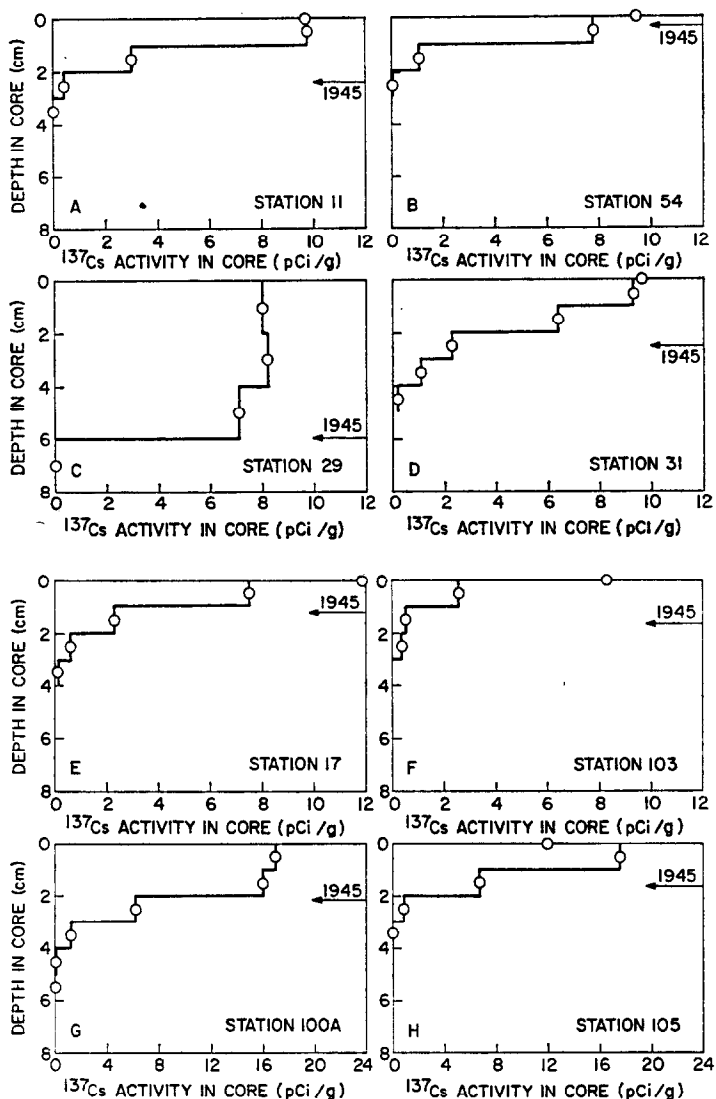


Fig. 4. Measured profiles of ^{137}Cs at all stations. The arrow in each case locates the depth, based on the best values of sedimentation rate from the ^{210}Pb measurements, below which no ^{137}Cs activity should be observed.

examined in terms of chemical remobilization of lead at the sediment-water interface. They suggested that ^{210}Pb , initially deposited under oxidizing conditions, is comparatively soluble in sediment pore water and can diffuse either outward into overlying water or into deeper, more reducing regions of the sediment where it is reprecipitated, perhaps as PbS . Presumably a similar mechanism could explain the ^{210}Pb profiles in selected fresh water lakes depending on their sediment geochemistry. However, in Lake Michigan cores with similar sediment characteristics taken in different locations do not show a predictable reduction in surficial ^{210}Pb . Moreover,

LELAND and SHUKLA (1973) find little evidence for lead mobility in sediments. It therefore seems more likely that the apparent low surficial concentrations are due to post-depositional redistribution of the sediment by physical mixing or bioturbation (CULLEN, 1973).

The effect appears to be greatest at Station 29 where the sedimentation rate is the greatest. To further examine the possibility of surficial mixing, a core sampled at 0.5 cm intervals was analyzed for ^{137}Cs at this station. The results, together with the estimated annual atmospheric flux of ^{137}Cs from fallout (HEALTH AND SAFETY LABORATORY, 1972) deposited in Lake Michigan, are shown in Fig. 5. Based on the value of the sedimentation rate of 0.0938 g/cm^2 from ^{210}Pb dating, the position of the 1963–64 maximum in fallout activity should be at 2.5 cm. However, the observed maximum occurs at 4.5 cm (Fig. 5b) indicating a significant downward mobility of ^{137}Cs at this station.

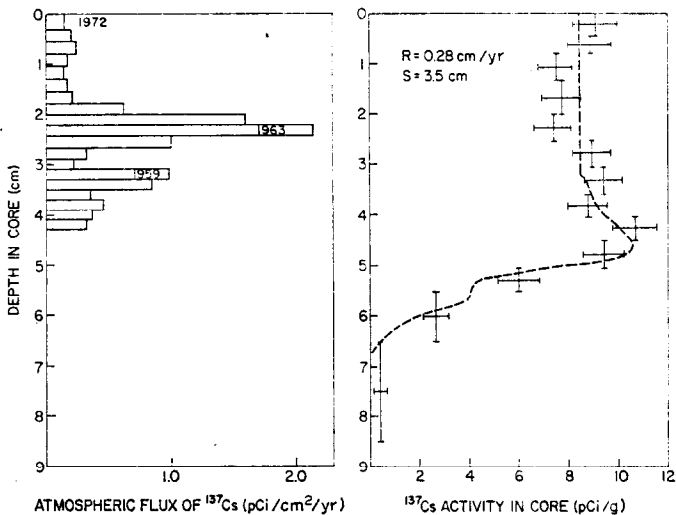


Fig. 5. (a) The estimated annual flux of ^{137}Cs to the lake surface from atmospheric fallout, plotted vs depth in the sediment using a time scale established from the ^{210}Pb measurements at Station 72–29.

(b) Measured and calculated profile at Station 72–29 for ^{137}Cs , calculated assuming steady-state vertical mixing. This model satisfactorily accounts for the difference between the observed depth of maximum activity in the core and that expected from Fig. 5a for undisturbed sediments.

This 2 cm displacement cannot be accounted for in terms of simple molecular diffusion because the diffusion coefficient of ^{137}Cs in sediment appears to be too low (DUURSMA and BOSCH, 1970). Recent measurements by RITCHIE *et al.* (1973) and PENNINGTON (1973) of ^{137}Cs in the sediments of reservoirs and lakes, as well as the results at other stations in this study, confirm its low diffusional mobility. This point is not absolutely clear as can be seen from the analysis of cores from the other Great Lakes by LERMAN (1973) and LIETZKE *et al.* (1973). Diffusion should not change the position of the peak activity as KRISHNASWAMI *et al.* (1971) point out in discussing the nature of ^{137}Cs profiles in cores from Lake Lemman. They suggest that

the maximum value of the activity at a greater depth than expected is due to physical or biological mixing. There should be a discrete horizon preserved in the sediment core for this nuclide from nuclear testing, whose location should be very sensitive to mixing effects.

While such mixing processes are undoubtedly particle-selective, it is still useful to consider a simple model in which a zone at the top of the sedimentary column is assumed to be completely homogeneous. As this zone of fixed thickness, corresponding to N yr of sedimentation, moves upward with respect to the basement, material is transferred from this zone to underlying sediment where no further mixing occurs (DAVIS, 1967).

To develop a mathematical representation it is convenient to simulate the growth of the sediment column above the fixed basement in annual increments starting with the onset of the introduction of radioactivity into the sediments. If the first input of radioactivity occurred k yr ago, then the deepest that activity will be measured in the sediment column will be at a depth corresponding to $k + N$ years.

The activity observed at this depth at the time of sampling will be (in pCi/g)

$$A_{k+N} = (fP_k/N\omega)e^{(1-k)\lambda} \tag{12}$$

where P_k is the intergrated annual flux (pCi/cm²) to the air-water interface and f is the flux normalization factor which is a measure of the efficiency of transfer of radioactivity through the water column to the sediment-water interface. $N\omega$ is the total mass of sediment solids in the mixing zone (g/cm²). The activity observed now at a depth corresponding to the deposition $k + N - 1$ yr ago, the second year of input is:

$$A_{k+N-1} = \frac{1}{N\omega} \left[fP_k \left(\frac{N-1}{N} \right) e^{-\lambda} + fP_{k-1} \right] e^{(2-k)\lambda} \tag{13}$$

The first term within the parentheses is the contribution of the first year of input which remained in the homogeneous zone. In general, the activity at a depth corresponding to $j + N$ yr of deposition is given by

$$A_{j+N} = \frac{f}{N\omega} \left[\sum_{n=j}^k \left(\frac{N-1}{N} \right)^{(n-j)} e^{-(j-n)\lambda} P_n \right] e^{(1-j)\lambda} \tag{14}$$

where $j \leq n \leq k$. The correspondence between the age of the sediment layer, $j + N$ yr, and the depth in the core is given by equation (14), (7) and (11) constitute a model for the prediction of the shape of measured profiles provided that the source term P_n is known. An iterative least squares method is used to obtain best values for the sedimentation rate, R_0 , the mixing depth, S (cm), and the normalization factor, f .

The result of using this model to predict the distribution of ¹³⁷Cs in core 72-29 is shown as the dashed line in Fig. 5b. Minimum variance was obtained using $R_0 = 0.28$ cm/yr, $S = 4.0$ cm and $f = 1.79$. Since the sedimentation rate is relatively large at this station, temporal variations in the ¹³⁷Cs input to the sediment appear to be preserved in the sediment column even with considerable surficial mixing.

Table 4. Comparison of parameters derived from ^{137}Cs and ^{210}Pb dating of Lake Michigan Sediments

Station	^{137}Cs				^{210}Pb				
	Sedimentation rate, R_0 (cm/yr)	Mixing depth, S (cm)	Flux normalization factor (f_{Cs})	Specific activity (f_{Cs}/ω)	Sedimentation rate, R_0 (cm/yr)	Mixing depth, S (cm)	Flux normalization factor (f_{Pb})	Specific activity (f_{Pb}/ω)	Flux ratio (f_{Pb}/f_{Cs})
11	0.04	1.0	0.472	23.8	0.10	0	1.07	54.0	2.26
29	0.28	3.4	2.329	24.8	0.28	4.0	4.299	45.6	1.84
31	0.05	2.0	0.706	40.1	0.07	0.8	0.874	49.6	1.24
17	0.078	0.2	0.242	20.0	0.066	0	0.900	74.6	3.72
100A	0.108	1.0	0.868	40.75	0.134	1.0	2.693	126.3	3.10
103	0.074	0	0.0796	5.40	0.080	0	1.600	108.0	20.0
105	0.053	0.8	0.604	39.40	0.083	0	1.350	87.9	2.23

Since the flux of ^{210}Pb at the air-water interface is assumed to be constant ($= P = 0.2$ pCi/cm² yr) equation (14) may be simplified. The expression within the brackets may be replaced by $P/[N - (N - 1)e^{-\lambda t}]$ which is the steady-state activity of ^{210}Pb in the homogeneous layer. The results of the three parameter least squares fit for the ^{210}Pb data on a different core from Station 72-29 is shown in Fig. 4D as the dashed-line fit. The parameters calculated for minimum variance are given in Table 4. It is evident that when the effect of surficial mixing is included in the model to describe the profiles, both ^{137}Cs and ^{210}Pb give consistent measures of the sedimentation rate and mixing depth at this station. The result of applying this model to the radionuclide data obtained at the other stations are also given in Table 4 and indicate that measurements of ^{137}Cs and ^{210}Pb profiles yield essentially the same values for sedimentation rate and mixing depth. The use of ^{137}Cs for dating is considerably more limited than the ^{210}Pb method and can be undertaken only where the sedimentation rate is sufficiently high to be compatible with inherent sampling resolution.

In this simple model the effect of the residence time of the radionuclide within the lake has not been considered. This will be important where the flux to the air-water interface is not constant. We have shown that the averaging effect of the aquatic system on the ^{137}Cs transferred from the atmosphere to the sediments would lead to a very different profile within the sediment core if the residence time in the water column is greater than two years (ROBBINS and EDGINGTON, 1973). Thus, it should be possible on the basis of even a single appropriate core, to determine if the averaging occurs in aquatic system or the sediment. The residence time of ^{137}Cs appears to be several years according to an estimate by WAHLGREN and NELSON (1973) based on the concentrations of this radionuclide in the surface water of Lake Michigan measured over a period of years. Therefore the structure of temporal variations in the flux of ^{137}Cs would be preserved through the time of transfer of the nuclide to sediments, and the assumptions made in the model may be reasonable.

Fluxes at the sediment-water interface

The data presented in Table 4 indicate that the measurements of a ^{210}Pb or a ^{137}Cs profile in a sediment core will provide values of the sedimentation rate and mixing depth which are essentially the same. However, the values of the flux

normalization factor, f [equation (14)], for these two radionuclides are different and the ratio of the two values, $f_{\text{Pb}}/f_{\text{Cs}}$, varies from station to station. In the light of the discussion in a prior section, it is unlikely that the observed spatial variations can be accounted for in terms of incomplete recovery of the uppermost part of the core. Furthermore, this variability is unlikely to be due to the variation in the concentration of atmospheric ^{210}Pb with latitude (JAWOROWSKI, 1966) or differing rainfall patterns, since these small effects are very likely to be averaged out in the water column. Moreover, stations at very comparable latitudes (72-11, 17 and 29) show widely differing values of the excess flux.

The flux at the sediment-water interface tends to be spatially conservative if the radioactivity remains unassociated with the bulk sedimentary material in the water column and therefore arrives independently at the bottom. This would be true also if the sedimentation rate and sediment composition were constant over the whole of the lake. Since the values of the sedimentation rate and the normalization factor, f , are highly variable, it may be concluded that ^{210}Pb and ^{137}Cs are scavenged by settling sediment particles in the water column. Under these conditions the apparent flux of ^{210}Pb and ^{137}Cs at the sediment-water interface would be proportional to the rate of sedimentation and their specific activities (f/ω) should be relatively constant provided that those components of the sedimentary material which complex with or absorb these two nuclides are uniformly distributed in the water column.

The values of the specific activity for ^{210}Pb and ^{137}Cs , given in Table 4, indicate that there are somewhat dissimilar particle associations or histories of transport and deposition of these two nuclides. While there is no systematic variation in the specific activity of ^{137}Cs , the values for ^{210}Pb are consistently lower at the inshore Stations (72-11, 29, 31 and 54 (Table 2) than those found for the offshore Stations (72-17, 100A, 103, 105). This difference may be accounted for in terms of the known properties of the sediments and the geochemistry of lead and cesium in aquatic systems.

Differences in the characteristics of inshore and offshore sediments are evident not only from the porosity profiles (Fig. 2) but also in terms of major changes in chemical composition. For example, there is a twofold decrease in the concentration of calcium and magnesium on going 5-25 miles westward offshore in southern Lake Michigan (ROBBINS and EDGINGTON (1973). In a recent review, LELAND and SHUKLA (1973) concluded that organic matter is more important in the complexation of lead in lake water and sediments than absorption on clays of hydrous oxides. On the other hand, the dominant mechanism for the removal of ^{137}Cs from the water column is ion-exchange with the clay component of sediment and soils (JENNE and WAHLBERG, 1968). Therefore, the spatial distribution of ^{137}Cs and ^{210}Pb in the sediments of Lake Michigan may reflect patterns of deposition of two major sedimentary components, clay and organic carbon respectively. Since organic carbon and $<2 \mu\text{m}$ clay are highly correlated in the sediments of Lake Michigan (SHIMP *et al.*, 1971), the sedimentary material should be separated according to particle size and composition, and the concentration of both radionuclides measured in each fraction.

The inshore values of f/ω observed for ^{210}Pb may be depressed because of dilution of the inshore sediments by terrigenous material which has a low excess concentration

of this nuclide and presumably a low content of organic carbon. The contribution of ^{210}Pb from sources other than direct atmospheric fallout, such as from rivers or erosion, is probably small, as can be seen from an estimate of the mean annual flux of excess ^{210}Pb to the sediments of the southern basin. Since the values of P/ω for the offshore stations are relatively constant and the inshore area of high sedimentation accounts for a small fraction of the area of the southern basin, the average value of the excess flux, \bar{P} , may be estimated from the average value of P/ω at the offshore stations, (19.35 pCi/g), and the average sedimentation rate, $\bar{\omega}$. The average value of the sedimentation rate may be estimated from the average depth of sediment, \bar{L} , deposited in the Waukegan Member over the last 7000 yr (\bar{T}). The value of \bar{L} may be derived from the data given in LINEBACK and GROSS (1971). Since $\bar{L} = 106$ cm and the average porosity of the sediments, ϕ' , is 0.8, the average sedimentation rate is given by equation (3) $\omega = (1 - \phi') \bar{\rho}_s \bar{L}/\bar{T}$ and $\bar{P} = 19.35 \times \bar{\omega} = 0.14$ pCi/cm² yr. This value agrees remarkably well, considering the approximations made, with the value of the atmospheric flux of 0.2 pCi/cm² yr for this latitude (JAWOROWSKI, 1966) and therefore supports the assumption that other sources of ^{210}Pb do not contribute a significant fraction to the total excess concentration found in the sediments.

CONCLUSIONS

^{210}Pb dating appears to be a valid and powerful technique for establishing the age of recently deposited sediments in the Great Lakes. The method gives measures of the sedimentation rate which are essentially in agreement with those derived from more laborious and less exact, but established palynological and stratigraphic methods. ^{210}Pb profiles studies to date show an expected exponential shape consistent with the assumption of uniform sedimentation rates over the last several hundred years and of secular equilibrium between supported ^{210}Pb and ^{226}Ra . Companion ^{137}Cs profiles indicate that the coring technique was probably recovering the uppermost levels of the deposit and that the mobility of both radionuclides within the sediments is small.

More speculative conclusions are based on the limited number of cores analyzed at the present time. It appears that, in general, modern sedimentation rates are not very different from historical ones, although there may be many localities where recent alterations in land use have measurably increased sedimentation rates. The excess ^{210}Pb seems to originate primarily from atmospheric fallout but a further inventory of the ^{210}Pb distribution over the lake bottom must be made to properly assess the significance of other sources. The correlation between ^{137}Cs and ^{210}Pb specific activities from location to location suggests that the mode of transport of both of these radionuclides are comparable involving attachment to settling particles of different composition. Post-depositional redistribution by physical mixing or biological processes can account for the observed low surficial activity of ^{210}Pb at several locations, as well as the appearance of ^{137}Cs at greater depths in the core than would be predicted from the inferred sedimentation rate alone.

The measurement of both ^{210}Pb and ^{137}Cs in sediment cores provided much more information than can be gained from either nuclide alone with regard to understanding the physical and chemical nature of the sedimentation process.

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