

Running Head: Finite Element Modelling of ^{210}Pb Profiles

This manuscript has been submitted to
Environ. Geol. Water Sci.
and the contents are subject to change.

This copy is to provide information
prior to publication.

**ESTIMATION OF MIXING RATES IN WESTERN LAKE
ONTARIO SEDIMENTS BY FINITE ELEMENT ANALYSIS
OF ^{210}Pb PROFILES**

Joshi, S.R.¹ and A.G. Bobba²

NWRI Contribution No. 86-34

¹ Environmental Contaminants Division
² Aquatic Physics and Systems Division
National Water Research Institute
Canada Centre for Inland Waters
Burlington, Ontario, Canada L7R 4A6

ESTIMATION OF MIXING RATES IN WESTERN LAKE ONTARIO SEDIMENTS BY FINITE ELEMENT ANALYSIS OF Pb-210 PROFILES

by

S.R.Joshi and A.G.Bobba

MANAGEMENT PERSPECTIVE/IMPLICATIONS OF RESEARCH

A finite element model is used to generate theoretical excess Pb-210 concentration/depth profiles that are "best-fit approximations" to directly measured profiles at five locations in western Lake Ontario. The results indicate that, while mixing (of old and new sediments) is prevalent at all 5 locations, its influence is severe at locations closer to the mouth of Niagara River. While these results can easily be rationalised in terms of known current mixing in the area, the findings seem to be incompatible with other data. For example, were mixing so dominant as the model indicates, the profiles of all other pollutants in the sediment cores would be severely obliterated. This, however, is not the case. The available profiles for heavy metals, radionuclides and organics are in fact rather well preserved and have been shown to follow the time pattern assigned by techniques which ignore the effect of mixing. Obviously, the so-called "Pb-210 methodology for deriving accurate mixing rates" has some errors. We postulate that two factors can easily give erroneous results: Firstly, the flux of atmospheric Pb-210 is not constant as is generally assumed. The variations in flux can give rise to the "kinks" which are then interpreted as due to "mixing". Secondly, the sedimentation rate may not be constant as is generally assumed.

ESTIMATION DES TAUX DE BRASSAGE DES SÉDIMENTS DANS LA PARTIE OUEST DU LAC ONTARIO AU MOYEN DE L'ANALYSE D'ÉLÉMENTS FINIS DES PROFILS DE Pb^{210}

par

S.R. Joshi et A.G. Bobba

PERSPECTIVE-GESTION/RÉPERCUSSIONS SUR LA RECHERCHE

On se sert d'un modèle à éléments finis pour produire des profils théoriques de concentrations excessives de Pb^{210} par rapport à la profondeur qui se rapprochent le plus des profils mesurés à cinq endroits de la partie ouest du lac Ontario. Les résultats révèlent que, même s'il se produit un brassage (des anciens sédiments avec les nouveaux) aux cinq endroits, son influence est très marquée aux endroits à proximité de l'embouchure de la rivière Niagara. On peut facilement expliquer ces résultats par le brassage que l'on reconnaît au courant dans cette zone; or, les résultats semblent incompatibles avec les autres données. Par exemple, si le brassage était aussi dominant que l'indique le modèle, les profils de tous les autres polluants dans les couches sédimentaires seraient gravement estompés. Pourtant, ce n'est pas le cas. Les profils dont on dispose pour les métaux lourds, les radionucléides et les matières organiques sont en fait bien conservés et suivent le modèle temporel assigné par des techniques qui ne tiennent pas compte de l'effet de brassage. Il est évident que la présumée méthodologie reposant sur le Pb^{210} pour dériver des taux précis de brassage comporte certaines erreurs. Nous avançons ce qui suit : deux facteurs peuvent facilement fausser les résultats. Tout d'abord, le flux de Pb^{210} dans l'atmosphère n'est pas constant, contrairement à ce que l'on a tendance à le croire généralement. Les variations de flux peuvent donner lieu à des écarts qui sont ensuite interprétés comme étant imputables au brassage. De plus, le taux de sédimentation peut ne pas être constant, contrairement à ce que l'on présume habituellement.

ABSTRACT

A finite element model is used to generate theoretical excess ^{210}Pb concentration/depth profiles that are "best-fit approximations" to directly measured profiles at five locations in western Lake Ontario. The "best-fit approximations" are derived using a minimum error analysis technique. The results indicate that, while mixing is prevalent at all the study locations, its influence is severe at locations closer to the mouth of the Niagara River. The non-occurrence of the implicit effects of sediment mixing in measured pollutant profiles at these locations is pointed out and it is postulated that the general assumptions of constant flux of excess ^{210}Pb and constant sedimentation rate in such models lead to unrealistic estimates of mixing.

RÉSUMÉ

On se sert d'un modèle à éléments finis pour produire des profils théoriques de concentrations excessives de Pb^{210} par rapport à la profondeur qui se rapprochent le plus des profils mesurés directement à cinq endroits de la partie ouest du lac Ontario. Ces approximations qui se rapprochent le plus des profils réels sont dérivées à l'aide d'une technique d'analyse des erreurs minimales. Les résultats révèlent que, même s'il se produit un brassage à tous les endroits visés par l'étude, son influence est très marquée à l'embouchure de la rivière Niagara. On signale que les effets implicites du brassage des sédiments dans les profils de polluants mesurés à ces endroits ne se font pas sentir et on avance que les hypothèses générales du flux constant de quantités supérieures de Pb^{210} et du taux de sédimentation constant de tels modèles donnent des prévisions irréalistes du brassage.

Introduction

In the recent past several studies have documented the presence of toxic organic chemicals (Kaiser and others, 1983; Onuska and others, 1983; Durham and Oliver, 1983) and heavy metals (Mudroch, 1983) in the sediments of western Lake Ontario at the mouth of Niagara River. A general lack of independently-derived estimates of sedimentation rates has precluded a complete analysis of pollutant profiles in these studies. The present article reports linear sedimentation rates derived from excess ^{210}Pb (that is total ^{210}Pb in the sediment less that supported by ^{226}Ra) measurements at five locations common to all above investigations. More importantly, since the study area is known to be influenced by strong currents (Murthy, 1969), we have further analyzed the measured excess ^{210}Pb profiles in terms of a finite element model that considers mixing as an integral part of the complex sediment transport process.

Methods

Sediment cores from five locations in western Lake Ontario (Fig. 1) were sectioned at 1-cm intervals and their physical properties (porosity and dry and bulk densities) measured using standard techniques (Durham and Joshi, 1980). The radionuclide concentrations were derived by measuring the 46.5 (^{210}Pb), and 186 and

352 keV (^{226}Ra) gamma-emissions using high-resolution, low-energy gamma-ray spectrometry system described elsewhere (Joshi, 1985a).

Governing Equations and Analysis

In order to develop formulations describing the behaviour of ^{210}Pb -bearing particles in the deposited sediments, certain assumptions are considered to be valid within the boundary conditions represented in Fig. 2. The essential boundary is defined by the sediment/water interface in view of the general realization that sediment pore water is much less susceptible to turbulence than the water overlying the deposited sediment. The assumptions necessary to develop the governing equations have been discussed earlier (Bobba and others, 1982, 1983) and lead to the following equations (Bukata and Bobba, 1984):

$$\frac{\partial C}{\partial t} = \frac{[\theta D_m + \rho_s K(1 - \theta) D_B]}{[\theta + \rho_s K(1 - \theta)]} \cdot \frac{\partial^2 C}{\partial z^2} - S \frac{\partial C}{\partial z} - \lambda C \quad \text{for } 0 \leq z \leq L \quad (1)$$

and

$$\frac{\partial C}{\partial t} = \frac{\theta D_m}{[\theta + \rho_s K(1 - \theta)]} \cdot \frac{\partial^2 C}{\partial z^2} - S \frac{\partial C}{\partial z} - \lambda C \quad \text{for } z > L \quad (2)$$

where C is the concentration of excess ^{210}Pb at depth z and time t , θ is the porosity of the sediment, ρ_s is the bulk density of the sediment, K is the adsorption coefficient, D_m is the molecular diffusion coefficient of ^{210}Pb , D_B is the mixing (current/physical/biological) coefficient, S is the linear sedimentation rate, λ is the radioactive decay constant for ^{210}Pb and L the depth beyond which mixing is thought to cease.

A previously described finite element technique (Bobba and others, 1983) is used to solve equations (1) and (2). Fig. 3 presents the measured and simulated profiles of excess ^{210}Pb in the five sediment cores. In the one-layer model, mixing is considered to extend the entire depth over which measurements were performed and the simulated curve is constructed using equation (1). In the two-layer model, the measured porosity profile defines the boundary between upper and lower layers and the bottom layer is considered to be devoid of mixing. The simulated profile is obtained using equation (1) for $z \leq L$ and equation (2) is applied for the second layer. The values of D_m and D_B used to obtain simulated profiles are selected by considering the minimum error between observed and simulated data. Fig. 4 illustrates this point for station 210 where minimum cumulative error for D_m and D_B is observed at 1.25 and 0.60 $\text{cm}^2 \text{ y}^{-1}$, respectively, for one-layer representation. The values of D_m and D_B given in Table 1 for all other locations were estimated similarly.

Discussion

Application of the total minimum error analysis technique to the comparisons illustrated in Fig. 3 reveals that one-layer model provides a closer fit to the measured excess ^{210}Pb profiles at stations 207, 208 and 209 while two-layer representation fits better the data for stations 206 and 210. This implies that, while mixing affects the deposited sediments at all five locations, its influence is much more significant at stations 207, 208 and 209. This is understandable considering these three stations are closest to the mouth of the Niagara River and are thus more prone to current mixing.

Notwithstanding the superior flexibility of this finite element model in comparison with numerous other prevailing models and easy rationalization of conclusion afforded in the present study in terms of the known physical characteristics of the study area, we are puzzled by the non-observance of the implications of the present study in measured pollutant profiles. For example, the results of the present study indicate that the sediment cores retrieved from stations 207, 208 and 209 are severely affected by mixing throughout the entire length of the core. Under these circumstances one would expect that the depositional records of various pollutants would be very highly obliterated (Goreau, 1977). The available data on the profiles of both organic (Durham and Oliver, 1983) and inorganic (Mudroch, 1983) pollutants, however, clearly show that mixing has had hardly any

effect on the time patterns of deposition of various pollutants at these locations.

In our opinion two modelling-related factors can easily vitiate efforts to reconcile ^{210}Pb -derived parameters with measured pollutant profiles in the study area. Firstly, the general assumption of constant flux of excess ^{210}Pb at the air/water interface and, by inference, at the sediment/water interface may not be valid. Variations in the flux of atmospheric ^{210}Pb have been known to occur even before the radioisotope gained popularity in sedimentation rate studies of the lakes (see, for example, Joshi and others, 1969). The fluctuations are discernible in data from locations as far apart as Greenland (Koide and others, 1982) and Antarctica (Koide and others, 1979). Variations in the concentrations of atmospheric ^{210}Pb are also evident in the limited data available for the Great Lakes region (IJC, 1983; D.J. Bazinet, Ontario Ministry of the Environment, personal communication, 1984; Joshi, 1985b). Such variations obviously can lead to inaccurate estimates of sedimentation rate and may indicate mixing even when it is absent. In reporting a 4.4-fold fluctuation in the annual ^{210}Pb atmospheric flux at Seattle, WA, USA, Nevissi (1985) has also alluded to this possibility.

Secondly, the assumption of constant sedimentation rate may encompass a rather oversimplified view of the study area which is known to have undergone small, but definite, changes in sediment inputs via the Niagara River. In our experience it is easier to eliminate the effects of variable sedimentation rate by evaluating

sedimentation rate data in terms of the constant rate of supply of excess ^{210}Pb model (Krishnaswamy and others, 1971; Oldfield and others, 1978) as against the commonly used constant initial concentration of excess ^{210}Pb model used in the present study. Both approaches, however, assume constant flux of excess ^{210}Pb at the sediment/water interface. Efforts to apply the constant rate of supply model to our data are currently underway. Our preliminary analysis of data indicates that a precise knowledge of the total excess ^{210}Pb in the sediment column is very critical in deriving sedimentation rates using this model and that such information is best obtained only when ^{210}Pb analytical data covering the previous 110 years or so are available.

In order to check the time patterns of pollutant deposition using the ^{210}Pb technique, especially in cases such as the present one where mixing is suspected, it seems equally important that another independent time marker should also be included. Nuclear fallout radionuclides with low molecular diffusion coefficients (such as $^{239,240}\text{Pu}$ and ^{241}Am) constitute such markers as, unlike other inorganic and organic pollutants, their production patterns are quite well defined. We are in the process of measuring the vertical profiles of fallout radionuclides as well and the results of such measurements and interpretations based on constant rate of supply model-derived sedimentation rates and revised mixing parameters will be reported at a later date.

Acknowledgements

The authors thank M.E. Fox for providing the sediment cores, S.P. Thompson for laboratory assistance and A. Mudroch and D.C.L. Lam for useful discussions.

Table 1. Estimated transport parameters for excess ^{210}Pb in western Lake Ontario sediments

Station	Linear Sedimentation rate, S (cm/yr)	Two-Layer model ^b					One-layer model ^b			
		D_{m1}	D_{B1}	D_{m2}	D_{B2}	L cm	D_{m3}	D_{B3}	L cm	
206	0.36	0.50	7.23	3.41	0	6	0.50	2.92	15	
207	1.41	1.45	95	41.2	0	4	1.50	95.2	24	
208	0.81	20.50	332	22.6	0	10	1.25	330	20	
209	0.96	30.00	7500	3506.5	0	8	1.65	7500	24	
210	0.19	0.55	0.65	0.75	0	3	1.25	0.60	10	

aValues derived following Durham and Joshi (1980) and Joshi (1985b).

bSubscripts 1, 2 and 3 refer to upper, lower and extended mixed layers, respectively.

References Cited

- Bobba, A.G., R.P. Bukata, S.R. Joshi, and R.W. Durham, 1982, Numerical simulation and analysis of contaminants in lake sediments: Paper presented at the 11th International Congress on Sedimentology, McMaster University, Hamilton, Canada, August 22-27.
- Bobba, A.G., D.C.L. Lam, and R.P. Bukata, 1983, Simulation of contaminant transport through sediments: Paper presented at the 26th Conference on Great Lakes Research, SUNY, Oswego, U.S.A., May 23-27.
- Bukata, R.P., and A.G. Bobba, 1984, Determination of diffusion coefficients associated with the transport of ^{210}Pb radionuclide in lake bed sediments: Environ. Geol., vol. 5, no. 3, p. 133-141.
- Durham, R.W., and S.R. Joshi, 1980, Recent sedimentation rates, ^{210}Pb fluxes and particle settling velocities in Lake Huron, Laurentian Great Lakes: Chem. Geol., vol. 31, p. 53-66,
- Durham, R.W., and B.G. Oliver, 1983, History of Lake Ontario contamination from the Niagara River by sediment radiodating and chlorinated hydrocarbon analysis: J. Great Lakes Res., vol. 9, no. 2, p. 160-168.
- Goreau, T.J., 1977, Quantitative effects of sediment mixing on stratigraphy and biogeochemistry: a signal theory approach: Nature, vol. 265, p. 525-526.
- International Joint Commission, 1983, 1983 Report on Great Lakes water quality. Appendix on radioactivity: International Joint Commission, Windsor, Canada, p. 27.

- Joshi, L.U., C. Rangarajan, and S. Gopalakrishnan, 1969, Measurement of lead-210 in surface air and precipitation: *Tellus*, vol. 21, p. 107-112.
- Joshi, S.R., 1985a, Determination of Thorium-228, Thorium-230 and Thorium-232 in sediments by anion exchange and nuclear spectrometry: *Anal. Chem.*, vol. 57, no. 6, p. 1023-1026.
- Joshi, S.R., 1985b, Recent sedimentation rates and ^{210}Pb fluxes in Georgian Bay and Lake Huron: *Sci. Total Environ.*, vol. 41, p. 219-233.
- Kaiser, K.L.E., M.E. Comba, and H. Huneault, 1983, Volatile halocarbon contaminants in the Niagara River and in Lake Ontario: *J. Great Lakes Res.*, vol. 9, no. 2, p. 212-223.
- Koide, M., R. Michel, E.D. Goldberg, M.M. Herron, and C.C. Langway, Jr., 1979, Depositional history of artificial radionuclides in the Ross Ice Shelf, Antarctica: *Earth Planet. Sci. Lett.*, vol. 44, p. 205-223.
- Koide, M., R. Michel, E.D. Goldberg, M.M. Herron, and C.C. Langway, Jr., 1982, Characterization of radioactive fallout from pre- and post-moratorium tests to polar ice caps: *Nature*, vol. 296, p. 544-546.
- Krishnaswamy, S., D. Lal, J.M. Martin, and M. Meybeck, Geochronology of lake sediments: *Earth Planet. Sci. Lett.*, vol. 11, p. 407-414.
- Mudroch, A., 1983, Distribution of major elements and metals in sediment cores from the western basin of Lake Ontario: *J. Great Lakes Res.*, vol. 9, no. 2, p. 125-143.

Murthy, C.R., 1969, Large-scale diffusion studies at Niagara River mouth, Lake Ontario: Proc. 12th Conf. Great Lakes Res., p. 635-651.

Nevissi, A.E., 1985, Measurement of ^{210}Pb atmospheric flux in the Pacific Northwest: Health Phys., vol. 48, no. 2, p. 169-174.

Oldfield, F., P.G. Appleby, and R.W. Battarbee, 1978, Alternative ^{210}Pb dating: results from the New Guinea Highlands and Lough Erne: Nature, vol. 27, no. 5643, p. 339-342.

Onuska, F.I., A. Mudroch, and K.A. Terry, 1983, Identification and determination of trace organic substances in sediment cores from the western basin of Lake Ontario: J. Great Lakes Res., vol. 9, no. 2, p. 169-182.

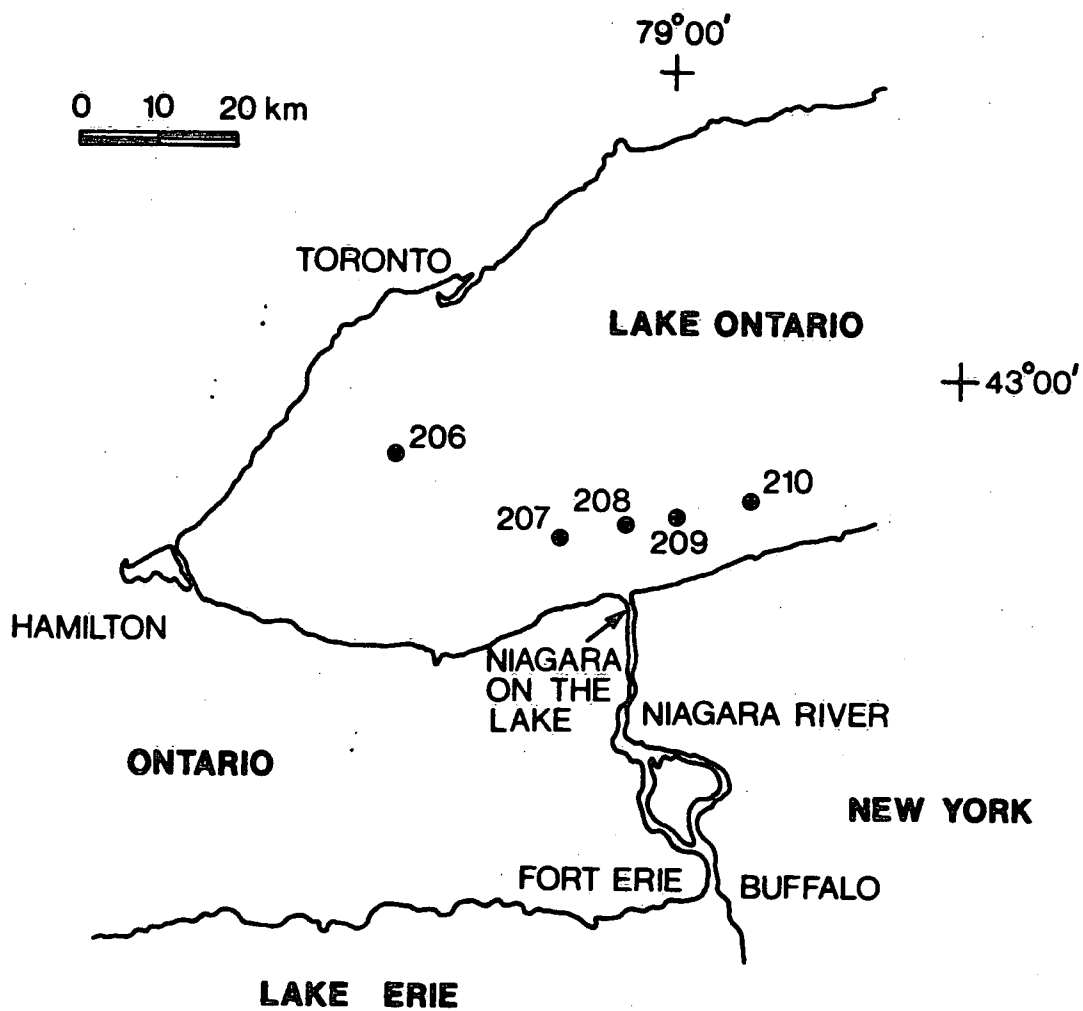
Figure Captions

Figure 1 Locations of sediment core sampling stations in western Lake Ontario

Figure 2 Conceptual representation of the boundary conditions used in the model

Figure 3 Comparative representation of simulated and observed excess ^{210}Pb profiles in sediment cores

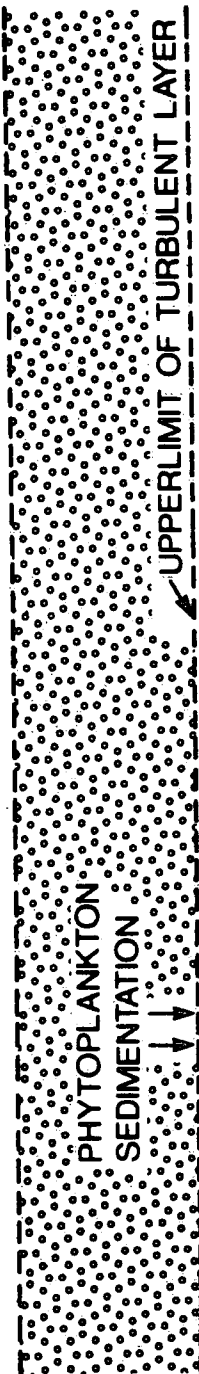
Figure 4 Error analysis of diffusion parameters obtained for station 210



LAKE SURFACE



LAKE WATER

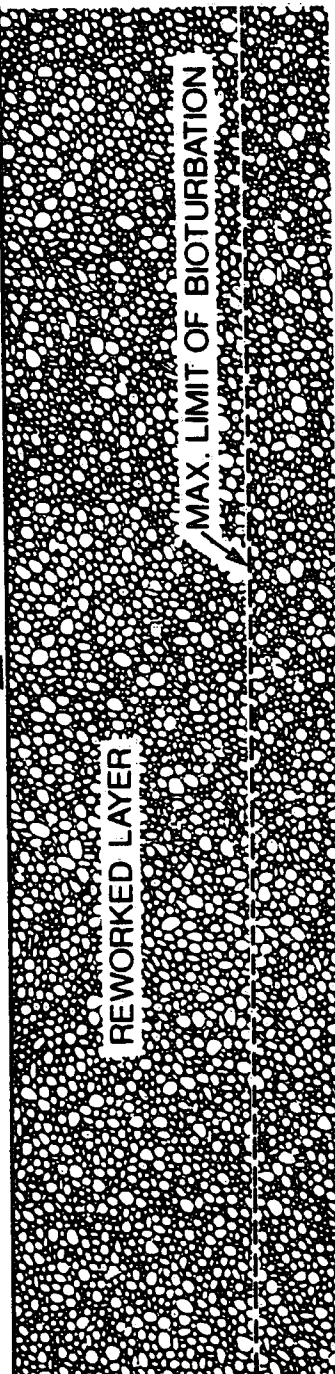


BENTHIC BOUNDARY LAYER

BEDLOAD TRANSPORT →



SEDIMENT SURFACE



SEDIMENT

EXCESS ^{210}Pb (Bq/g dry sediment)

