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LEAD-210 SEDIMENTATION IN LAKE ONTARIO

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NWRI Contribution No. 91-06

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MANAGEMENT PERSPECTIVE

The measurement of depth profile of ^{210}Pb, a naturally-occurring radionuclide with a half-life of 22.3 years, provides a suitable means for assigning dates to sediment core segments and in deriving sedimentation rates. The interpretation of sedimentary ^{210}Pb profile, however, can be complicated by the mixing of old and new sediment. Estimates of mixing in Lake Ontario sediments have been previously derived by an analysis of porosity (water content) profiles. These estimates report much higher mixing than is indicated by the reported pollutant profiles in sediments off the mouth the Niagara River.

In the present study we suggest that realistic mixing estimates are obtained when sedimentary ^{210}Pb, rather than porosity, profiles are considered. We also report the first estimates of mixing in any aquatic system assuming both mixing and sedimentation occur simultaneously. (Previously mixing has been assumed to occur in the absence of sedimentation.) The presently derived estimates suggest very little mixing in Lake Ontario sediments in agreement with the reported pollutant profiles.

This work has been extended to obtain a first estimate of the Lake Ontario area over which the ^{210}Pb-bearing particles are depostied. The results indicate that the particles are spread over an area of about 441 km^2 (range 220-938 km^2). The calculation assumes that the sedimentation rate remains unchanged throughout this area. In practice, however, lower sedimentation rates prevail as one goes farther away from the Niagara River mouth. This estimate, therefore, represents the minimum area over which the Niagara River-supplied sediment is spread in Lake Ontario.

A more significant aspect of calculations performed during this investigation is that the annual Niagara River suspended sediment load is found to be about 1.8 million tonnes. This value is lower by a factor of about 2.5 than that (4.6 mt) used commonly. This observation should be further validated from ongoing WQB-OR measurements. The obvious implication of this new estimate is that both sediment and pollutant loadings to Lake Ontario need downward revision.

PERSPECTIVES DE LA DIRECTION

La détermination du profil de concentrations de ^{210}Pb, un nucléide naturel d'une demi-vie de 22,3 années, constitue un moyen approprié de dater des portions de carottes de sédiments et de mesurer les taux de sédimentation. Toutefois, l'interprétation des profils de concentrations de ^{210}Pb dans les sédiments peut être complexe en raison du brassage des sédiments anciens et nouveaux. Des estimations du brassage des sédiments dans le lac Ontario ont été obtenues antérieurement au moyen d'une analyse des courbes (profils) de porosité (teneur en eau). Ces estimations sont beaucoup plus élevées que les valeurs de brassage obtenues au moyen des profils de concentrations des matières polluantes dans les sédiments tout près de l'embouchure de la rivière Niagara.

Dans la présente étude, on soutient que l'on peut obtenir des estimations réalistes du brassage à l'aide du profil des concentrations de ^{210}Pb dans les sédiments plutôt qu'avec les courbes de porosité. Le document fournit également les premières estimations de brassage dans tout système aquatique en supposant au départ que le brassage et la sédimentation s'effectuent de façon simultanée (antérieurement, on présumait que le brassage et la sédimentation ne se produisaient pas en même temps). Les estimations présentées dans ce rapport révèlent qu'il y a très peu de brassage des sédiments dans le lac Ontario, ce qui se conforme aux profils des concentrations de polluants signalées.

On a étendu la portée de l'étude afin de faire la première estimation de la superficie sur laquelle des particules contenant du ^{210}Pb se déposent dans le lac Ontario. D'après les résultats, les particules sont dispersées sur une superficie d'environ 441 km^2 (gamme de 220 à 238 km^2). Aux fins des calculs, on présume que le taux de sédimentation est identique sur toute cette superficie. Cependant, en réalité, les taux de sédimentation diminuent au fur et à mesure qu'on s'éloigne de l'embouchure de la rivière Niagara. En conséquence, cette estimation représente la superficie minimale sur laquelle les sédiments provenant de la rivière Niagara s'étendent dans le lac Ontario.

Un aspect plus significatif des calculs effectués au cours de cette étude est la charge annuelle de sédiments en suspension dans la rivière Niagara, qui atteint environ 1,8 million de tonnes. Cette valeur est inférieure d'un facteur d'environ 2,5 à la charge habituellement employée (4,6 tonnes métriques). Les mesures effectuées en permanence par RO-DQE devraient permettre de vérifier avec exactitude cette observation. De toute évidence, cette nouvelle estimation révèle que tant les charges de sédiments que les charges de polluants dans le lac Ontario doivent être rectifiées à la baisse.

ABSTRACT

The sedimentation rates and diffusive sediment mixing coefficients at several Lake Ontario locations have been derived from measurements of unsupported ^{210}Pb profiles in sediment cores. The values of mixing coefficients obtained in the present study are significantly lower than those obtained previously through an analysis of porosity profiles. The present estimates, however, are consistent with the rather well-preserved pollutant profiles at some of these locations. It is observed that the more realistic value of the mixing coefficient, obtained by inclusion of the sedimentation rate parameter, follows the sign opposite to that for the constant obtained by regression analysis of the porosity data. Further work is required to delineate this apparent relationship between two important physical characteristics of deposited sediments.

Analysis of available suspended sediment data shows that Niagara River supplies about 1.8 million tonnes of sediment annually to Lake Ontario. This value is significantly lower than that (4.6 mt yr^{-1}) used previously in constructing sediment and pollutant budgets for Lake Ontario. From the presently derived sedimentation rate and suspended solid discharge estimates, an average value of 441 km^2 (range 220-938 km^2) is obtained for the minimum area of Lake Ontario over which the Niagara River-supplied fine sediment is deposited.

RÉSUMÉ

Les taux de sédimentation et les coefficients de brassage des sédiments diffus ont été calculés à plusieurs endroits dans le lac Ontario au moyen des profils de concentrations de ^{210}Pb non vérifiés dans des carottes de sédiments. Les coefficients de brassage obtenus au cours de la présente étude sont nettement inférieurs à ceux calculés antérieurement par l'analyse des courbes de porosité. Toutefois, les présentes estimations se conforment aux profils de concentrations de polluants, qui sont plutôt bien préservés à certains endroits. On observe que la valeur plus réaliste du coefficient de brassage, obtenue en incluant le taux de sédimentation, a un signe opposé à celui de la constante obtenue au moyen de l'analyse de régression des données de porosité. Il faudrait poursuivre les travaux afin de caractériser cette relation apparente entre deux caractéristiques physiques d'importance des sédiments déposés.

L'analyse des données accessibles portant sur les sédiments en suspension révèle que la rivière Niagara fournit annuellement environ 1,8 millions de tonnes de sédiments au lac Ontario. Cette valeur est notablement moins élevée que celle employée antérieurement (4,6 t.m. an^{-1}) pour établir les bilans de sédimentation et des matières polluantes dans le lac Ontario. D'après estimations du taux de sédimentation et des charges de matières solides en suspension obtenus au cours de cette étude, on calcule que la superficie minimale sur lesquelles se déposent les sédiments fins de la rivière Niagara dans le lac Ontario est d'une valeur moyenne de 441 km^2 (plage de 220 à 938 km^2).

Introduction

Recently, Joshi and Bobba (1989) have reported on the unsupported ^{210}Pb (that is total ^{210}Pb in the sediment less that supported by ^{226}Ra) profiles in five sediment cores from western Lake Ontario. A finite element model that considers mixing as an integral part of the sediment transport process was used to derive estimates of sediment mixing at these locations. The analysis showed the presence of severe mixing at locations closer to the mouth of the Niagara River. The virtual non-occurrence of the implicit effects of mixing in reported (Durham and Oliver 1983; Joshi 1988a and 1988b) pollutant profiles at these locations was pointed out and it was postulated that unrealistic estimates of mixing arise from the general assumptions of constant flux of unsupported ^{210}Pb at the sediment/ water interface and constant sedimentation rate in such models.

Additionally, it may be suggested that this particular approach itself is of limited use in estimating mixing rates as it does not adequately account for the known behaviour of ^{210}Pb in the aquatic environment. Specifically, this model (Bukata and Bobba 1984) derives mixing parametrs using both the measured porosity and solid-phase unsupported ^{210}Pb profiles in a sediment core. While the water content is an integral physical property of the system, its association with unsupported ^{210}Pb is only marginal since this radionuclide is largely associated with the sedimenting solids (Durham and Joshi 1980; Van Hoof and Andren 1989). In this respect, the conventional approach is more realistic in that it relies only on the measured solids phase depth profile of unsupported ^{210}Pb.

The present communication thus re-examines our previously reported (Joshi and Bobba 1989) data in terms of the conventional approach assuming mixing is constant throughout the sediment core. Data for six additional sites have also been included. All the eleven cores are also assigned dates using two models including one based on the assumption of a variable sedimentation rate. And, finally, we report the first estimates of sediment deposition areas in Lake Ontario using recently available data on the suspended solid concentrations in the Niagara River.

Methods

Sediment cores were retrieved during 1981 and 1982 (Table 1) from locations shown in Figure 1. The unsupported ^{210}Pb activities and physical parameters such as density, porosity and sample thickness, etc., were derived using methods described earlier (Durham and Joshi 1980; Joshi 1987; Joshi 1989). The measured ^{210}Pb activities were decay-corrected to the time of sediment core collection.

Results

Mixing Coefficients

Following Krishnaswami and Lal (1978), three major physical processes, viz., radioactive decay, sedimentation and sediment mixing, are expected to influence the concentration-depth profile of unsupported ^{210}Pb in a sediment core. If sedimentary particle mixing is considered as a diffusive process, the time variation in the concentration of unsupported ^{210}Pb is given by the diagenetic equation

$$\frac{\partial}{\partial x}\left(K\frac{\partial}{\partial x}\rho C\right) - S\frac{\partial}{\partial x}(\rho C) - \lambda\rho C = \frac{\partial}{\partial t}(\rho C) \qquad (1)$$

where K is the mixing or diffusion coefficient ($cm^2\ yr^{-1}$), ρ is the <u>in situ</u> density of the sediment ($g\ cm^{-3}$), C is the concentration of unsupported ^{210}Pb ($Bq\ g^{-1}$) at time t and depth x below the sediment/water interface, S is the linear sedimentation rate ($cm\ yr^{-1}$), and λ the decay constant of ^{210}Pb ($0.0311\ yr^{-1}$). Assuming steady-state conditions, i.e., $\frac{\partial C}{\partial t} = 0$, and K, S and ρ to be constant with time and depth, equation (1) can be rewritten as

$$K\frac{\partial^2 C}{\partial x} - S\frac{\partial C}{\partial x} - \lambda C = 0 \qquad (2)$$

Three solutions of this differential equation can be written (Krishnaswami and Lal 1978) with the general boundary condition $C(x) = C(0)$ for $x = 0$ and $C(x) = 0$ for $x \to \infty$ and by imposing three specific conditions as follows

<u>Case I</u> (K = 0)

The solution of equation (2) is then given by

$$C(x) = C(0) \cdot \exp\left(-\frac{\lambda}{S} \cdot x\right) \qquad (3)$$

This formulation assumes a constant initial concentration (CIC) of unsupported ^{210}Pb at the sediment/water interface and is referred to as the CIC model for deriving sedimentation rate, S. The values of S given in Table 2 have been corrected for the compressive effects of depositing sediments as described earlier (Durham and Joshi 1980; Joshi 1985). This correction, in essence, eliminates the assumption of constant ρ in deriving equation (2). Table 2 also gives our revised estimates of the sedimentation rate at stations 207, 208 and 209.

Case II (S = 0)

The solution of equation (2) is then given by

$$C(x) = C^{+}(0) \cdot \exp\left[\left(-\frac{\lambda}{K}\right)^{1/2} \cdot x\right] \quad (4)$$

This formulation is commonly employed to report the value of the diffusion coefficient. This diffusion coefficient is designated as K in Table 2.

Case III (K ≠ 0, S ≠ 0)

The solution of equation (2) is then given by

$$C(x) = C^{*}(0) \circ \exp(\alpha \circ x) \quad (5)$$

where $$\alpha = \frac{S - (S^2 + 4K\lambda)^{1/2}}{2K} \tag{6}$$

or $$K = \frac{(\lambda + \alpha \cdot S)}{\alpha^2} \tag{7}$$

This diffusion coefficient is obtained by using the value of S given by equation (3) and is denoted as K' in Table 2.

The above formulations assume that diffusion is present throughout the sediment core.

Sediment Core Section Age

Our methodology for estimating sediment core section age has been fully described earlier (Joshi 1985; Joshi and others 1988). Briefly, the CIC age t (in years) of a core section is derived following equation (3), i.e., $t = \frac{x}{S}$, using compaction-corrected values of surface sedimentation rate (Joshi 1985). Following the assumption of a constant rate of supply (CRS) of unsupported ^{210}Pb to sediment, the age t' (in years) of a sediment core segment at depth x below the sediment/water interface is given by

$$t' = \frac{1}{\lambda} \ln \frac{A(\infty)}{A(x)} \tag{8}$$

where $A(\infty)$ represents the total unsupported ^{210}Pb (Bq cm^{-2}) in the sediment column and A(x) that beneath sediment of age t' (Joshi and others 1988).

Area of Sediment Deposition Zone

The area over which the Niagara River-supplied sediment is deposited can be estimated as

$$\text{Deposition Area} = \frac{\overline{La}}{\omega} \qquad (9)$$

where $\overline{La}$ (mg yr^{-1}) denotes the mean annual suspended sediment discharged by the Niagara River and ω (mg cm^{-2} yr^{-1}) is the mass sedimentation rate. $\overline{La}$ was calculated from the Niagara River discharge data (Environment Canada 1985), DD (m^3 sec^{-1}), and the limited available (U.S. Geological Survey 1975-78; K. Kuntz, personal communication 1987) suspended solid, SS (mg L^{-1}), measurements over n years using the relation

$$\overline{La} = \frac{3.1536 \times 10^{10}}{n} \sum_{i=1}^{n} DD_i \; SS_i \qquad (10)$$

A value of about 1.8×10^{15} mg yr^{-1} is obtained for $\overline{La}$ from the data given in Table 3 and is used in deriving estimates of depositional areas given in Table 4. It should be noted that the 1975-78 suspended solid concentrations reported by the U.S. Geological Survey (Table 3) are mean values from six to eight measurements over a year at the Fort Niagara location while the remaining concentrations are mean values of weekly or biweekly measurements over a year at Niagara-on-the-Lake. The values of ω given in Table 4 were obtained following the CIC model in which it is assumed that C(0) in equation (3) is constant and is given by $C(0) = \frac{P}{\omega}$ where P is the flux of unsupported ^{210}Pb (Bq cm^{-2} yr^{-1}) at the sediment/water interface. This definition of the CIC model also assumes that both P and ω are constant.

Discussion

The results given in Table 2 show that the mixing (diffusion) coefficeints obtained on the basis of unsupported ^{210}Pb depth profiles are significantly different from those derived by examination of porosity profiles (Joshi and Bobba 1989). In particular, the values of K' appear to be consistent with the reported organic (Durham and Oliver 1983) and radioactive (Joshi 1988a and 1988b) pollutant profiles which seem to be quite undisturbed and devoid of any significant mixing. Table 2 also lists the derived values of β, a constant in the expression

$$\phi(x) = \phi(o) \cdot e^{-\beta x} \qquad (11)$$

where $\phi(x)$ and $\phi(o)$ denote the sediment porosity at depth x = x and x = 0, respectively (Durham and Joshi 1980). As expected, the measured porosity profiles yield positive values of β, except for stations C and 208. A negative value of β would imply that contrary to expectation, the sediment porosity increases with depth in the sediment core. On the other hand, since all diffusive-type models also yield positive values for the diffusion coefficient, it would be expected that β, K and K' would yield values of the same sign. This is however not borne out by the data given in Table 2 where one finds that although all values of K are indeed of a positive sign, those for K' and β assume opposite signs. Further work is required to decipher an apparent relationship between these two sensitive parameters. In particular, the assumptions of constant K and S in solving equation (1) and in deriving other relevant equations may be questionable.

The CRS model, on the other hand, relies on the assumptions of variable S and ρ (Joshi and others 1988), but is unable to give an estimate of the diffusion coefficient. The age-depth relations in the two models thus essentially differ in that the CIC ages assume a constant S while the CRS ages assume a variable S since fluctuations in ρ in both models have been accounted for and mixing is considered to have a similar impact on both ages. In the present study, both the CIC and CRS models give similar estimates of sediment segment age in several cases. In others, the age estimates tend to differ with an increase in depth in sediment core. We have earlier (Joshi and Shukla 1990) shown that the discrepancy between the two ages arises from the mathematical treatment of analytical data and that this discrepancy is estimable by the relation

$$t' - t = \frac{1}{\lambda} \ln \left[\frac{P_{CIC}}{P_{CRS}}\right] \qquad (12)$$

where P_{CIC} and P_{CRS} denote the fluxes of unsupported ^{210}Pb at the sediment/water interface obtained with the assumptions of the CIC and CRS models, respectively. Our estimates of age discrepancy, derived by using equation (12) are given in Table 5. Our preference for a particular model-derived age in cases where significant discrepancy exists must await the development of relevant expressions for error analysis of measurements.

The data given in Table 5 also shows that significantly lower values of P are obtained for stations 93 and 210 when compared with other locations in the Niagara River area. The values for these two locations also fall somewhat short of an independent estimate (Joshi 1985) of the direct atmospheric flux of unsupported ^{210}Pb (0.025 Bq cm^{-2} yr^{-1}) in the Great Lakes region. This observation may be taken to suggest that these two sites are not significantly impacted by the Niagara River plume. Therefore, these two stations have not been included in obtaining preliminary estimates of the Niagara River sediment deposition zone (Table 4). Stations 64, 403, 14 and 206 have also been excluded from such calculations as they are far removed from the mouth of the Niagara River. The estimated areas of the Niagara River sediment deposition zone given in Table 4 are clearly sediment core-dependent, as follows from equation (9). The areas range between 220 and 938 km^2 with an average of about 441 km^2. Undoubtedly, some fraction of the sediment is carried farther off by the prevailing currents. Therefore, the present estimate will need further refinement when more data on the sedimentation rates at the shelf locations become available.

It should be noted that the above estimates are based on an annual suspended solid load of 1.8 x 10^6 tonnes yr^{-1}, a value considerably lower than a previous estimate of 4.6 x 10^6 tonnes yr^{-1} (International Joint Commission 1969; Kemp and Harper 1976). Ongoing Environment Canada suspended sediment measurements at Niagara-on-the-Lake confirm (K. Kuntz, personal communication 1990) that the lower value used in the present study is more representative of long-term trends. The previous estimate appears to have been based on some measurements during

1966-67 only (International Joint Commission 1969). The data given in Table 3 clearly suggest that annual mean suspended solid concentrations in the Niagara River can vary by a factor of about 2.5, a value similar to the ratio (2.6) of the previous and present annual loading estimates. An obvious corollary to this observation provides that both sediment and pollutant loads to Lake Ontario via the Niagara River need re-examination and possible downward revision.

Also noteworthy is the fact that the mass sedimentation rates in the present study are derived from ^{210}Pb measurements which assume a strong affinity of this radionuclide for sedimenting particles (Durham and Joshi 1980). Many of the known pollutants are known to exhibit preferential association with finer sedimentary particles. Recent studies by Van Hoof and Andren (1989) on the partitioning of ^{210}Pb in Lake Michigan dissolved and size-fractionated particulate phases show that nearly all the particulate ^{210}Pb is contained in the 70-0.45 μm fraction with the vast majority being in the finer ($<$21 μm) particles. With the reasonable assumption that a similar situation prevails in Lake Ontario, and keeping the similarity between ^{210}Pb and numerous chemical pollutants in perspective, it may be suggested that much of the Niagara River-supplied fine sediment and the associated pollutants are largely deposited in the inferred zone.

Acknowledgements

The authors thank the National Water Research Institute's Research Support Division staff and the crew of the

CSS Limnos for obtaining the sediment cores. S.P. Thompson, E.A. Kokotich and J.A. FitzGerald are thanked for sample preparation and analyses. K. Kuntz (Water Quality Branch, Ontario Region) kindly provided unpublished data on suspended solids in the Niagara River.

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Table 1. Sediment core sampling dates and locations in Lake Ontario

Station	Location		Collection Date
	Lat. (° ' "N)	Long. (° ' "N)	
Rochester Basin			
64	43 31 34	76 55 49	July 26, 1982
Mississauga Basin			
403	43 36 05	78 14 14	July 29, 1982
Niagara Basin			
14	43 23 35	79 29 13	July 19, 1982
206	43 24 11	79 27 46	September 7, 1982
Niagara River Area (Inshore Zone)			
93	43 19 39	78 52 04	April 24, 1981
C	43 20 39	78 59 25	April 24, 1981
D3	43 20 09	79 04 15	August 19, 1981
210	43 21 53	78 51 15	September 8, 1982
209	43 20 42	78 59 30	September 8, 1982
208	43 20 14	79 03 51	September 9, 1982
207	43 19 15	79 09 04	September 7, 1982

Table 2. Derived linear sedimentation rates and diffusion coefficients

Station	Linear Sedimentation Rate S (cm yr^{-1})	Diffusion Coefficient (cm^2 yr^{-1}) K	K'	β (cm^{-1})
Rochester Basin				
64	0.45	6.50	-1.47	2.2×10^{-3}
Mississauga Basin				
403	0.17	0.97	-0.21	4.6×10^{-3}
Niagara Basin				
14	0.46	6.92	-1.40	2.0×10^{-3}
206	0.36	4.10	-0.67	2.8×10^{-3}
Niagara River Area (Inshore Zone)				
93	0.16	0.81	-0.17	1.8×10^{-2}
C	1.74	97.20	2.20	-4.6×10^{-4}
D3	0.55	9.65	-2.00	4.5×10^{-3}
210	0.19	1.13	-0.22	1.2×10^{-2}
209	1.53	75.05	-5.60	1.2×10^{-3}
208	1.67	88.96	4.81	-9.2×10^{-4}
207	1.04	34.47	-4.17	2.3×10^{-3}

Table 3. **Niagara River discharge and suspended solid data**

Year	Mean Suspended Solid Concentration[a] (mg L^{-1})	Mean Discharge[b] (m^3 sec^{-1})
1974-75	7.9	6890
1975-76	14.0	6760
1976-77	6.4	6720
1977-78	5.5	6100
1979	8.6	6300
1980	9.4	6570
1981	6.9	6260
1982	10.7	6290

[a] Values for the period 1974-78 were computed from the data reported by the U.S. Geological Survey (1975-78); values for the period 1979-82 were provided by K. Kuntz (Environment Canada, Burlington).

[b] Values obtained from Environment Canada (1985).

Table 4. **Mass sedimentation rates and estimated sediment deposition areas in Lake Ontario**

Station	Mass Sedimentation Rate ω (mg cm^{-2} yr^{-1})	Sediment Deposition Area (km^2)
Rochester Basin		
64	103	NA[a]
Mississauga Basin		
403	40	NA
Niagara Basin		
14	121	NA
206	72	NA
Niagara River Area (Inshore Zone)		
93	66	NA
C	817	220
D3	192	938
210	76	NA
209	626	288
208	810	222
207	334	539

[a] NA, not applicable (see text).

Table 5. Estimates of flux of unsupported ^{210}Pb at the sediment/water interface and age discrepancy afforded by the CIC and CRS models

Station	P ($Bq\ cm^{-2}\ yr^{-1}$) CIC	CRS	Predicted Age Discrepancy t'-t (years)
Rochester Basin			
64	0.069	0.071	-0.9
Mississauga Basin			
403	0.043	0.043	-
Niagara Basin			
14	0.057	0.057	-
206	0.070	0.074	-1.8
Niagara River Area (Inshore Zone)			
93	0.017	0.017	-
C	0.178	0.149	5.7
D3	0.057	0.059	-1.1
210	0.021	0.021	-
209	0.181	0.201	-3.4
208	0.210	0.171	6.6
207	0.120	0.140	-4.9

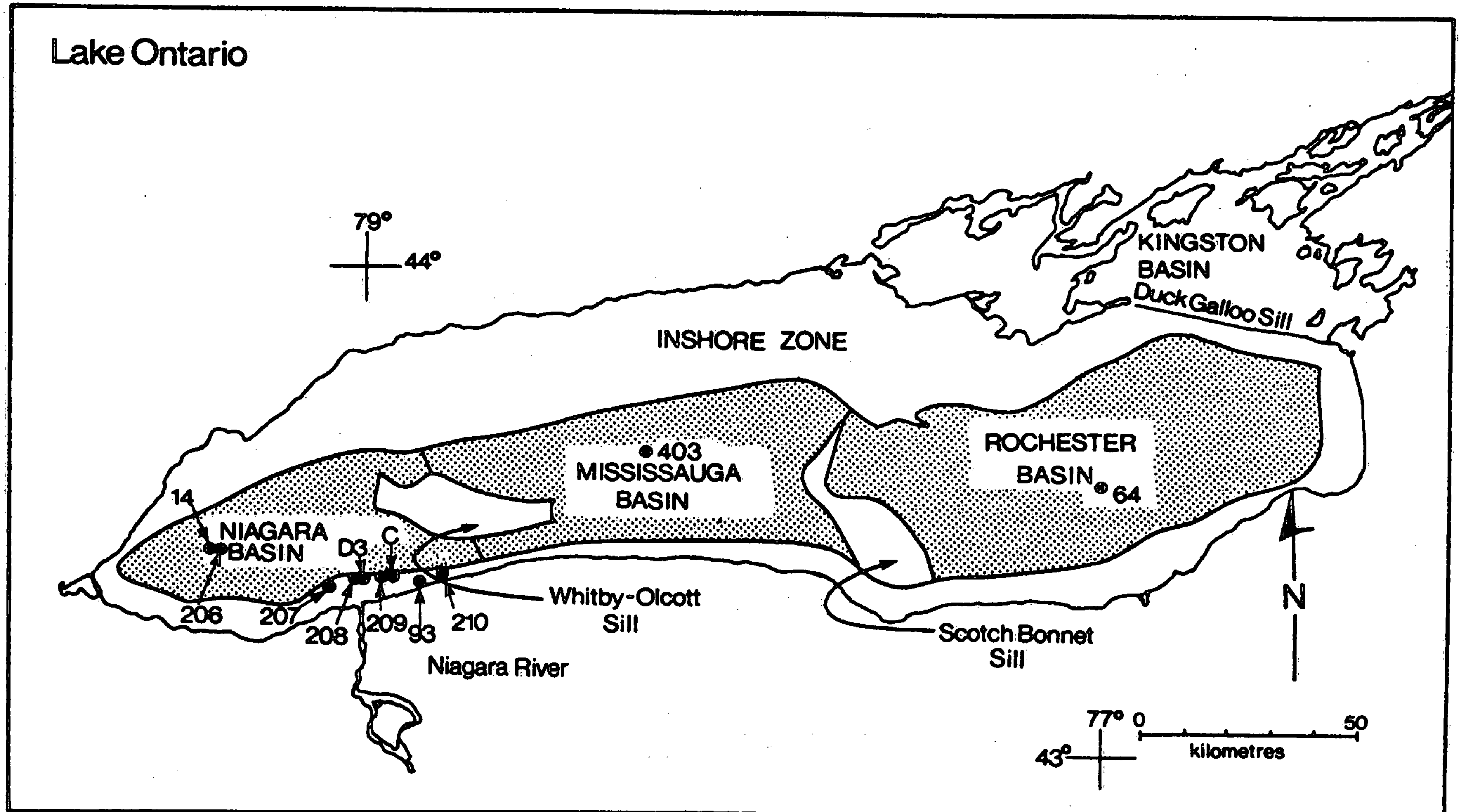

Figure 1. Sediment core samplings in Lake Ontario.

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