PRELIMINARY INVESTIGATION OF TOXIC CHEMICALS IN THE SEDIMENTS OF GREAT SLAVE LAKE, NORTHWEST TERRITORIES, CANADA

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ABSTRACT

A preliminary investigation was carried out in the summer 1987 to determine the geochemistry and distribution of trace elements and organic contaminants in sediments from selected areas in Great Slave Lake, Northwest Territories, Canada. The objective of the investigation was to assess the potential of the lake as a sink for contaminants and further transport of sediment-associated contaminants into the Mackenzie River. Sediment dating indicated a very high sedimentation rate (46.5 g·cm⁻² per year) at a 110-m water depth in the vicinity of the Slave River mouth. The geochemical composition of the sediment showed the deposition of similar material at all sampling stations. The concentrations of trace elements (Cu, Ni, Co, Cr, V, Pb and Zn) were uniform in sediment profiles. surficial sediments were enriched by arsenic. The concentrations of PCB's in collected sediments (1.46 to 22.76 ng·g⁻¹) were about one to two orders of magnitude lower than those found in Lake Ontario. However, the distribution pattern of PCB's showed a greater content of lower chlorinated biphenyls in sediments from Great Slave Lake than in Lake Ontario. The concentrations οf other chlorinated hydrocarbons (0.01

1.00 ng·g⁻¹)were up to two orders of magnitude lower in Great Slave Lake sediments than from those in Lake Ontario. However, concentrations of several compounds, particularly chlordane and hexachlorobenzene, were considerably high and uniform in sediment profiles. These results are in agreement with the findings and suggestions of other researchers who have studied the distribution of chlorinated hydrocarbons in the sub-arctic and arctic ecosystem.

KEY WORDS: Great Slave Lake, sediments, geochemistry, organochlorine contaminants.

RÉSUMÉ

Durant l'été de 1987, on a entrepris une recherche préliminaire pour déterminer la géochimie et la distribution des oligo-éléments et des contaminants organiques dans les sédiments de certaines régions du Grand Lac de l'Esclave. Territoires-du-Nord-Ouest, au Canada. Cette étude visait évaluer dans quelle mesure ce lac pouvait servir de lieu d'évacuation des contaminants et de relais pour le transport des contaminants associés aux sédiments dans le fleuve Mackenzie. En procédant à la datation des sédiments, on a obtenu un taux de sédimentation très élevé (46,5 g.cm⁻² par an) à une profondeur d'eau de 110 m au voisinage de l'embouchure de la rivière de l'Esclave. D'après la composition géochimique des sédiments, les matériaux alluvionnaires étaient similaires dans toutes les stations d'échantillonnage. Les concentrations d'oligo-éléments (Cu, Ni, Co, Cr, V, Pb, et Zn) étaient uniformes dans les profils sédimentaires. Cepéndant, les sédiments superficiels étaient enrichis par de l'arsenic. Les concentrations de BPC dans les sédiments receuillis (1,46 à 22,76 ng.g-1) étaient d'environ un ou deux ordres de grandeur inférieures à celles observées dans le Toutefois, la distribution des BPC montrait que les lac Ontario. sédiments du Grand Lac de l'Esclave contenaient plus de biphényles plus faiblement chlorés que ceux du lac Ontario. concentrations d'autres hydrocarbures chlorurés (0,01 à ng.g⁻¹) dans les sédiments du Grand Lac de l'Esclave étaient inférieures de deux ordres de grandeur au maximum par rapport aux sédiments du lac Ontario. teneurs des Cependant, les concentrations de plusieurs composés, en particulier le chlordane et l'hexachlorobenzène étaient considérablement élevées uniformes dans les profils sédimentaires. résultats Ces concordent avec les conclusions et suggestions chercheurs qui ont étudié la distribution des hydrocarbures chlorurés dans l'écosystème subarctique et arctique.

MOTS-CLÉS: Grand Lac de l'esclave, sédiments, géochimie, contaminants organochlorés.

EXECUTIVE SUMMARY

A preliminary investigation was carried out in the summer 1987 to determine the geochemistry and distribution of trace elements and organic contaminants in sediments from selected areas in Great Slave Lake, NWT. The objective of the investigation was to assess the potential of the lake's sediments for transport of contaminants into the Mackenzie River.

Surface sediment samples and cores were collected at three stations in the lake. Selected sections of the cores were used for chemical analyses. One core from the deepest part of the lake's west basin was dated by Pb-210 and Cs-137.

Echo sounding of the bottom showed a steep slope at the mouth of the major tributary, the Slave River. This slope extended from about 20 to 110 m water depth towards the deepest point in the lake's west basin. A visual examination of sediment cores indicated mixing and possible turbidity currents at this area.

Sediment dating indicated a very high sedimentation rate (46.5 $g/cm^2/year$), and/or intensive mixing of the sediment at a 110-m water depth in the vicinity of the Slave River mouth.

The geochemical composition of the sediment showed the deposition of similar material at all sampling stations. The concentrations of trace elements (Cu, Ni, Co, Cr, V, Pb and Zn) were uniform in sediment profiles with the exception of arsenic. The enrichment of arsenic in surficial sediments at two stations indicated a recent input of this element into the lake, most likely from the emission or wastes from gold mining operations on the north shore of the lake.

The concentrations of PCB's in the sediments were about one to two orders of magnitude lower than those found in Lake Ontario. However, the distribution pattern of PCB's showed a greater content of lower chlorinated biphenyls in sediments from Great Slave Lake than in Lake Ontario. More studies need to be carried out to identify if greater concentrations of lower chlorinated biphenyls in sub-surficial sediments originated from environmentally induced dechlorination processes of PCB's (bacterial degradation, photolysis) or from different sources (riverine and atmospheric input).

The concentrations of other chlorinated hydrocarbons were up to two orders of magnitude lower in Great Slave Lake sediments than from those in Lake Ontario. However, concentrations of several compounds, particularly chlordane and hexachlorobenzene, were considerably high and uniform in sediment

profiles. These results are in agreement with the findings and suggestions of other researchers who have studied the distribution of chlorinated hydrocarbons in the sub-arctic and arctic ecosystem.

It was concluded that Great Slave Lake sediments are a potential source of organic contaminants for transport into the Mackenzie River, and that further comprehensive investigation of the distribution of organic contaminants in suspended and bottom sediments from the Slave River-Great Slave Lake-Mackenzie River system would enhance present limited knowledge on the contribution of organics to Canadian sub-arctic and arctic regions from different sources, and transport and global cycling of these persistent toxic contaminants.

RÉSUMÉ DE L'ÉTUDE

Durant l'été de 1987, on a entrepris une recherche préliminaire pour déterminer la géochimie et la distribution des oligo-éléments et des contaminants organiques dans les sédiments de certaines régions du Grand Lac de l'Esclave, dans les Territoires-du-Nord-Ouest, au Canada. Cette étude visait à évaluer dans quelle mesure les sédiments du lac pouvaient servir à transporter les contaminants dans le fleuve Mackenzie.

Dans trois stations établies dans le lac, on a prélevé des échantillons et carottes de sédiments superficiels. On a soumis à des analyses chimiques certaines sections des carottes. On a pu dater l'une des carottes extraites de la partie la plus profonde du bassin occidental du lac, et ce à l'aide du Pb-210 et du Cs-137.

Un sondage acoustique du fond a permis de repérer une pente abrupte à l'embouchure du principal affluent, la rivière de l'Esclave. Cette pente s'étendait de 20 m environ à 110 m de profondeur en direction du point le plus profond dans le bassin occidental du lac. Un examen visuel des carottes sédimentaires révélait qu'il y avait dans cette zone un brassage d'eau et peut-être des courants de turbidité.

La datation des sédiments a permis de constater que le taux de sédimentation était très élevé (46,5 g/cm²/an), et/où qu'il y avait un brassage intensif des sédiments à une profondeur d'eau de $110\,$ m au voisinage de l'embouchure de la rivière de l'Esclave.

D'après la composition géochimique des sédiments, les matériaux alluvionnaires étaient similaires dans toutes les stations d'échantillonnage. Les concentrations d'oligo-éléments (Cu, Ni, Co, Cr, V, Pb, et Zn) étaient uniformes dans les profils sédimentaires, à l'exception de l'arsenic. L'enrichissement des sédiments superficiels par de l'arsenic dans deux stations indique qu'il y a eu récemment un apport de cet élément dans le lac; il provient très probablement des effluents des exploitations de mines d'or situées sur la rive nord du lac.

Les concentrations de BPC dans les étaient d'environ un ou deux ordres de grandeur receuillis inférieures à celles observées dans le lac Ontario. Toutefois, la distribution des BPC montrait que les sédiments du Grand Lac de l'Esclave contenaient plus de biphényles plus faiblement chlorés que ceux du lac Ontario. Il faut entreprendre d'autres études pour savoir si les plus fortes concentrations de ces biphényles chlorés constatées dans les sédiments situés sous la surface proviennent des processus de déchloruration des BPC dans l'environnement (dégradation bactérienne, provoqués photolyse) ou de différentes sources (apports riverain atmosphérique).

Les concentrations d'autres hydrocarbures chlorurés dans les sédiments du Grand Lac de l'Esclave étaient inférieures de deux ordres de grandeur au maximum par rapport aux teneurs des sédiments du lac Ontario. Cependant, les concentrations de plusieurs composés, en particulier le chlordane et l'hexachlorobenzène étaient considérablement élevées et uniformes dans les profils sédimentaires. Ces résultats concordent avec les conclusions et suggestions d'autres chercheurs qui ont étudié la distribution des hydrocarbures chlorurés dans l'écosystème subarctique et arctique.

En conclusion, les sédiments du Grand Lac de l'Esclave constituent une voie de transport possible des contaminants organiques dans le fleuve Mackenzie. D'autres études exhaustives de la distribution des contaminants organiques dans les sédiments en suspension et de fond prélevés dans le réseau hydrographique de la rivière de l'Esclave, du Grand Lac de l'Esclave et du fleuve Mackenzie amélioreraient nos connaissances limitées sur la contamination des régions subarctiques et arctiques du Canada par les substances organiques provenant de différentes sources ainsi que sur le transport et sur le recyclage global de ces contaminants toxiques persistants.

MANAGEMENT PERSPECTIVE

A preliminary study of geochemistry and concentrations of inorganic and organic contaminants showed similar concentrations of trace elements (Cu, Ni, Co, Cr, V, Pb and Zn) in sediments collected from Great Slave Lake, NWT. These concentrations represented the background levels in the lake's drainage The concentrations of As were greater in the surficial sediments indicating recent input of this element into the lake. The concentrations of PCB's in Great Slave Lake sediments were one to two orders of magnitude lower than those found in The distribution pattern of PCB's and other Lake Ontario. chlorinated hydrocarbons in Great Slave Lake sediments indicated different sources, most likely the Slave River, the major lake's tributary, and atmospheric deposition. Great Slave Lake sediments appear to be potential storage of organic contaminants which may be transported further by sediment resuspension into the Mackenzie River. A comprehensive investigation of the distribution of organic compounds in the Slave River-Great Slave Lake-Mackenzie River system would contribute to the present limited knowledge of the transport of these toxic contaminants into Canadian sub-arctic and arctic regions, and their global cycling through different ecosystem compartments.

PENSIFECTIVE DE CESTION

Une étude préliminaire de la composition géochimique des sédiments recueillis dans le Grand Lac de l'Esclave et de leur teneur en contaminants inorganiques et organiques a montré que, dans ces sédiments, les concentrations d'oligo-éléments (Cu, Ni, Co, Cr, V, Pb et Zn) étaient similaires. Celles-ci correspondaient aux concentrations naturelles observées dans le bassin d'alimentation du lac. Les concentrations d'As étaient supérieures dans les sédiments superficiels, ce qui indique un apport récent de cet élément dans le lac. Les concentrations de BPC dans les sédiments du Grand Lac de l'Esclave étaient d'environ un ou deux ordres de grandeurs inférieures à celles observées dans le lac Ontario. La distribution des BPC et d'autres hydrocarbures chlorurés dans les sédiments du Grand Lac de l'Esclave indiquait qu'il y avait d'autres sources, très probablement la rivière de l'Esclave, le principal affluent du lac, et les dépôts atmosphériques. Les sédiments du Grand Lac de l'Esclave semblent être un lieu possible d'entreposage des contaminants organiques qui peuvent être transportés plus loin jusque dans le fleuve Mackenzie par les sédiments en suspension. D'autres études exhaustives de la distribution des contaminants organiques dans le réseau hydrographique de la rivière de l'Esclave, du Grand Lac de l'Esclave et du fleuve Mackenzie amélioreraient nos connaissances encore limitées sur le transport de ces contaminants toxiques dans les régions subarctiques et arctiques du Canada et sur leur recyclage global à travers différents écosystèmes.

INTRODUCTION

The global cycling and deposition of toxic organic chemicals and metals is well recognized as a major source of such compounds to the environment, and, in general, is recognized as a serious concern in North America and Europe. The Chernobyl nuclear incident made it very clear how rapidly chemicals can circumnavigate the earth and be deposited in distant areas.

The pollution of the aquatic environment is widespread and occurs in areas without known sources of the contaminants involved. Deposition of many compounds from the atmosphere has been established as a major route for the contamination of marine and freshwater ecosystems in remote areas of the world.

Chlorinated and polycyclic hydrocarbons have been used, sometimes extensively, in industrialized locations and in agriculture. Nowadays, these persistent organic chemicals have been found in the environment not only at the locations of their use but also in the remote areas of the world including arctic and anarctic regions. Organochlorine residues were detected in seabird eggs collected in Canadian arctic (Noble and Elliot, 1986). Impaired fish liver quality has been reported by local fisheries on the lower Mackenzie River (Nagy et al., 1987) and toxaphene was detected in a fish sample from the Mackenzie River (B. Olding, IWD, Yellowknife, NWT, personal communication).

A preliminary study was carried out in the summer of 1987 to determine the geochemistry and concentrations of trace elements and organic contaminants in sediments from selected areas in Great Slave Lake, NWT. The objective of the study was to assess the role of the lake in global cycling of contaminants and the potential of the lake's sediments for transporting contaminants through the Mackenzie River into the southern Beaufort Sea.

MATERIALS AND METHODS

Study Area

Great Slave Lake with an area of 26,900 km² is a major component of about 6,000 km long Mackenzie River drainage system and the fourth largest lake in Canada (Fig. 1). Maximum depth of the lake is 162 m in the west, and 614 m in the east lake's basins. Limnology and geological setting of the lake were described by Rawson (1950) and Allan (1979). The Slave River, a 415-km long major tributary, supplies about 87% of the total annual discharge to the Great Slave Lake (Rawson, 1950). The Slave River combines the contribution of two southern sub-basins of the Mackenzie River system, the Athabasca and Peace River basins, and enters the lake by the Slave Delta. The lake's only outflow is the Mackenzie River (Fig. 2). Hay River, 700 km

long, Yellowknife River and few smaller streams are the other tributaries. The lake stabilizes the flow, water temperature and water levels of the entire upper Mackenzie River, and is a sink for the suspended material discharged by the tributaries, particularly the Slave River with mean suspended load 30 x 10⁶ tons/year (Mackenzie River Basin Committee, 1981). The City of Yellowknife with a population of 11,000 is located on the northern shore of the lake. There are few small towns along the lake's shore. Mining, particularly gold and Pb-Zn mines, is the main industry around the lake. However, heavy oil and tar sand deposits and about 1 million hectares of farmland are in the Athabasca and Peace River areas (Mackenzie River Basin Committee, 1981).

Sediment Sampling and Sample Preparation

The location of the sampling stations was selected according to previous investigation of geochemistry of Great Slave Lake sediments by Allan (1979). To locate fine-grained sediments suitable for coring the bottom in the vicinity of selected sampling stations was surveyed by echo sounding equipment mounted on the vessel. In addition, echo sounding was carried out on a line extending from the deepest area in the west basin to the mouth of the Slave River.

Fisheries and Oceans vessel Christie Bay operated by staff members of the Department of Fisheries and Oceans in Hay River, NWT, was used for the sampling operation.

Surface sediment samples were obtained by a mini-Ponar grab sampler at five sampling stations. Five sediment cores about 60 cm long were collected at three sampling stations (Fig. 1) using a Benthos corer with plastic liners. cores were subsampled into 1 cm sections down to 20 cm sediment depth, and into 2 cm sections below 20 cm sediment depth. Subsamples from two cores from each station were composited into hexane-prewashed glass jars for the determination of selected Subsamples from three cores from each organic contaminants. station were composited into plastic bags for geochemical analyses, dating and the determination of particle size distribution. All samples in the glass jars were immediately frozen, shipped to the laboratory at the Canada Centre for Inland Waters (CCIW), Burlington, Ontario, where they were freeze dried and used for the determination of organic contaminants. One portion of the subsamples composited into plastic bags was used for the particle size analyses and the second portion was frozen, freeze dried and homogenized by grinding to 189 µm particle size for geochemical analyses.

Chemical Analyses

Sediment geochemistry and metal concentrations

The concentrations of major elements (Si, Al, Fe, Ca, Mg, K, Na, Ti, Mn and P) and trace elements (Cu, Cr, Co, Pb, Ni, V, Zn and As) were determined in all samples by X-ray fluorescence spectrometry using the method described by Mudroch (1985). The precision of the analysis was determined by analysing five pellets made from a homogenized sediment sample. The accuracy of the analysis was verified by running Canadian Reference Standards Syenite SY^{-2} and soils SO^{-2} and SO^{-4} , and comparing the analytical results with the stated reference The concentration of values for major and trace elements. organic matter was determined as loss on ignition at 550°C. The mineralogical composition of sediments was investigated by powder X-ray diffraction using Cu-target with a Ni-filter. The particle size distribution was determined by the method described by Duncan and LaHaie (1979).

Chlorinated Hydrocarbons

Chlorinated hydrocarbons were determined in selected sediment subsamples from the following depths in the cores: 0 to 1 cm, 4 to 5 cm, 5 to 6 cm, 9 to 10 cm and 14 to 15 cm at

each station; 19 to 20 cm at station 3, and 26 to 28 cm and 40 to 42 cm at station 4.

The analytical procedure for the determination of chlorinated hydrocarbons consisted of 12 hours Soxhlet extraction by dichloromethane. The extracts were subjected to a fractionation by column chromatography prior to analysis using the method described by Carey and Hart (1986). Each of the resulting fractions was subjected to analyses using a dual-column capillary gas chromatography method with 30 m long DB5 and DB17 columns and electron capture detectors. Reproducibility of the analysis on replicate samples was +10%. National Research Council, Canada, standard material CLB-1, a mixture of 51 PCB congeners, was used as the standard in the analysis. The accuracy of the determination of PCB's was confirmed by analysis of Sediment Reference Material for Total Polychlorinated Biphenyls obtained from Quality Assurance and Method Section, National Water Research Institute, Burlington, Ontario (Lee and Chau, 1987). Chlorinated hydrocarbons including PCB isomers analyzed in Great Slave Lake sediments are listed in Appendix 1.

Sediment Dating

The total Pb-210, Ra-226 and Cs-137 levels in the sediment core sections from station 4 were measured by

low-energy gamma-ray spectrometry using techniques described by Joshi (1987). The contribution of atmospherically-delivered or excess Pb-210 (hereafter referred to as Pb-210) was obtained by subtracting the amount of Pb-210 supported by Ra-226 from total Pb-210. The sediment age was calculated by the method described previously (Mudroch and Joshi, 1988).

RESULTS AND DISCUSSION

Bottom Morphometry and Sediment Geochemistry

The echo sounding revealed a very gentle sloping of the lake bottom from the Hay River mouth towards station 3 (Fig. 1). The echo sounding from station 4 (water depth about 110 m) towards the mouth of the Slave River (water depth about 20 m) showed steep sloping of the bottom from deep water towards the Slave River bar (Fig. 3). The steep slope, the configuration of the bottom below the bar and fine lenses of a darker material in the sediment profile at station 4 suggested sediment slumping and turbidity currents in this part of the lake. The bar at the Slave River mouth was firm, consolidated material, most likely sand.

Generally, the sediments from all sampling stations consisted of very fine grey-brown silty clay, with a surface layer (1 to 5 cm) less consolidated than the underlying

sediments. Few amphipods were found at the sediment surface at each sampling station.

The samples contained 77 to 92% clay-size particles (2 µm). The rest of the particles was silt (particle size 2 to 63 µm). Major minerals were quartz, illite, chlorite, kaolinite and feldspars. Other minerals in the sediments were calcite and small quantities of many different minerals originating from complex bedrock geology of the lake drainage basin (Allan, 1979).

The concentrations of major elements were uniform from the surface to the bottom of each sediment core and similar for all three coring stations (Table 1), indicating a continuous deposition of geochemically similar material. Low concentrations of organic C in the sediment profile indicated the deposition of mineral particles and limited contribution of autochtonous organic matter to the sediments in this oligotrophic lake.

All trace elements but As were uniformly distributed in the sediment profiles at all coring stations (Table 2), and represented background concentrations in Great Slave Lake sediments derived from regional geology of the lake's drainage basin. However, As increased in the surficial sediment from 5 to 13 μ g/g and 6 to 14 μ g/g at stations 1 and 3, respectively. At station 4 the concentrations of As were uniform (12 to

14 $\mu g/g$) within 42 cm sediment layer. The increase of As in surficial sediments at stations 1 and 3 agreed well with the observations of Allan (1979) who found an increase of As concentrations (up to 14 $\mu g/g$) in the sediments at central west basin of the lake. The source of As enrichment might be effluents or emissions from gold mines on the north shore (Mudroch et al., 1988). Uniform concentrations of As in the sediment profile at station 4 reflected a fast deposition of recent sediment with a greater As content or mixing of the sediments at this area.

Sediment Dating

The sedimentation rates at station 4 were calculated using both CIC and CRS models. Both indicated that the sediment at this area was very young: 14 cm sediment section was about 0.23 years of age using both models. The results from both models were virtually the same. When a diffusive mixing parameter was used, the mixing-influence age of the 14-cm sediment layer was only about 0.02 years. The Pb-210 profile was very atypical as little change was observed in activity with depth in core, suggesting again a high sedimentation rate. The CIC model gave a mass sedimentation rate of about 46.5 g/cm²/y¹. The Cs-137 activity was very low compared with the levels attributable to early 60's fallout. Possibly this activity was due to

the 1986 nuclear reactor accident in Chernobyl, U.S.S.R. This activity has been detected in the Canadian north earlier (Joshi, 1987).

The sediment age determined by CIC, CRS and diffusional models with relevant parameters is shown in Table 3. The atypical Pb-210 profile, the concentrations of Cs-137, the morphometry of the bottom and lenses of different colour material found by the visual inspection of the cores indicated a high sedimentation rate and intensive sediment mixing at station 4.

Estimated mean sediment inflow from the Slave River into the lake is 30×10^6 tons/year, of which 10% is sand and the rest is fine-grained material (Mackenzie River Basin Committee, 1981). It appears that the majority of this material becomes deposited on the bottom of the lake at the area adjacent to the river delta. If the suspended load would become deposited over an area approximately 50 km^2 , the thickness of the sediment deposited at station 4 would be about 7 cm. This estimate is in agreement with our determination of the sedimentation rate at station 4.

Organic Contaminants

PCB's

Total concentrations of PCB's in selected sections of sediment cores collected at three stations in Great Slave Lake are compared to those found in the sediments from the deepest depositional basin of Lake Ontario in Table 4. Generally, the concentrations of PCB's in Great Slave Lake sediments were 10 to 100 times lower than those in Lake Ontario. The greatest concentration of PCB's in Great Slave Lake (22.76 ng/g) was found at the 26 to 28 cm sediment depth at station 4. Considering a sedimentation of the 17-cm sediment layer in about one month, the 26 to 28 cm section would be deposited at station 4 between March and April. According to Allan (1979), in this geographical setting, peak sediment load is prior to peak spring discharge and the actual sediment load during early spring snowmelt considerably exceeds the load generated by a rainfall event. The major tributary of Great Slave Lake, the Slave River, most likely affects the sediment quality at station 4. Consequently, the increase of the PCB's concentrations in the 26 to 28 cm sediment section at this station may reflect increased input of PCB's from the Slave River during the snowmelt runoff.

The monitoring of water quality in the Athabasca River revealed detectable concentrations of PCB's. Maximum concentrations of Arochlors 1248, 1254 and 1260 were 0.165, 0.019 and

0.007 µg/L, respectively, in nonfiltered water from the Athabasca River, and 1.70 and 2.40 µg/L of Arochlor 1248 and 1260, respectively, in water from lakes in the Athabasca River basin (Mackenzie River Basin Committee, 1985). These findings indicate a transport of PCB's into Great Slave Lake via Slave River tributaries. In addition to industrial sources in the tributaries drainage basin, including potential sources at the heavy oil and tar sand deposits, accumulation of PCB's deposited over the winter from atmospheric sources most likely contributes to the load during snowmelt which is the period of the largest transport of suspended material by the Slave River into the lake.

The distribution pattern of PCB's in the sediments from Great Slave Lake and that of the sediments from the deepest depositional basin of Lake Ontario are shown in Figs. 3a-3d. Tetrachlorobiphenyls represented 44 to 60% of the total PCB's in the sediment from station 4 in Great Slave Lake, followed by 8 to 39% tri-, 11 to 28% penta-, and 4-24% hexachlorobiphenyls. Dichlorobiphenyls (about 6%) were found only at the 26 to 28 cm sediment depth at this station. At stations 1 and 3 tetrachlorobiphenyls represented about 60% of total PCB's in the surface 1 cm sediment section, and the distribution pattern of PCB's was similar to that of sediments from station 4. However, in the deeper parts of sediment cores from stations 1 and 3,

dichlorobiphenyls represented 23 to 47% of total PCB's, followed by tri- and tetrachlorobiphenyls. Penta- and hexachlorobiphenyls represented only small portion of total PCB's (Figs. 4a and 4b).

This distribution pattern of PCB's in the sediments from Great Slave Lake suggested different PCB's sources at stations 1 and 3 from station 4 indicating again that the area at station 4 was most likely affected by the Slave River. concentrations of As and distribution pattern of PCR's showed that surficial sediments at stations 1 and 3 were similar to those from station 4, and suggested most likely sediment transport from the Slave River mouth towards the outlet of the Mackenzie River. However, the distribution pattern of PCB's in deeper sediment sections from stations 1 and 3, particularly greater concentrations of lower chlorinated biphenyls, indicated different sources from those affecting the distribution pattern of PCB's at station 4, or dechlorination processes in the sediment column (Figs. 4a-4c). Brown et al. (1987) showed that dechlorination processes producing lower chlorinated biphenyls can be mediated by a different population of anaerobic bacteria with its own distinctive pattern of PCB's congener selectivity. They found that synthetic 2,3,4,5,6-pentachlorobiphenyl can be partially dechlorinated by a 32-week anaerobic incubation with unsterilized, but not with sterilized, upper Hudson sediments, indicating microbial mediation. The distribution pattern of PCB's in Great Slave Lake sediments indicated the possibility of these processes. The concentrations of tetrachlorobiphenyls found in recently deposited sediments from station 4 and surficial oxidized sediments from stations 1 and 3 were greater than those in subsurface sediments (most likely anaerobic). However, subsurface sediments had greater concentrations of di- and trichlorobiphenyls.

Photolysis by solar near-ultraviolet light is another route for the dechlorination of higher to lower chlorinated biphenyls, which are more slowly photolyzed. Modeling studies revealed that solar photolysis may reduce the levels of heavy chlorinated PCB's in large lakes or oceans with half-times of a year or two (Bunce et al., 1978).

Lower chlorinated biphenyls are more readily biodegraded by aerobic bacteria (Bedard et al., 1986), and more water soluble than higher chlorinated ones. This is an important fact regarding the further assessment of distribution and transport of PCB's into the Mackenzie River. However, more studies have to be conducted before drawing a conclusion regarding the distribution pattern of PCB's in Great Slave Lake sediments.

The distribution pattern of PCB's in Great Slave Lake sediments was different than that in the deepest depositional

basin of Lake Ontario where tetrachlorobiphenyls represented about 60% of total PCB's. In addition, the concentrations of lower chlorinated PCB's in Lake Ontario were considerably lower than in Great Slave Lake (Fig. 4d).

Other Chlorinated Hydrocarbons

The concentrations of other chlorinated hydrocarbons determined in Great Slave Lake sediments were in the range of 10^{-5} to $10^{-3} \mu g/g$. The concentrations of individual compounds in the sediments from three sampling stations are shown in Table 5. Generally, the concentrations of determined compounds decreased with increasing sediment depth at stations 1 and 3. The concentrations of pentachlorobenzene and A-chlordane were consistent in sediment sections below 4 cm depth at station 1and 3, respectively (Fig. 5). Generally, the concentrations of determined compounds were similar from the surface to 42 cm depth at station 4. The concentrations of determined chlorinated hydrocarbons in surficial sediments from Great Slave Lake were up to two orders of magnitude lower than in Lake Ontario (Table 6) but the concentration of A-BHC which was similar in both sediments. The distribution pattern of compounds listed in Table 6 and plotted in Fig. 6 shows that A-BHC represented a

large portion of the total quantity of determined compounds in Great Slave Lake sediments.

The most important mechanism of dispersal of neutral organochlorine contaminants in the global environment is the long-range atmospheric transport (Bidleman et al., 1981). According to Norstrom and Muir (1986), primary emission of organochlorines probably dominated inputs to the global environment when these compounds were intensively used. After this use has been severly restricted in most developed nations during 1970's, complex exchange process between the atmosphere, land and surface waters is perhaps the dominant factor in input loadings of PCB and many pesticides at mid-latitudes in the northern hemisphere. Atmospheric deposition may be important at sub-arctic latitudes in the path of air mass movement from midlatitudes, since annual precipitation at sub-arctic latitudes is greater than in arctic regions. Several organochlorines have been identified in Arctic snow and ice at concentrations of 1 ng/L, except for A-HCH (McNeely and Gummer, 1984) detected around 3 ng/L in arctic snow by Tanabe et al. (1983). Levels of PCB's and DDT were in sub-ng/L concentrations (Norstrom and Muir, 1986).

The concentration pattern of PCB's in Great Slave Lake sediments indicated a deposition of considerably greater quantities of lower than higher chlorinated biphenyls at this area.

This finding agrees with the statement of Ottar (1981) that organochlorines, in particular more volatile low chlorinated PCB, can be re-emitted to the atmosphere after an initial deposition by sublimation and transferred from warmer to colder regions, while the heavier chlorinated biphenyls have probably not yet reached this stage. Consequently, greater concentrations of biphenyls with lower chlorine content in Great Slave Lake sediments may reflect these processes.

Chemical analyses of the Arctic aerosol showed that considerable amounts of pollutants were brought into the Arctic region in winter (Ottar, 1981). This may explain the different distribution pattern and concentrations of PCB's in the sediments at station 4 from stations 1 and 3 in Great Slave Lake. The estimation of sediment age at station 4 showed that the 14 cm sediment layer was deposited in April during the ice and snow melt. Greater concentrations of lower chlorinated biphenyls in subsurface sediments from stations 1 and 3 may reflect the emission of these compounds during higher summer temperatures from the mid and lower latitudes of the northern hemisphere, and their transport and deposition at the sub-arctic However, the transport of PCB's by the Slave River suspended load has to be considered a potential source of PCB's to Great Slave Lake. In addition, the dechlorination of PCB'c may be an important process in the lake's sub-surface sediments.

Many of the other chlorinated hydrocarbons found in Great Slave Lake sediments may originate from pesticide use in the agricultural area of the prairies. Unfortunately, only limited information is available on the type and quantities of pesticides previously used.

Consistent concentrations of A-chlordane in the sediment profile at all sampling stations reflects an intensive use, high persistency and atmospheric transport of this compound. According to Noble and Elliot (1986) technical chlordane, which contains 10.6% cis- and 9% trans-chlordane, 7% nonachlor, and 8% heptachlor, was used extensively in the U.S.A. and Canada as an insecticide. In most animals, chlordane is readily metabolized to compounds of which oxychlordane is the most persistent (Menzies, 1980). Oxychlordane was found in polar bears in greater concentrations than any other organochlorine, and chlordane compounds were more prevalent in seabirds from Canadian arctic than further south (Noble and Elliot, 1986). Chlordane levels were higher in northern latitudes in Europe although its use was greater in the south, suggesting that volatile chlordane was readily transported northward by atmospheric processes Our results on the distribution of organo-(Ottar, 1981). chlorines in Great Slave Lake sediments agree with these findings. In Great Slave Lake sediments, A- and G-chlordane were

two of the few compounds present in the subsurface sediments at all three sampling stations.

Total concentrations of organochlorines in the water of the Athabasca and Peace Rivers were below detection limit with the exception of A- and G-BHC (Mackenzie River Basin Committee, 1985). Greatest concentrations of these compounds (up to 10 ng/L) in the spring and late summer or fall were attributed to the time of use of pesticides and the chemistry of the compounds. These compounds become readily adsorbed on particulate matter, which explains the greatest concentrations in the spring when suspended sediment concentrations are elevated and pesticide residues become flushed from the watershed during snowmelt and by surface runoff. Consequently, pesticides in Great Slave Lake sediments originated most likely from their application in the drainage basin and transport by Athabasca and Peace Rivers via the Slave River into the lake.

The other compound frequently detected in the sediments was heptachlor epoxide. This compound is considered the most persistent metabolite of heptachlor. The presence of heptachlor epoxide in the eggs of the Ivory gull, an arctic resident, indicates that this contaminant is transported to the arctic (Noble and Elliot, 1986).

Limited information exists on the distribution and transport of other chlorinated hydrocarbons in the sub-arctic

and arctic regions. Hexachlorobenzene (HCB) originally used as a fungicide in Canada and the U.S.A. and occurring also as a byproduct of some industrial processes (Hallet et al., 1982) was found in elevated levels east of Newfoundland and in the arctic (Noble and Elliot, 1986). Ottar (1981) noted that HCB was readily transported to northern latitudes from sources further south. The lack of geographic variation of HCB levels in polar bears from the Canadian arctic found by Norstrom also suggested that atmospheric rather than ocean transport was the main vector (Noble and Elliot, 1986). These findings support our results of the distribution of HCB, which was found in all sediment samples collected during our study in Great Slave Lake.

Dieldrin was used since the early 60's in North America with reduced use in Canada during the 60's and after 1971. According to Noble and Elliot (1986) less volatile and more water-soluble dieldrin tends to be removed from the atmosphere by precipitation and dissolution at the air/water interface of large water bodies. Consequently, this compound was considered a more local contaminant usually associated with riverine/land runoff (Norstrom et al., in preparation). In Great Slave Lake sediments dieldrin was found only in the surface 6 cm at station 4, indicating its transport into the lake by the Slave River.

A more comprehensive investigation of the distribution of chlorinated hydrocarbons in suspended and bottom sediments from the Slave River-Great Slave Lake-Mackenzie River system would enhance present limited knowledge on contribution from different sources and the transport and global cycling of these toxic persistent organic contaminants.

SUMMARY AND CONCLUSIONS

- 1. A preliminary investigation was carried out in the summer of 1987 to determine the geochemistry and distribution of trace elements and organic contaminants (PCB's and other chlorinated hydrocarbons) in the sediments from Great Slave Lake, NWT. The objective of the investigation was to assess the role of the lake in the global cycling of contaminants and the potential of the lake's sediment for transport of contaminants into the Mackenzie River.
- 2. Echo sounding of the lake bottom showed a steep slope at the Slave River mouth extending from about 20 to 110 m water depth, and confirmed the presence of fine-grained sediments at selected sampling station. Sediment cores were collected at three stations in the lake. The cores were subsampled

for the determination of sediment geochemistry, organic contaminants and sediment dating.

- 3. Sediment dating by Pb-210 and Cs-137 showed very high sedimentation rate (46.5 g/cm²/year) at the sampling station located at the deepest area (approximately 110 m) of lake's west basin oposite the Slave River mouth.
- 4. All sediment samples consisted of fine-grained (63 µm particle size) sediments. A uniform concentration of major elements in the sediment profile showed deposition of similar material at all three sampling stations. The concentrations of trace elements were similar in all sediment profiles and most likely represented the background levels in the sediments derived from the Great Slave Lake drainage basin. On the contrary, the concentration of As increased towards the surface of the sediment at two sampling stations indicating an input of As into the lake, most likely from the emission or wastes from gold mines located on the north shore of the lake.
- 5. The concentrations of PCB's in Great Slave Lake sediments were one to two orders of magnitude lower than those in Lake Ontario. However, the distribution pattern of PCB's showed

that lower chlorinated biphenyls were present in greater amounts in Great Slave Lake sediments than in Lake Ontario. The distribution pattern of PCB's in the sediments from three stations in Great Slave Lake indicated different sources of these contaminants or dechlorination processes in sub-surface sediments.

- The concentrations of other sixteen chlorinated hydrocarbons were up to two orders of magnitude lower in Great Slave Lake sediments than those in Lake Ontario. However, the concentrations of chlordane and hexachlorobenzene were considerably high and uniform in sediment profile at all sampling stations.
- 7. A uniform distribution of PCB's and other determined chlorinated hydrocarbons in sediment profiles from the station located near the deepest area of the lake's west basin opposite the Slave River mouth may be the result of high sedimentation rate, sediment mixing and transport of these contaminants by the Slave River into the lake.
- 8. It was concluded that Great Slave Lake sediments are a potential storage of chlorinated hydrocarbons which may become transported by sediment resuspension into the

Mackenzie River. However, a comprehensive investigation of the distribution of organic contaminants in suspended and bottom sediments from the Slave River-Great Slave Lake-Mackenzie River system would improve present limited knowledge of transport of these persistent toxic contaminants into the Canadian sub-arctic and arctic. The results of the investigation indicated that sedimentary deposits of freshwater lakes in remote areas can provide a record of global cycling of many contaminants.

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FIGURE CAPTIONS

- Figure 1. Sediment sampling stations in Great Slave Lake
- Figure 2. Drainage subbasins of the Mackenzie River basin
- Figure 3. Bottom morphometry from station 4 to the Slave River mouth
- Figure 4a. Distribution of PCB's in sediment from station 1
- Figure 4b. Distribution of PCB's in sediment from station 3
- Figure 4c. Distribution of PCB's in sediment from station 4
- Figure 4d. Distribution of PCB's in sediment from deepest depositional basin of Lake Ontario
- Figure 5. Distribution of A-chlordane and pentachlorobenzene in the sediments
- Figure 6. Distribution of selected chlorinated hydrocarbons in sediments from Great Slave Lake and Lake Ontario

TABLE 1. Concentration range of major elements in sediments (% dry weight)

Parameter	Station 1	Station 3	Station 4
S10 ₂	67.5 - 68.8	65.7 - 67.3	66.9 - 68.4
A1 ₂ 0 ₃	16.9 - 17.5	17.4 - 18.8	17.1 - 18.5
Fe ₂ O ₃	6.91 - 7.35	7.02 - 7.79	5.96 - 6.67
MgO	1.05 - 1.15	1.05 - 1.29	1.56 - 2.35
CaO	1.65 - 1.81	1.62 - 1.77	1.59 - 1.67
Na ₂ O	0.60 - 0.65	0.66 - 0.68	0.49 - 0.66
K ₂ 0	3.05 - 3.40	3.07 - 3.38	2.81 - 3.04
T102	0.90 - 0.95	0.96 - 1.03	0.94 - 0.99
MnO	0.09 - 0.15	0.11 - 0.34	0.07 - 0.10
P ₂ O ₅	0.15 - 0.17	0.17 - 0.19	0.14 - 0.16
Organic C	1.00 - 1.25	1.05 - 1.33	1.25 - 1.58

TABLE 2. Concentration ranges of trace elements in sediment (µg/g dry weight)

Parameter	Station 1	Station 2	Station 3
Cu	28 - 31	30 - 41	20 - 30
Ni	50 - 56	50 - 60	47 - 52
Co	9 - 14	8 - 14	8 - 15
Cr	110 - 132	115 - 141	108 - 133
V	168 - 175	174 - 184	157 - 179
Pb	25 - 32	27 - 41	21 - 30
Zn	132 - 148	137 - 150	130 - 150
As	5 - 13	6 - 14	12 - 14

TABLE 3. Sediment age by CIC, CRS and diffusional models and relevant parameters

CORE - GSE

The density of the dry sediment (g·cm ⁻³) The number of data points for which activity is given The error, CE (Bq·g ⁻¹) in the radioactivity measurement The limited depth, XL (cm) up to which activity is given Porosity constant B (cm ⁻¹) Porosity at X = 0 CD for porosity fitting Slope of Cd (Bq·g ⁻¹) VS X (cm) without compaction Activity (Bq·g ⁻¹) at X = 0, without compaction CD for activity, CX fitting Depth up to which Pb is present, Xm (cm) Integrated activity, Al (Bq·cm ⁻²) between XL and XM Sedimentation rate, So (mm yr ⁻¹) by CIC Mass flux, Rl (mg cm ⁻² yr ⁻¹) by CIC Diffusion coefficient, K (cm ² ·yr ⁻¹) neglecting the So CD for CIC fitting Diffusion coefficient, Kl (cm ² ·yr ⁻¹) considering the So	0.005 0.741 0.455 -0.001 0.083 0.0003 1653.872 109.558 733.468 46533.78

Slice #	Depth (cm)	CIC-AGE (yr)	CRS-AGE (yr)	*D
1	0.66	0.009	0.009	0.0008
2	1.86	0.027	0.033	0.008
3	2.94	0.040	0.060	0.020
4	4.05	0.056	0.083	0.026
5	5.17	0.073	0.100	0.027
6	6.30	0.090	0.115	0.024
7	7.47	0.107	0.127	0.019
8	8.61	0.125	0.144	0.019
9	9.73	0.142	0.161	0.019
10	10.83	0.160	0.183	0.024
11	11.93	0.177	0.209	0.031
12	13.03	0.195	0.233	0.038
13	14.13	0.213	0.257	0.044
14	15.24	0.231	0.279	0.048

^{*}D = difference between CIC and CRS-AGES

TABLE 3 (cont'd). Sediment age by CIC, CRS and diffusional models and relevant parameters

Standard deviation, for Eq. #20, in slope Standard deviation, for Eq. #20, in intercept Slope of, the Eq. #20, straight line Intercept of, the Eq. #20, straight line CD of the line represented by Eq. #20	= 568941.8 = 22.151 = 4.1 x 10 ⁵ = 427.294 = 5.4 x 10 ⁵ = 0.968 = 4.5 x 10 ⁵ = 81001.53
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Slice #	Age Without Diffusion (yr)	Age With Diffusion (yr)
•	0.009	0.0007
7	0.026	0.002
2	0.041	0.003
3	0.037	0.004
4 5	0.037	0.006
=	0.073	0.007
6	0.108	0.009
8	0.125	0.010
9	0.123	0.011
10	0.160	0.013
11	0.177	0.014
12	0.195	0.014
13	0.213	0.017
14	0.231	0.019

TABLE 4. Total concentrations of PCB's in Great Slave Lake and Lake Ontario sediments (ng/g)

GREAT SLAVE LAKE

Station 1

0-1 cm 4-5 cm 5-6 cm 9-10 cm 14-15 cm 9.05 5.11 4.71 4.12 3.36

Station 3

0-1 cm 4-5 cm 5-6 cm 9-10 cm 14-15 cm 19-20 cm 5.88 1.46 3.59 5.57 2.67 3.92

Station 4

0-1 cm 4-5 cm 5-6 cm 9-10 cm 14-15 cm 26-28 cm 40-42 cm 3.81 3.86 3.47 4.27 2.19 22.76 8.45

LAKE ONTARIO (Eastern Basin)

0-1 cm 4-5 cm 14-15 cm 586.5 646.5 170.0

TABLE 5. The range of concentrations of selected organochlorines in Great Slave Lake sediments (ng/g)

	Station 1	Station 3	Station 4
Hexachlorobutadiene	0.02-0.23	0.01-0.17	0.04-0.20
1,2,3,4-tetrachlorobenzene	0.06-0.85	0.10-1.01	0.20-0.78
Pentachloroanisol	0.06-0.31	0.04-0.48	0.16-0.54
Lindane	0.04-0.09	0.06-0.12	0.01-0.34
pp-DDE	0.03-0.34	0.01-0.16	0.05-0.21
DDD	0.01-0.33	0.01-0.20	0.01-0.26
pp-DDT	0.01-0.50	0.01-0.25	0.04-0.27
Heptachloroethane	0.01	0.01-0.02	0.01-0.24
Heptachlor	0.01	0.01-0.06	0.01-0.17
Heptachloroepoxide	0.01-0.33	0.01-0.06	0.01-0.18
Dieldrin	0.01	0.01	0.01-0.09

TABLE 6. Concentrations of chlorinated hydrocarbons in surficial sediments of Lake Ontario and Great Slave Lake (ng/g)

	LAKE ONTARIO* Eastern Basin	GREAT SLAVE LAKE			LAKE
		Station 1	Station 3	Station 4	
HCBD	7.9	0.1	0.2	0.2	
A-BHC	1.1	1.0	0.2	0.3	
HBC	52	0.4	0.9	1.0	
Lindane	0.5	0.2	0.1	0.1	
G-chlordane	1.9	0.1	0.1	0.2	
DDE	30	0.1	0.1	0.4	
DDD	20	0.1	0.1	0.3	
DDT	16	0.1	0.1	0.5	

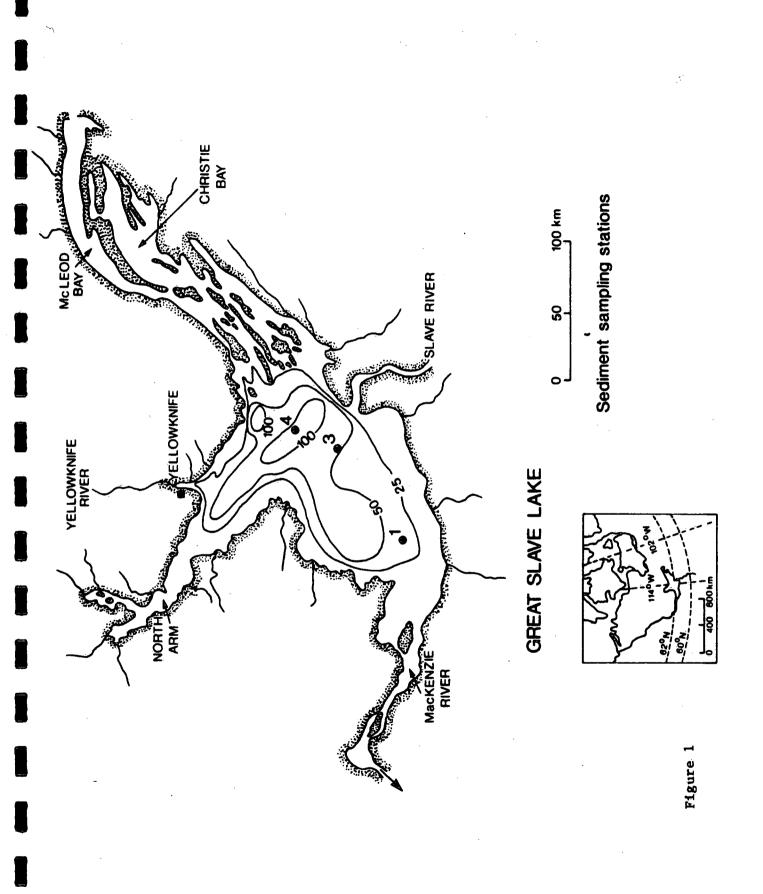
^{*} Oliver, B.G., unpublished data, 1987.

APPENDIX 1

Chlorinated Hydrocarbons Analysed in Great Slave Lake Sediments

PCB iso	mers ¹ :	Other chlorinated hydrocarbons:
15	154	hexachloroethane
18	156	hexachlorobutadiene
31	159	1,2,3,4-tetrachlorobenzene
40	170	pentachlorobenzene
44	171	A-BHC
49	173	hexachlorobenzene
52	180	pentachloroanisole
54	182	lindane
60	183	heptachlor
77	185	<u>-</u>
86	187	heptachlor epoxide
87	189	G-chlordane
101	191	A-chlordane
103	194	dieldrin
105	195	p,p-DDE
114	196	DDD
118	200	p,p-DDT
121	201	
128	202	
129	203	
137	205	
138	206	
141	207	•
143	208	
151	209	
153		

¹ IUPAC No. (Ballschmiter, K. and Zell, M. 1980). Analysis of polychlorinated biphenyls (PCB) by glass capillary chromatography. Fres. Z. Anal. Chem. 302:20-31.



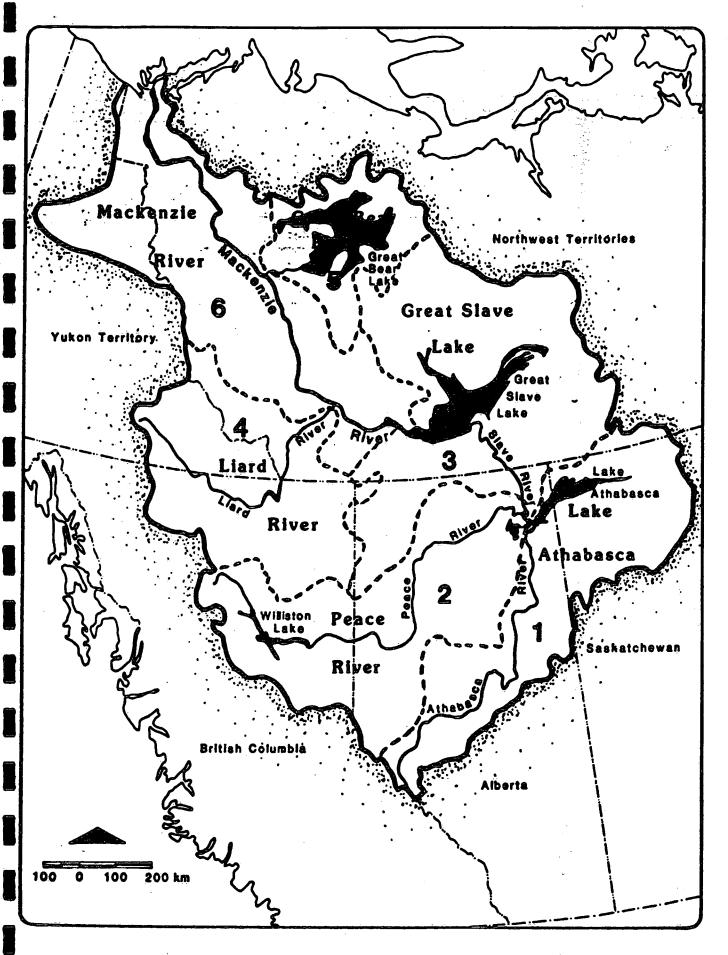
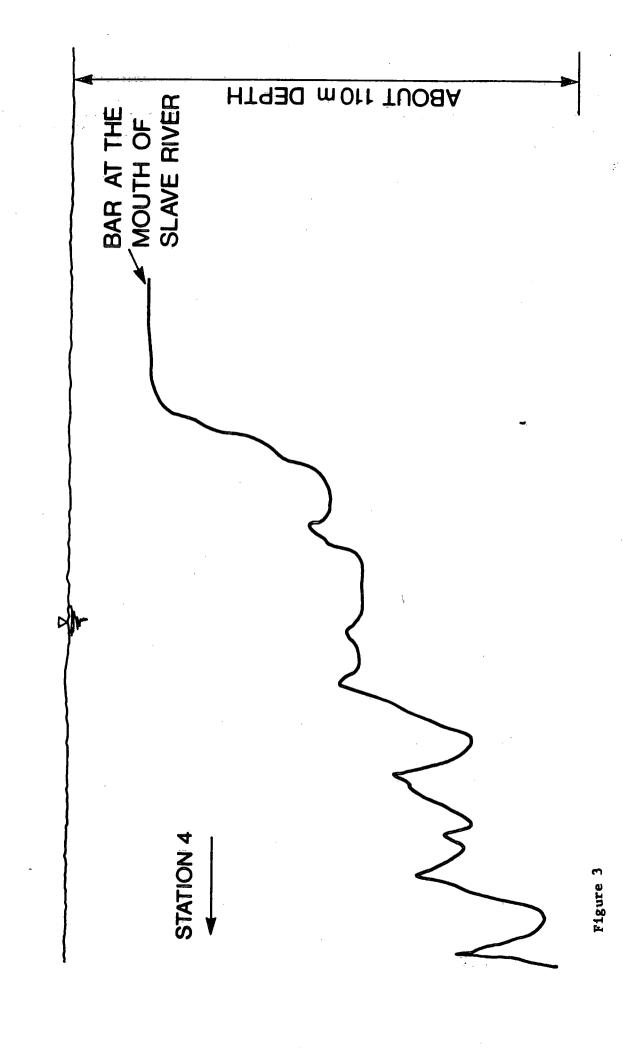


Figure 2



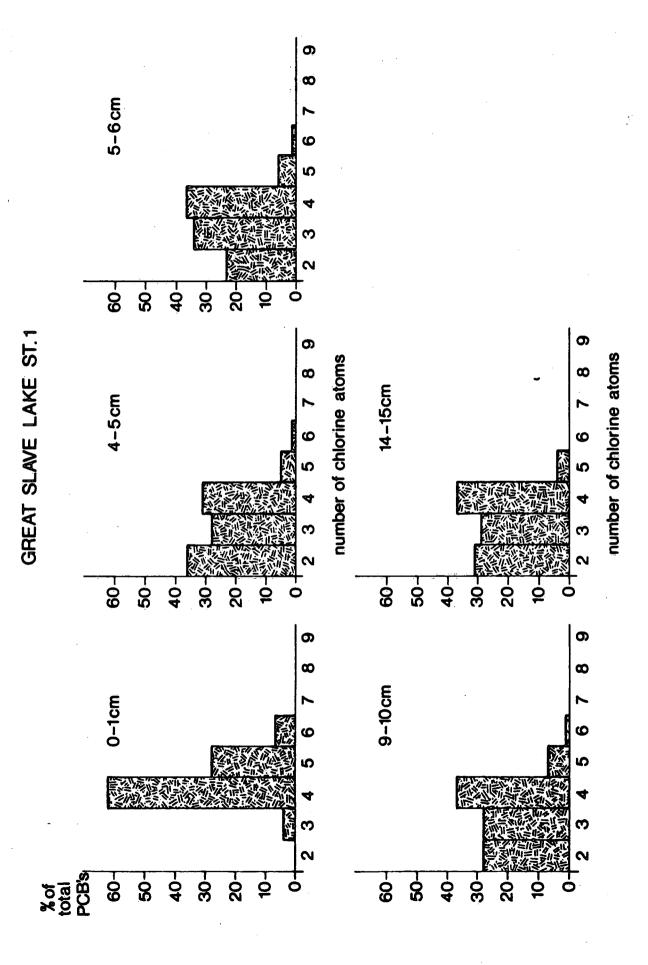


Figure 4a

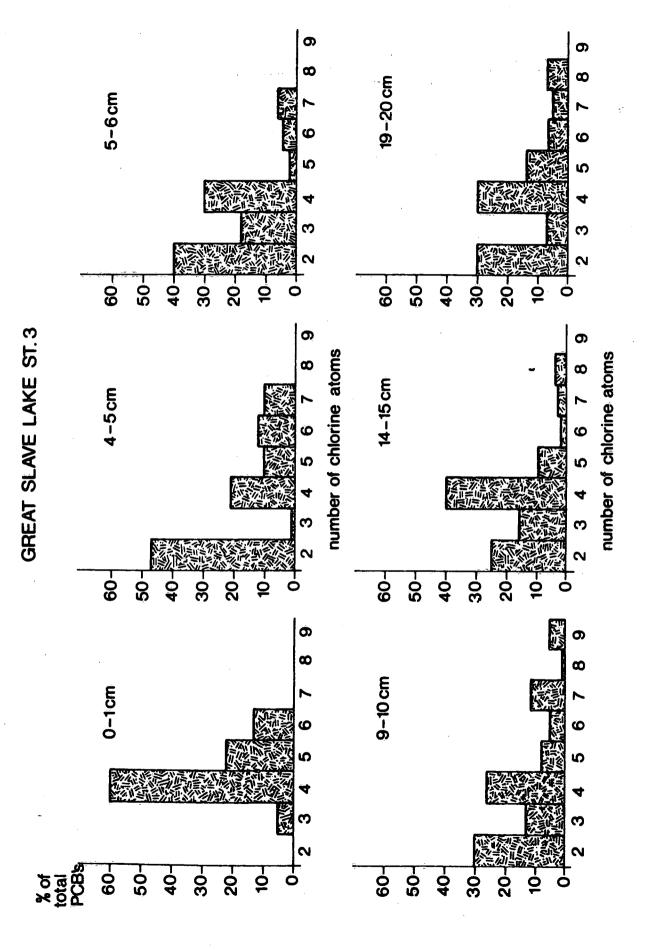


Figure 4b

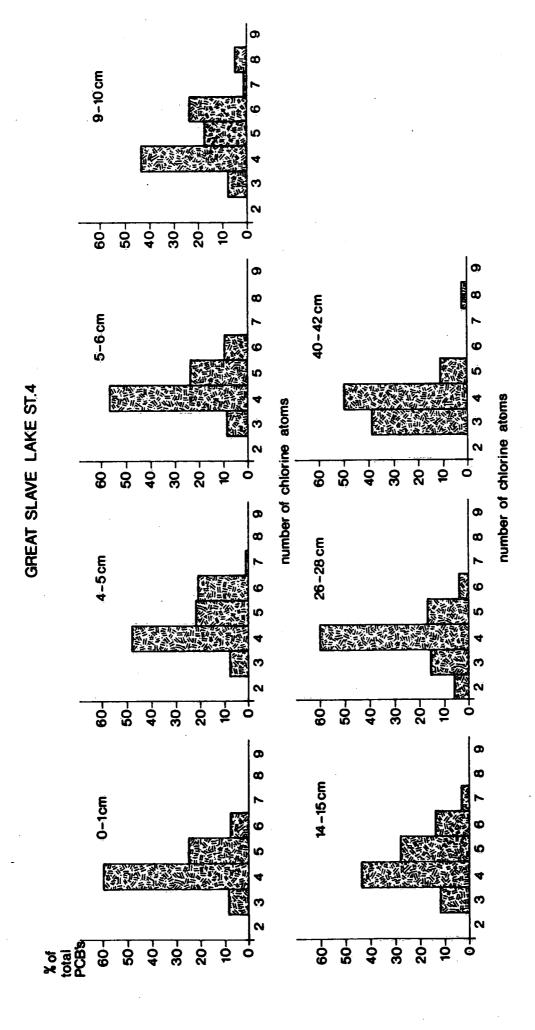
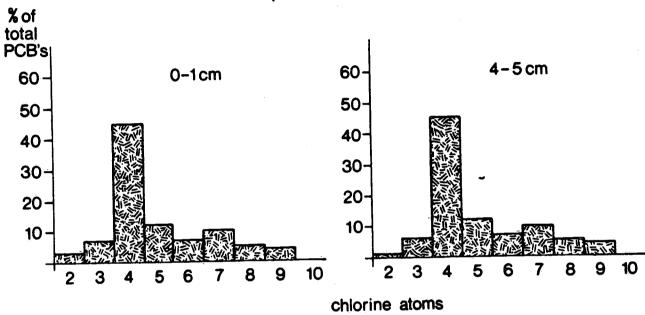


Figure 4c

LAKE ONTARIO-EASTERN BASIN (deepest point)



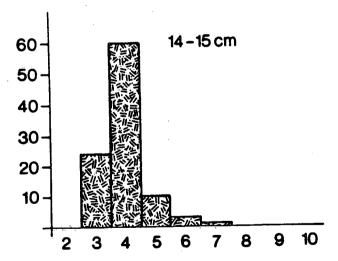


Figure 4d

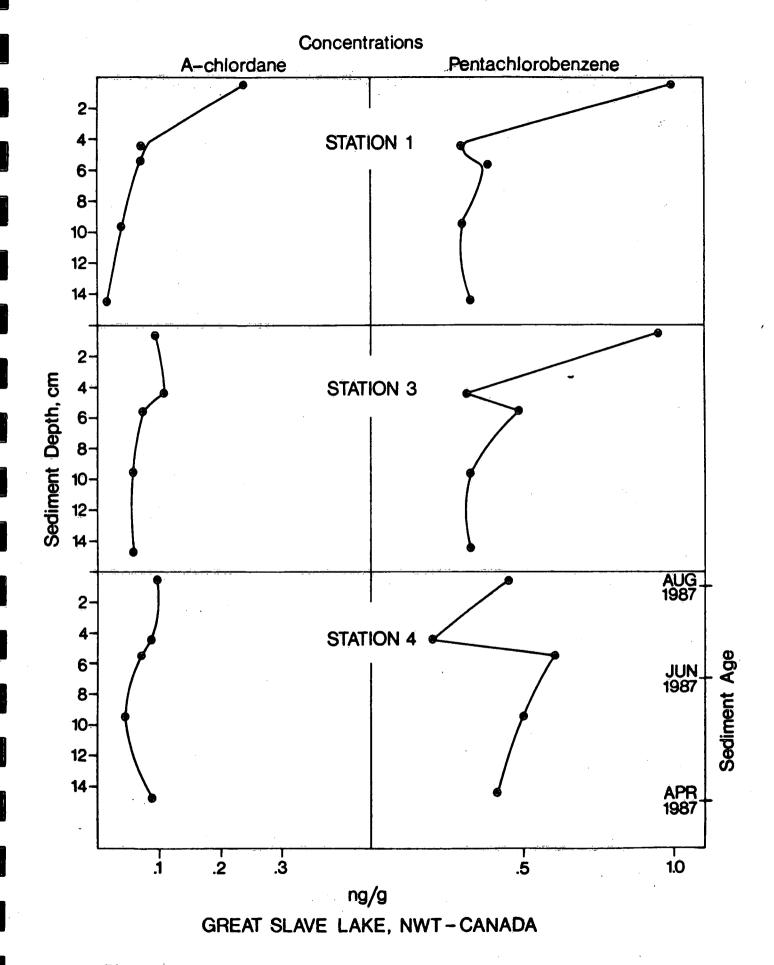
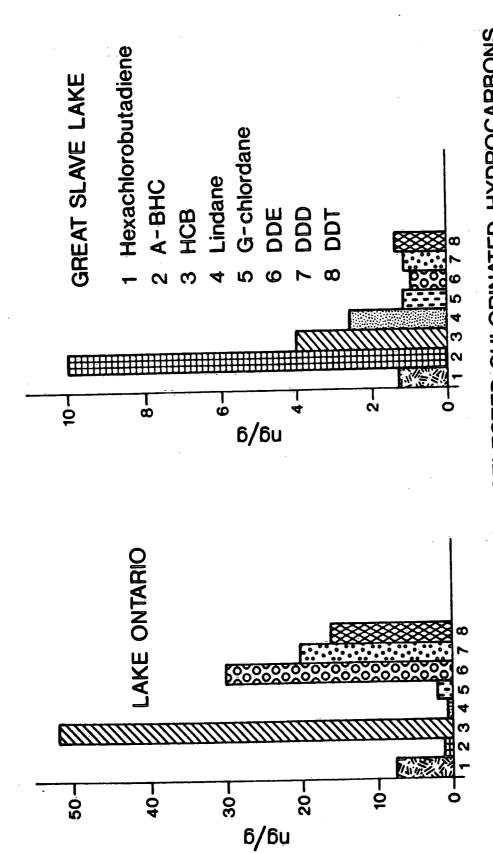


Figure 5



CONCENTRATION PATTERN OF SELECTED CHLORINATED HYDROCARBONS IN SURFICIAL SEDIMENTS FROM LAKES ONTARIO AND GREAT SLAVE