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TOXIC CONTAMINANTS IN WATERS AND SEDIMENTS OF THE GREAT LAKES

R.J. Allan

NWRI Contribution No. 91-19

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MANAGEMENT PERSPECTIVE

This report was prepared for 4th International Conference on the Conservation and Management of Lakes, held in Hangzhou, Peoples Republic of China, in 1990. The proceedings of this Conference will be published in late 1991 or 1992.

The report is a brief summary that provides highlights of a much longer report titled "An Overview of Toxic Contaminants in Water and Sediments of the Great Lakes", which was released in the spring of 1991 by the Departments of the Environment, Fisheries and Oceans and Health and Welfare.

This extensive and comprehensive report is in three volumes; a Summary and two Technical Volumes and contains information on fish, wildlife and humans. Water and sediment contaminants are discussed in Volume 1.

The major conclusions of this review were the following:

1. The data from the 1980s on water concentrations for toxic metals may be too high due to contamination problems in standard laboratories and inaccurate for toxic organics because of small sample sizes and high detection limits. As a consequence, downward trends in concentrations of toxic metals and organics in water which are now in the low part per trillion range may not be as great as they appear.

2. Analysis of bottom sediments show that concentrations of both toxic metals and organics have declined in recent years. In some cases however, concentrations are now leveling off, perhaps due to continued atmospheric and other non-point source inputs.

3. In the open waters of the lakes, concentrations of toxic metals and organics are presently within the Great Lakes Water Quality Objectives for protection of most sensitive users (usually aquatic biota). Occasionally objectives are exceeded in the connecting channels and nearshore areas. The concentrations of the chemicals reviewed are well within drinking water standards even before processing through drinking water treatment plants.

These government reports can be obtained free of charge from the office of the Regional Director General, Ontario Region, Environment Canada, 25 St. Clair Avenue East, Toronto, Ontario M4T 1M2.

PERSPECTIVES DE LA DIRECTION

Le présent rapport a été préparé pour la Quatrième conférence internationale sur la conservation et la gestion des lacs, tenue à Hangzhou (République populaire de Chine), en 1990. Le compte rendu de cette conférence sera publié à la fin de 1991 ou en 1992.

Le rapport est un bref résumé qui indique les principaux points d'un rapport beaucoup plus volumineux intitulé "Les produits chimiques toxiques dans les Grands Lacs et leurs effets connexes" qui a été publié au printemps de 1991 par Environnement Canada, Pêches et Océans Canada et Santé et Bien-être social Canada.

Ce rapport complet et détaillé se présente en trois volumes : un résumé et deux volumes techniques; il fournit des informations sur les poissons, la faune et les humains. Les contaminants de l'eau et des sédiments sont traités dans le Volume I.

Voici les principales conclusions du rapport :

1. Il est possible que les données des années 1980 sur les concentrations de métaux toxiques dans l'eau soient trop élevées en raison de problèmes de contamination dans les laboratoires ordinaires et imprécis pour les produits organiques toxiques en raison de la petite taille des échantillons et des seuils de détection élevés. Donc, des tendances à la baisse des concentrations de métaux toxiques et de produits organiques dans l'eau qui sont maintenant dans la fourchette inférieure du nombre de parties par 10^{-12} (picogramme) peuvent ne pas être aussi importantes qu'elles paraissent.

2. L'analyse des sédiments de fonds révèle que les concentrations de métaux toxiques et de composés organiques ont diminué au cours des dernières années. Toutefois, dans certains cas, les concentrations se stabilisent, à cause peut-être des apports continus de l'atmosphère et d'autres sources diffuses.

3. Dans les eaux libres des lacs, les concentrations de métaux toxiques et de produits organiques correspondent à l'heure actuelle aux objectifs de qualité de l'eau des Grands Lacs pour la protection des utilisateurs les plus sensibles (en général le biote aquatique). Il arrive parfois que les objectifs soient dépassés dans les voies interlacustres et les zones pré-côtières. Les concentrations des produits chimiques étudiés sont bien en deçà des normes pour l'eau potable même avant leur traitement dans des usines d'épuration de l'eau potable.

On peut se procurer gratuitement ces rapports gouvernementaux en s'adressant à : Bureau du Directeur général régional, Région de l'Ontario, Environnement Canada, 25, avenue St. Clair est, Toronto (Ontario) M4T 1M2

RÉSUMÉ

Au cours des dernières années, des techniques d'analyse sensibles ont été mises au point afin de mesurer de très faibles concentrations de contaminants. Jusqu'en 1980, on possédait peu de données fiables sur les concentrations de métaux et de produits chimiques organiques toxiques dans les Grands Lacs. Grâce aux progrès à l'heure actuelle, nous pouvons maintenant déceler avec fiabilité la présence de métaux, de pesticides et de produits chimiques organiques industriels de l'ordre de partie par 10^{-12}, et dans certains cas, de parties par 10^{-15}. Les concentrations actuelles de produits chimiques toxiques dans les eaux libres des Grands Lacs sont dans la fourchette inférieure du nombre de parties par 10^{-12} et sont en deçà des objectifs établis dans le cadre de l'Accord canado-américain relatif à la qualité des eaux des Grands Lacs. Ces objectifs, fixés afin de protéger l'utilisateur le plus sensible (le biote aquatique) sont bien en deçà des normes canadiennes et internationales relatives à l'eau potable. Des analyses des sédiments de fonds des lacs révèlent que l'apport maximal de produits chimiques organiques de synthèse et de métaux lourds est survenu dans les années 1960 et 1970, suivi par des baisses dans les années 1980. Ces diminutions témoignent d'un renforcement des règlements relatifs à l'utilisation de pesticides organochlorés, des BPC, du plomb et du mercure. D'après une évaluation générale des concentrations de produit chimique toxique dans les eaux et les sédiments, les lacs Ontario, Michigan et Érié étaient les plus pollués avec les concentrations les plus fortes de produits chimiques toxiques et persistants. Les lacs Supérieur et Huron étaient les moins pollués, à l'exception de quelques secteurs locaux.

TOXIC CONTAMINANTS IN WATERS AND SEDIMENTS OF THE GREAT LAKES[1]

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ABSTRACT

In recent years, sensitive analytical techniques have been developed to measure very low concentrations of contaminants. Until 1980, there were few reliable data on concentrations of toxic metals and organic chemicals in Great Lakes waters. Improvements now permit the reliable detection of metals, pesticides and industrial organic chemicals in parts per trillion and in some cases parts per quadrillion. The present concentrations of toxic chemicals in the open waters of the Great Lakes are in the low parts per trillion range and are within the objectives of the Canada-U.S.A. Great Lakes Water Quality Agreement. These objectives, set to protect the most sensitive user (aquatic biota) are well below Canadian and international drinking water standards. Analyses of lake bottoms sediments indicate that the peak input of synthetic organic chemicals and heavy metals to the lakes occurred in the 1960's and 1970's followed by declines in the 1980's. These declines reflect the increased regulation of the use of organochlorine pesticides, PCBs, lead and mercury. Based on an overall appraisal of toxic chemical concentrations in waters, and sediments, Lakes Ontario, Michigan and Erie were the most contaminated with the highest concentrations of toxic and persistent chemicals. Lakes Superior and Huron were the least contaminated, with the exception of some local areas.

INTRODUCTION

The Great Lakes constitute the largest body of freshwater on earth in spite of the size of these water bodies (Figure 1 and Table 1), man has contaminated all of them with toxic chemicals. Some toxic chemicals from agricultural, urban, and industrial sources are distributed throughout the entire Great Lake's ecosystem.

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[1]Paper presented at "Hangzhou '90", the 4th International Conference on the Conservation and Management of Lakes, Hangzhou, People's Republic of China, 1990.

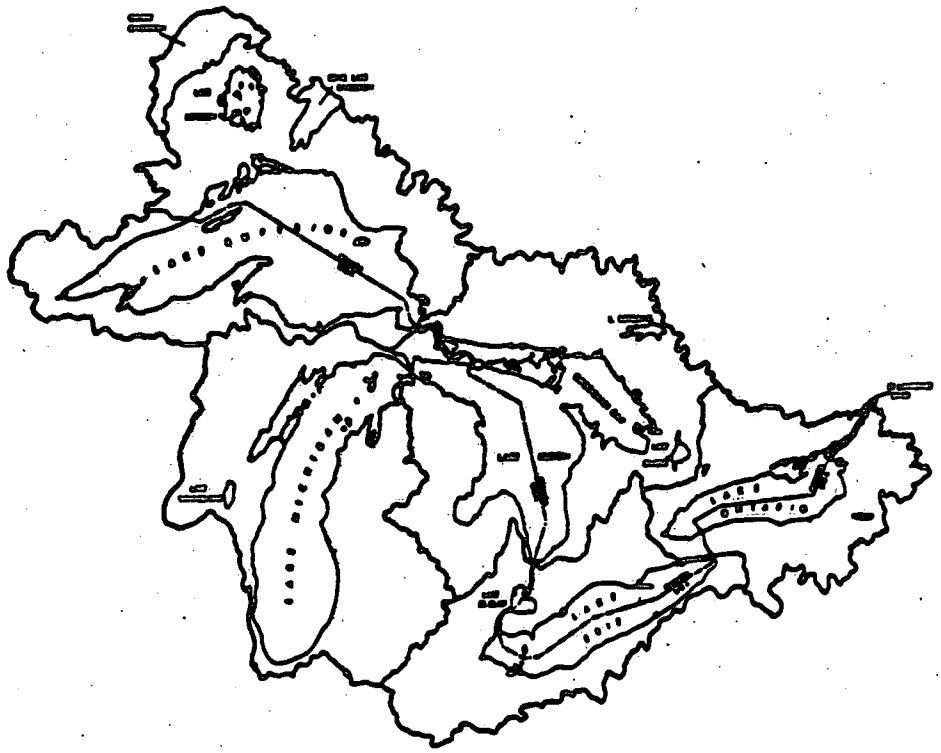

Figure 1 The Great Lakes Drainage Basin

The available information on this toxic chemical contamination of the Great Lakes, although far from complete, is greater than exists for any other set of freshwater lakes in the world (Allan, 1988). Several comprehensive studies of toxic chemicals in various sections of the Great Lakes (Allan et al., 1983; Niagara River Toxics Committee Report (NRTC), 1984; Chau et al., 1985; Lawrence, 1986; Upper Great Lakes Connecting Channels Study (UGLCCS, 1989) have added significantly to a rapidly expanding database over the last decade. Much earlier studies, for example, between 1968 and 1975, of bottom sediment surveys of some toxic metals and organic chemicals on all of the lakes and the composite geochemical maps produced were extremely valuable for making long-term comparisons of the pollution status of the lakes (Thomas and Mudroch, 1979; Thomas, 1981; Thomas and Frank, 1983). Mudroch et al. (1988) summarized sediment concentrations of some metals and PCBs in each lake. During the early 1980's, toxic metals in the water column of each of the Great Lakes were progressively more accurately measured (Rossmann, 1983, 1984, and 1986). Recently, Stevens and Neilson (1989) have used large volume water samples from Lakes Superior, Huron, Erie and Ontario for toxic organic chemical measurements. Strachan and Eisenreich (1988) selected the

	Superior	Michigan	Huron	Erie	Ontario	Totals
Elevation (km)	183	176	176	173	74	
Length (km)	563	494	332	388	311	
Breadth (km)	257	190	245	92	85	
Average Depth (m)	147	85	59	19	86	
Maximum Depth (m)	405	281	229	64	244	
Volume (km^3)	12,100	4,920	3,540	484	1,640	22,684
Area:						
Water (km^2)	82,100	57,800	59,600	25,700	18,960	244,160
Land Drainage Area (km)	127,700	118,000	134,100	78,000	64,030	521,830
Total (km^2)	209,800	175,800	193,700	103,700	82,990	765,900
Shoreline Length (km)	4,385	2,633	6,157	1,402	1,146	17,017
Retention Time (years)	191	99	22	2.6	6	
Population:						
U.S. (1980)	558,100	13,970,900	1,321,000	11,347,500	2,090,300	29,287,800
Canada (1981)	180,440	-	1,051,119	1,621,106	4,551,875	7,404,540
Totals	738,540	13,970,900	2,372,119	12,968,606	6,662,175	36,692,340
Outlet	St. Mary's River	Straits of Mackinac	St. Clair River	Niagara River Welland Canal	St. Lawrence River	

Table 1 Physical Features of the Great Lakes (after Botts and Krushelnick, 1989)

best toxic chemical concentrations in Great Lake water and sediments to calculate mass balances for each lake. These references are very selective. A comprehensive listening is available in Allan and Ball (1990). This paper discusses toxic chemical concentrations in the waters and sediments of the open Great Lakes. Spatial and temporal trends in these abiotic media also discussed where possible.

BRIEF HISTORY OF CONTAMINATION

The recent history of toxic chemical contamination events in the Great Lakes began in the early 1960's with toxic metals, such as Hg and Pb, and pesticides such as DDT and dieldrin, followed by detection of other organochlorine pesticides and then industrial organic chemicals such as PCBs (Williams, 1975; D'Itri, 1988). In 1968, mercury (Hg) was detected in high concentrations in sediments from Lakes Ontario and Huron (Thomas, 1972; 1973). Concurrently, high concentrations of Hg were discovered in Lake St. Clair fish (Fimreite et al., 1971). In 1971, the Michigan Public Health Department issued a warning on PCBs in lake trout and salmon in Lake Michigan. Soon thereafter, the insecticide mirex, was discovered in fish in the

Bay of Quinte in Lake Ontario (Kaiser, 1974). In late 1979, chlorinated dioxins were detected in herring gull eggs from colonies in Saginaw Bay, Michigan. In 1982, toxaphene was detected in fish from Lake Superior. Between these major localised chemical crises, subsequent sampling revealed that most of these chemicals were in fact found throughout the entire Great Lakes system.

DISCUSSION

Although the toxic chemicals referred to here are those which have caused most public concern about contamination of the lakes, they are also those traditionally analyzed for because of the evolving state of analytical methodology for sediments, versus biota, versus water. Since 1980, the collection of suspended particulates has permitted easier detection of toxic metals and hydrophobic toxic organic chemicals in the water column. However, at the low very suspended solids concentrations common throughout the Great Lakes system, even the most hydrophobic organic chemicals are expected to be primarily in the dissolved phase (Allan, 1986).

Reliable, quantitative data for toxic contaminants in water are still scarce. Large volumes of water (200 litres) are presently extracted to detect concentrations in the low ppt and ppq ranges. Present detected dissolved concentrations in water are often an order of magnitude or more beneath former routine detection levels. The values for whole water, and especially for the dissolved phase, are highly variable in space and time and must still be viewed with caution as they were, and still are, often near the limits of analytical detection. Also, because of the complex, time consuming and costly procedures for sample collection, preparation and analyses, few measurements for toxic organic chemicals have been made in all parts of the Great Lakes system.

The historical dissolved toxic metals data for waters from the open lakes is now questioned because metals are ubiquitous in sampling platforms and in laboratories in dust. There is increasing evidence for Pb and Cd, but also for other metals, that the higher historical water concentrations are thus an artifact of contamination rather than the true ambient levels. This means that the true historical water phase toxic metal concentrations in the Great Lakes were probably far lower than previously reported. Because of these analytical developments, there are questions as to the validity of comparing historical with recent toxic metals concentrations in water. However, bottom sediment and most suspended, particulate, toxic chemical concentrations are considered reliable for both toxic metals and organic chemicals. Of particular value, sectioned, radiodated

sediment cores have revealed the long term trends in chemical inputs to the lakes since the depth of coring can be extended to pre-industrial and even to pre-colonial times.

The requirement to detect extremely low concentrations in water has been driven mostly by an objective to understand the relationship of chemical concentrations in water to those in fish rather than from drinking water considerations. The sediment analyses have been driven by a need to include the several orders of magnitude higher concentrations of toxic chemicals in bottom sediments versus water in cause-effect linkages and to quantify the burial of chemicals in lake sediments as a self-cleansing process. Both water and sediment analyses are a requirement to model chemical fate or concentrations for mass balances so as to determine major sources of toxic chemicals to the lakes and thus to prioritize toxic chemical control actions.

The following sections represent the basic conclusions contained in the much longer report (Allan and Ball, 1990) referred to earlier. This longer report contains many complex tables of basic data and other summary diagrams on which these conclusions are based.

Toxic Chemical Concentrations in Water

There is considerable variation in toxic chemical concentrations in waters throughout the Great Lakes. The total concentrations of metals in water, from highest to lowest, were Lake Michigan, Lake Erie, Lake Ontario, Lake Huron, and Lake Superior (Rossman and Barres, 1988). In the mid-1980's, Lake Ontario generally had higher concentrations of toxic organic chemicals in whole water than Lakes Superior, Huron, and Erie (Stevens and Neilson, 1989). The mean concentrations of CBs and lindane ranked highest in Lake Ontario. Concentrations of PCBs, p,p'-DDE, CBs and a-BHC were higher in Lakes Ontario and Erie than Lakes Superior and Huron, while water concentrations of CBs, dieldrin, and PCBs were similar in Lakes Superior and Huron. Dieldrin, lindane, and toxaphene were found to be ubiquitous in lake waters. Mirex is generally not detected in waters in any of the Great Lakes. Because of the historical problems with contamination for toxic metals and detection limits for the toxic organics, only a few illustrative values for water are given in Table 2. Perhaps of greater relevance is a comparison of toxic chemical water concentrations in Lake Ontario with water quality and drinking water guidelines (Table 3), which shows that the ambient water levels are far lower than the drinking water guidelines.

LAKE	CONTAMINANT		
	Pb	Cd	PCBs
	ppt (ng/l)		
SUPERIOR	770 (1977)	120 (1977)	1.8 (1979)
	39 (1983)	27 (1983)	0.4 (1986)
HURON	38 (1980)	16 (1980)	0.9 (1980) 0.6 (1986)
ERIE	1,180-26,310 (1979) 910 (1981)	370-10,660 (1979) 98 (1981)	<100 (1981) 1.4 (1986)
ONTARIO	310-400 (1979) 40 (1985)	<1,000-15,000 (<1981) 96 (1985)	2-50 (1981) 0.4-5.2 (1986)

[1]These values are extracted from very large databases. They are "illustrative" only and should be treated as such. Most are 'means' or 'medians' of many measurements.

Table 2 Illustrative Toxic Contaminant Concentrations in Canadian Great Lakes Open Water

Bottom Sediment Concentrations

In the bottom sediment surveys of the late 1960's-early 1970's (Thomas and Mudroch, 1979), toxic metal concentrations of Pb, Hg, and Cd in the lower lakes (Ontario and Erie) were higher over greater areas of the lake bottom than for the three upper Great Lakes. Pb enrichment in Lake Superior was less than for the other Great Lakes. Information about As concentrations in Great Lakes sediments is scarce. In 1980, levels of HCB in the sediments were much higher in Lake Ontario than in Lakes Superior, Huron and Erie. PAH concentrations in sediments are about 10 times lower in Lake Superior than the other Great Lakes, reflecting both its remote location and the primarily atmospheric source of these chemicals. There has been an indication of recent declines in the level of B(a)P in Lake Superior. Lake Michigan has areas of high levels of PAH concentrations in the bottom sediments. The main source of dioxins and furans to the Great Lakes sediments is through the atmosphere via municipal and/or industrial combustion of chlorinated organic products present in various wastes. There was a predominance of OCDD and HpCDF in bottom sediments. The highest concentrations of OCDD were in Lake Ontario followed by Lake Erie and then Lakes Michigan and Huron. Lake Ontario had elevated levels of OCDF.

(all concentrations in ppt or ng/L)

CHEMICAL	CONCENTRATION IN LAKE ONTARIO WATER	IJC (WATER) GUIDELINES[1]	SOME DRINKING WATER GUIDELINES			
			EPA	CANADA	GNOE	WHO
TOTAL PCBs	1.4 (1986 mean, n=31)	100	-	-	3,000[2]	-
TOTAL DDT	0.12 (1986 median, n=14)	3	-	30,000	30,000	1,000,000
DIELDRIN	0.33 (1986 mean, n=31)	1[3]	-	700	700	30
LINDANE	1.3 (1986 mean, n=31)	10	-	-	-	4,000[4]
HCB	0.06 (1986 mean, n=31)	DL[5]	50,000-175,000	-	-	10
B(a)P	0.3 (estimate < 1986)	10	-	10	-	-
Toxaphene	ND (1983, n=14)	8	5,000	5,000	-	-
TCDD	ND (1984, Niag.R, n=14)	0.01[6]	1[7]	-	-	-
Pb	28 (1985 median, n=22)	2,000-5,00	-	50,000	50,000	50,000
Hg	10 (1985 median, n=22)	200	2,000	1,000	1,000[8]	1,000

Notes:

1. The IJC Guidelines are for protection of the most sensitive aquatic life in the lakes and thus much lower than drinking water guidelines.
2. Not detected in 1,020 Ontario drinking water samples[9] at a detection limit of 20 ppt.
3. Aldrin/dieldrin total.
4. Found in 20% of Ontario drinking water at concentrations of up to 11 ppt.
5. Detection limit.
6. 2,3,7,8-TCDD.
7. One day child for 2,3,7,8-TCDD.
8. Maximum value of 687 Ontario drinking water samples was 380 ppt.
9. Samples referred to here and in 4 and 8 are not necessarily from the Great Lakes.

Table 3 Toxic Chemical Concentrations in the Great Lakes Compared to IJC Ambient Water Quality and Various Drinking Water Guidelines

Toxic Chemical Trends

Trends for toxic chemicals in water and sediments are downwards relative to the concentrations measured ten to twenty years ago. Concentrations in lake bottom sediment cores (Figures 2 and 3) indicate that the major influx of toxic metals and persistent toxic organic chemicals to the Great Lakes aquatic ecosystem took place in the nineteen sixties and seventies. These peak concentrations were followed by declines into the early eighties. Some of these declines have been dramatic once control action has taken place, for example, PCBs and mirex in Lake Ontario. As revealed by the cores, these periods of peak inputs of chemicals to the lakes often occurred prior to their first detection in the lake. Based on an assessment on overall concentrations of toxic and persistent chemicals in abiotic aquatic media, the most chemically contaminated Great Lakes were Lakes Ontario, Michigan, and Erie. Lake Superior was and is the least contaminated of the Great Lakes. Lake Huron is intermediate, being one of the least contaminated of the lakes with the exception of some local areas.

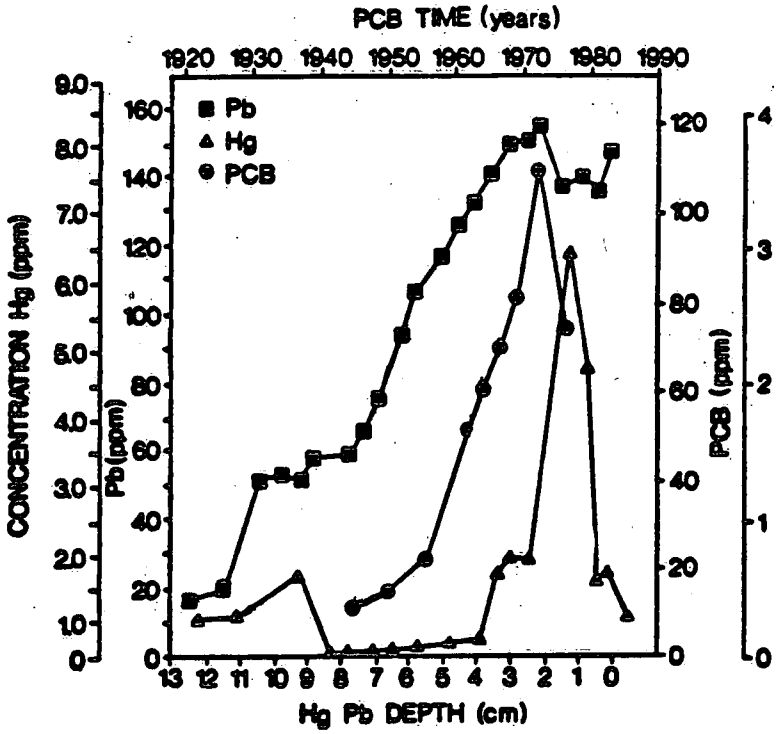

Figure 2 Distribution of Hg, Pb, PCBs in Lake Superior Sediment Cores (After Rossman, 1986; Charles and Hites, 1987)

In Lake Superior water, lead (Pb), mercury (Hg) and cadmium (Cd) concentrations cannot be used to determine trends. Arsenic (As) concentrations in whole water have declined since the 1970's. There has been a dramatic reduction in PCB concentrations in waters and sediments of Lake Superior since the early to mid-1970's. Pb and Hg are decreasing in the surface sediments, as shown by recent core analyses. In Lake Michigan, historic data for concentrations of Pb, Hg and Cd cannot be used to determine trends in the water. Arsenic concentrations in water have declined since the 1970's. Pb has decreased in the sediments from high levels in the 1950's-1960's and Cd has shown similar declines from highs in the 1940's-1950's. PCBs in sediments have declined from peak levels in the 1960's-1970's. In Lake Huron, historic data for Hg, Cd, and As in water cannot be used to determine trends. Dissolved Pb concentrations have declined. In Lake Huron cores taken prior to 1980, there was surface enrichment of Pb, Hg, Cd and As. In Lake Erie, historic Pb data could not be used to determine trends in the water. However, Hg, Cd and As concentrations in water are decreasing as are those of PCBs. Cores collected in the early 1970's showed anthropogenic increases of Pb, Hg and PCBs. More recent surface sediments show declines in concentrations of Pb,

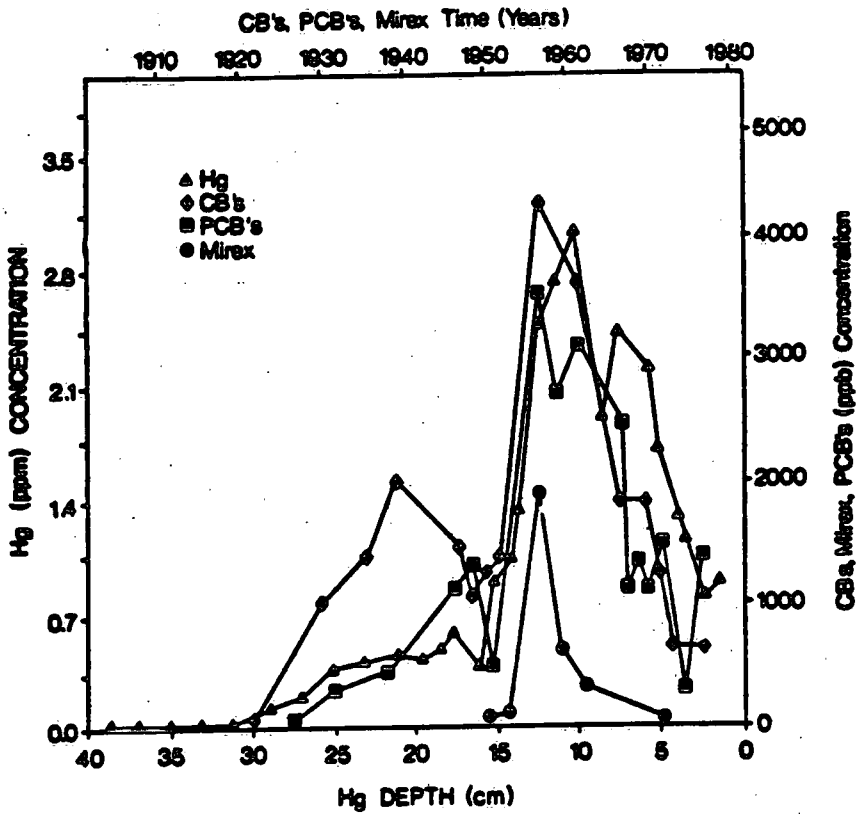

Figure 3 Distribution of Hg, Mirex, CBs, and PCBs in Lake Ontario Sediment Cores (After Durham and Oliver, 1983; Eadie et al., 1983)

As, DDT and metabolites, dioxins and furans. PCBs may not be declining in some surficial sediments. Peak levels of Hg in the sediments of Lake Ontario occurred in the early 1950's. By the end of the 1970's, Hg levels had returned to pre-1940 levels. In Lake Ontario, sediment core analyses have shown that anthropogenic enrichment of Pb, Hg, Cd, DDT, CBs, HCBD, OCs, mirex, PCBs and PAHs in bottom sediments has decreased but is now leveling off.

Present State of the Environment

In the last eight years (since 1982), no other toxic chemicals which fit the criteria of widespread occurrence in all five Great Lakes, high toxicity, and long persistence, similar to most of

the chemicals mentioned above, have been detected. Although other chemicals, for example, highly toxic but rapidly degraded pesticides can be detected, the scale of the lakewide contamination problem in space and time is unlikely to ever be as bad as it once was. In the future, to a greater extent than in the past, toxic chemical control actions to reduce chemical loads to the Great Lakes Basin will have to take into account non-point sources of chemicals, especially atmospheric sources; in-situ contaminated sediment sources; agricultural, forestry, and urban sources; and groundwater sources as well as point sources.

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