The Use of Ambient Monitoring to Estimate the Atmospheric Loading of Persistent Toxic Substances to the Great Lakes

(Phase II of Transportation and Deposition of Persistent Toxic Substances to the Great Lakes Basin: Deposition Monitoring Element)

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by

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### Preface

This report, on the use of ambient monitoring to estimate the atmospheric loading of persistent toxic substances to the Great Lakes, was commissioned by the IJC International Air Quality Advisory Board, the "IJC Air Board".

It is the last in a series of four closely related reports prepared for the IJC Air Board. The first three reports deal with (1) the capability of specific persistent toxic substances to be subjected to long range atmospheric transport; (2) the status and capabilities of associated emissions inventories; and (3) modeling the atmospheric transport and deposition of persistent toxic substances to the great lakes. A summary of the four components has also been prepared.

These reports were prepared as background documents for the IJC-sponsored Joint International Air Quality Board and Great Lakes Water Quality Board Workshop on Significant Sources, Pathways and Reduction/Elimination of Persistent Toxic Substances, to be held May 21-22, in Romulus Michigan. It is expected that the discussion at the Workshop will serve to elaborate upon and extend the analysis presented in this background report.

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## List of Abbreviations

AES         ANR         BVES         CN         COA         DNR         EC         EHD         EPA         HACMP         HQR         IADN         LCS         LMMBS         LVL         NAPS         NWRI         OMOEE         ox         PAH         part.         PCDD/F         pre	National Air Pollution Surveillance Network, Environment Canada National Water Research Institute Ontario Ministry of Environment and Energy oxychlordane polycyclic aromatic hydrocarbons particulate phase of atmosphere polychlorinated dibenzo- <i>p</i> -dioxins and dibenzofurans precipitation precipitation
precip	
	volatile organic compounds

## 1. INTRODUCTION

Atmospheric deposition is an important pathway for the entry of many pollutants to the Great Lakes. This report describes attempts to estimate the net atmospheric loading to the Great Lakes through the use of ambient pollutant measurements. Many examples of these estimates will be discussed in this report. Some of the central examples of these types of estimates include the analyses by Eisenreich *et al.* (1981), Strachan and Eisenreich (1988), Eisenreich and Strachan (1992), and Hoff *et al.* (1996).

The methodology used to estimate atmospheric loading to one or more of the Great Lakes from ambient measurements can be briefly summarized in the following way. First, atmospheric deposition is considered to occur by both wet and dry pathways, i.e., in both the presence and absence of precipitation. Loading from *wet deposition pathway* is estimated from the precipitation rate and the concentration of pollutant in the precipitation. The estimation of loading by the *dry deposition pathway* is somewhat more complicated. In essence, the estimation is based on the concentration of pollutant in the air above the lake, and, as discussed below, for gaseous pollutants (as opposed to pollutant which exists on particles in the atmosphere), the loading estimate also depends on the concentration of pollutant in the water near the surface of the lake. The rate of dry deposition also depends on the meteorological conditions above the lake, and in some cases, the hydrodynamic characteristics of the water near the lake's surface. Reviews of some or all of these phenomena include those by Arimoto (1989), Bidleman and McConnell (1995), Hoff (1994) and Hoff et al. (1996), and Slinn et al. (1978).

The approach used to estimate loadings from ambient measurements is discussed in Section 2 of this report.

## A. Overall Scope of this Analysis

This analysis is primarily limited to U.S. and Canadian government efforts which have monitored one or more of the compounds or groups of compounds listed in Table 1, below, in the Great Lakes region, and/or have used monitoring data to estimate the net atmospheric loading of particular pollutants to the Great Lakes.

In a few cases, particularly relevant non-governmental monitoring/loading efforts and/or efforts for other compounds have been included, but, comprehensive coverage of these other types of analyses was beyond the scope of this analysis.

The analysis began by attempting to assemble the universe of air, precipitation, and water monitoring programs in the Great Lakes region, with basic details about each program (i.e., locations, sampling periods, compounds measured). It turned out that

this effort required a substantial amount of resources. The information presented in this report regarding the universe of monitoring programs relative to the BVES compounds represents the bulk of the work that could be performed in this limited analysis.

Unfortunately, in essentially all cases, it was not possible to obtain and/or analyze monitoring data in the course of this analysis, due to lack of time and/or lack of data availability.

As a general observation, the establishment and maintenance of a clearinghouse for information about and data from monitoring programs in the Great Lakes region would certainly be a useful tool for the public, the research community and the regulatory community.

In the course of this work, it was discovered that there appear to be several parallel efforts underway to assemble the universe of monitoring programs in Canada and the U.S. Details of these efforts were not available for this study, but, it is hoped that they may be available in the future.

Finally, data from two recent research efforts undertaken in the Great Lakes region under the auspices of the U.S. EPA — the AEOLOS project and the Lake Michigan Mass Balance Study — were not generally available for this analysis. These data are being assembled and analyzed by others and will be presented by them elsewhere. It must be noted that these projects would appear to be very relevant to the subject of this report. The public release of the results of these studies is eagerly anticipated.

In addition, two important documents regarding the subject of this report are expected to be released soon but are not currently available. These are:

- Proceedings of the Conference on Atmospheric Deposition to the Great Waters, sponsored by the Air and Waste Management Association, the Great Lakes Center for Environmental Research & Education at Buffalo State College, and the U.S. EPA Great Lakes National Program Office, held October 28-30, 1996, in Niagara Falls, NY.
- Atmospheric Deposition to the Great Lakes and Coastal Waters, edited by Joel Baker, University of Maryland, to be published by the Society of Environmental Toxicology and Chemistry.

Both of these documents are expected to contain very relevant information relative to the subject of this study.

## **B.** Compounds Being Considered

The IJC International Air Quality Advisory Board selected a target list of 27 chemicals or chemical groups to be considered in this analysis, including twelve Level I substances or groups and fifteen Level II substances or groups:

- Level I substances are the 11 Critical Pollutants identified by the IJC's Great Lakes Water Quality Board, plus two additional Critical Pollutant identified by the Lake Superior LaMP and the Lake Ontario Toxics Management Plan (octachlorostyrene and chlordane).
- Level II Substances are those substances identified by the Canada-Ontario Agreement respecting the Great Lakes Basin Ecosystem (COA) as "Tier II" chemicals, plus additional substances of concern identified by LaMP and RAP processes and the Great Lakes Water Quality Guidance in the U.S.

A list of the compounds or groups included is given in Table 1, with the Level indicated in parentheses following the name of the compound.

Table 1. Compounds and Compound Groups Targeted in the Binational Virtual EliminationStrategy (BVES) for Persistent Toxic Substances in the Great Lakes Basin(Envr. Canada and U.S. EPA, 1996) (Level indicated in parentheses)

#### **METALS / ORGANOMETALLICS**

#### Alkylated Lead (I)

including, but not necessarily limited to: tetra-, tri- and di-ethyl lead, tetra-, tri- and di-methyl lead

#### Cadmium and Cadmium Compounds (II)

including, but not necessarily limited to: cadmium, cadmium oxide, cadmium dichloride, cadmium sulfide

#### Mercury and Mercury Compounds (I)

including, but not necessarily limited to: elemental mercury, mercury dichloride, mercury oxide, monomethyl mercury, and particulate mercury

#### Tributyltin Compounds (II)

#### **ORGANOCHLORINE BIOCIDES**

Aldrin / Dieldrin (I) Chlordane (I) DDT / DDD / DDE (I) Endrin (II) Heptachlor / Heptachlor Epoxide (II) Hexachlorocyclohexanes ( $\alpha$ , $\beta$ , $\delta$ , and  $\gamma$ ) (II) Methoxychlor (II) Mirex (I) Pentachlorophenol (II) Toxaphene (I)

#### INDUSTRIAL / MISCELLANEOUS

4-Bromophenyl Phenyl Ether (II) 3,3'-Dichlorobenzidene (II) Hexachloro-1,3-Butadiene (II) 4,4'-Methylene bis (2-Chloroaniline) (II) Octachlorostyrene (I)

#### **CHLOROBENZENES**

1,4-dichlorobenzene (II) Tetrachlorobenzenes (several congeners) (II) Pentachlorobenzene (II) Hexachlorobenzene (I)

# POLYCHLORINATED DIBENZO-P-DIOXINS and DIBENZOFURANS

2,3,7,8-TCDD and 2,3,7,8-TCDF (I) 1,2,3,7,8-PeCDD (I) 1,2,3,4,7,8-HxCDD (I) 1,2,3,6,7,8-HxCDD (I) 1,2,3,7,8,9-HxCDD (I) 1,2,3,4,6,7,8-HpCDD (I) OCDD (I) 1,2,3,7,8-PeCDF (I) 2,3,4,7,8-PeCDF (I) 1,2,3,4,7,8-HxCDF (I) 1,2,3,6,7,8-HxCDF (I) 1,2,3,7,8,9-HxCDF (I) 2,3,4,6,7,8-HxCDF (I) 1,2,3,4,6,7,8-HpCDF (I) 1,2,3,4,7,8,9-HpCDF (I) OCDF (I)

#### POLYCHLORINATED BIPHENYLS (PCB'S)

PCB's (I) [there are 209 PCB congeners]

#### POLYCYCLIC AROMATIC HYDROCARBONS

Benzo[a]Pyrene (I) Dinitropyrenes (several congeners) (II)

plus PAH's as a group (II) including but not limited to: Phenanthrene, Anthracene Benz[a]Anthracene, Perylene Benzo[g,h,i]Perylene

To form a group of PAH's for this analysis, the following additional PAH's were added, consisting of the remaining compounds in the EPA's 16-PAH list & the ATSDR 17-PAH list:

Naphthalene, Acenaphthene Acenaphthylene, Fluorene, Pyrene Fluoranthene, Chrysene, Benzo[b]Fluoranthene, Benzo[j]Fluoranthene Benzo[k]Fluoranthene, Benzo[e]Pyrene Dibenz[a,h]Anthracene, Indeno[1,2,3-c,d]Pyrene

## 2. OVERVIEW of the USE of AMBIENT MONITORING of PERSISTENT TOXIC SUBSTANCES to ESTIMATE NET ATMOSPHERIC LOADINGS to the GREAT LAKES

As discussed above, briefly, in the introduction, in the use of ambient monitoring data to estimate loadings of pollutants to the Great Lakes through the atmospheric pathway, both wet and dry deposition phenomena are considered.

## A. Wet Deposition

To estimate wet deposition flux, pollutant concentrations measured in sampled precipitation are multiplied by precipitation rates, e.g. (with one possible set of a consistent units)<sup>1</sup>:

(1a)	["L <sub>wet</sub> ",	wet deposition loading <i>(g/year)</i> ] =
		[" $C_p$ ", concentration of pollutant in precipitation (g/m <sup>3</sup> )]
	х	["R <sub>p</sub> ", precipitation rate ( <i>m</i> /year)]
	х	["A", area of lake $(m^2)$ ].

Using the symbols defined in the above relation, the loading can be expressed as:

(1b)  $L_{wet} = C_p R_p A$ 

Examples of wet deposition flux estimates to the Great Lakes are included in analyses by Eisenreich *et al.* (1981), Strachan and Eisenreich (1988), Gatz et al. (1989), Eisenreich and Strachan (1992), Voldner and Alvo (1993), Chan *et al.* (1994), and Hoff *et al.* (1996).

## B. Dry Deposition: Particle-Phase Material

To estimate dry deposition flux from ambient measurements, vapor and particleassociated fractions of a given pollutant are typically considered separately. First, the fraction of a given pollutant that is expected to exist in the vapor phase in the atmosphere, and the fraction that is expected to exist in the particle phase are estimated, either from direct measurement of theoretical considerations.

<sup>&</sup>lt;sup>1</sup>. For the reader's convenience, terms used by Hoff *et al.* (1996) are given the same symbol in this analysis. For terms not used by Hoff *et al.* (1996), a closely related or new symbol is used, as appropriate.

The particle-phase fraction of a given pollutant is generally considered to be deposited at a rate defined by an assumed deposition velocity, and the deposition rate is estimated using ambient measurements, e.g., (with one possible set of consistent units):

(2a)	["L <sub>drv.pa</sub>	<sub>art</sub> ", particle-associated dry deposition loading <i>(g/year)</i> ] =
	5,11	["C <sub>a</sub> ", total concentration of pollutant in atmosphere $(g/m^3)$ ]
	х	$[``\phi_a"$ , fraction of atmospheric pollutant associated with atmospheric
		particles (dimensionless)]
	v	[" $v$ ," dependition valuative (m(vacr)) (generally on ecourted value)]

x [ "v<sub>d</sub>", deposition velocity (*m*/year) (generally an assumed value)]

x ["A", area of lake, 
$$(m^2)$$
]

Using the symbol defined in the above relation, the loading can be written as:

(2b) 
$$L_{dry,part} = C_a \phi_a v_d A$$

Examples of approaches for estimating the dry deposition flux of particleassociated pollutants include those by Hicks and Williams (1980), Slinn and Slinn (1980), Williams (1982) and Schmidt (1982). Discussions of the appropriate value to use for the average deposition velocity for particle phase material are given by Eisenreich *et al.* (1981), Strachan and Eisenreich (1988), Eisenreich and Strachan (1992), and Hoff *et al.* (1996).

As is frequently noted, this approach is regarded as somewhat uncertain, as the exact "average" value of the dry deposition velocity for particulate phase material is not known. Further, as the dry deposition velocity will obviously depend on the particle size distribution in the atmosphere, and the size distribution for the particle-associated material of different pollutants will generally vary from pollutant to pollutant, the use of a single average deposition velocity for all particulate pollutants is obviously somewhat of an oversimplification.

In most of the ambient air monitoring that takes place in the Great Lakes region, particle size distributions are not routinely measured. Measurements of pollutant-specific particle size distributions are even less common.

## C. Dry Deposition: Vapor-Phase Material ("Gas Exchange")

The estimation of the deposition rate (or volatilization rate) of the vapor-phase fraction of a given pollutant to a lake is generally estimated in the following way. The net direction and driving force for the flux of pollutant is considered to result from the degree of departure from thermodynamic equilibrium between the air and water near the lake surface. This equilibrium is generally assumed to be governed by Henry's Law, a commonly used convention with trace, volatile species in water-air systems. For a given pollutant, the air and water phases are considered to be at equilibrium if the

concentrations satisfy the following condition (again, with one possible set of consistent units):

- (3a) [" $C_{w,dissolved}$ ", truly dissolved water phase concentration of pollutant in the water below the surface  $(g/m^3)$ ] =
  - [" $C_{a, vapor}$ ", vapor phase concentration of pollutant in air above the water surface  $(g/m^3)$ ]
  - x ["R", gas constant ( $Pa-m^3 / mol {}^{\circ}K$ )]
  - x ["T", temperature  $(^{\circ}K)$ ]
  - ÷ ["H", Henry's Law coefficient for the pollutant (*Pa-m<sup>3</sup>/mole*)]

Similar to the situation in the atmosphere, a pollutant can exist associated with particles in the water (i.e., suspended sediment) or in a truly dissolved state. It is the truly dissolved pollutant that is relevant to the above thermodynamic equilibrium condition. Using the symbols defined in the above relation, the equilibrium condition above can be defined expressed as:

(3b) 
$$C_{w,dissolved} = C_{a,vapor} R T / H$$

When the actual pollutant concentrations in the air and water do not satisfy the above equation, the system is said to depart from equilibrium, and the degree of departure, or the *thermodynamic driving force* due to the concentration imbalance, " $\Delta_c$ ", is expressed by the difference between the two sides of "equation" 3b above:

(4) 
$$\Delta_c = C_{a, vapor} (RT / H) - C_{w,dissolved}$$

As is typically done in many mass transfer situations, the net rate of flux in a non-equilibrium situation is estimated as the product of the thermodynamic driving force and a mass transfer coefficient,<sup>2</sup> e.g.,

(5)  $L_{dry,vapor} = \Delta_c K_{OL} A = [C_{a,vapor} (RT/H) - C_{w,dissolved}] K_{OL} A$ 

where  $K_{oL}$ , the mass transfer coefficient has units of m/yr (to be consistent with the above discussion).  $K_{oL}$  depends on the degree of mixing and diffusion on both sides of the surface, i.e., in both the water phase and the air, and is often parameterized as a function of the wind speed at a particular height above the water surface.

It can be seen from equation (5) above that the net flux of dry, vapor "deposition" can be positive (from the air to the water) or negative (from the water to the air),

<sup>&</sup>lt;sup>2</sup>. An analogous approach is taken in heat transfer situations, where a heat transfer coefficient is used.

depending on the relative concentrations of a given pollutant in the vapor and truly dissolved in the liquid phase.

There have been many studies that have attempted to estimate the direction and rate of gas exchange of different pollutants with one or more of the Great Lakes. Examples include:

- Hoff et al. (1996): a range of compounds measured in the IADN program;
- Hoff *et al.* (1993): toxaphene;
- Achman *et al.* (1993), Jeremiason *et al.* (1994), Hornbuckle *et al.* (1995), and Honrath *et al.* (1997): PCB's;
- Baker and Eisenreich (1990): PAH's and PCB's;
- McConnell *et al.* (1993) and Ridal *et al.* (1996):  $\alpha$ -HCH and  $\gamma$ -HCH.

Bidleman and McConnell (1995) have recently reviewed the gas-exchange phenomenon.

## D. Droplet Resuspension

While it is somewhat poorly understood at the present, it is also possible for water droplets to be "ejected" from the lake. While some of these droplets would so large that they would quickly fall back into the lake, some are small enough to be carried aloft into the atmosphere above the lake. The water in these droplets will strive to reach thermodynamic equilibrium with the water vapor in the ambient air (as characterized, for example, by the relative humidity) and most or all of the water in the droplets will evaporate relatively quickly. Much of the inorganic and organic material contained in the droplets when they were first formed will remain in the new aerosol particles, even after the water has partially or completely evaporated. These new particles, then, can be seen as a way in which pollutants in the lake can be resuspended into the atmosphere, in a particle-related fashion. A qualitative "term" is added to the loading equation to represent this phenomena.

Most estimates of atmospheric loading do not currently attempt to make quantitative estimates of this phenomenon.

## E. Fog Deposition

Another rather poorly characterized deposition phenomenon is that related to fog. The depositional behavior of fog droplets will be different from particle-phase deposition under non-fog conditions. It is unlikely that fog deposition is reliably

measured in conventional precipitation sampling programs. A qualitative "term" is added to the loading equation to represent this phenomena.

Most estimates of atmospheric loading do not currently attempt to make quantitative estimates of this phenomenon.

## F. Indirect Atmospheric Loading

In addition to the direct atmospheric loading to a given lake, it is recognized that atmospheric deposition to land areas in a lake's watershed can contribute, indirectly, to the lake's pollutant loading. Pollutants deposited in the watershed can be washed by precipitation runoff directly to a lake or to a tributary which empties into a lake. It is believed that these indirect processes are probably less important than the direct deposition phenomena described above. However, much less is known about indirect atmospheric loading, and it is difficult to even make semi-quantitative estimates at this time.

Most estimates of atmospheric loading do not currently attempt to quantitatively include this phenomenon.

## G. Overall Atmospheric Loading

The total, direct, net atmospheric loading to a given lake or lake subsection is the sum of the wet and (net) dry deposition amounts estimated above, i.e.:

(6a)  $L_{total,direct} = L_{wet} + L_{dry,part} + L_{dry,vapor} + [fog deposition] - [droplet resuspension]$ 

(6b) 
$$L_{total,direct} = A \{ C_p [R_p] + C_a [\phi_a v_d + (1-\phi_a)(K_{OL}RT/H)] - C_{w,dissolved} [K_{OL}] \} + [fog deposition] - [droplet resuspension]$$

A particle-associated fraction  $\phi_w$  in the water phase can be defined as in the air phase, above, and  $C_w$  can be defined as the total water concentration, analogous to the total air concentration Ca. Using these definitions, equation 6b can be rewritten as:

(6c)  $L_{total,direct} = A \{ C_p [R_p] + C_a [\phi_a v_d + (1-\phi_a)(K_{OL}RT/H)] - C_w (1-\phi_w)[K_{OL}] \} + [fog deposition] - [droplet resuspension]$ 

Adding a "conceptual" term to represent indirect loadings and slightly rearranging equation 6, above, the following expression is obtained, giving the total atmospheric loading to a lake:

- (7)  $L_{total} = A R_p C_p$ (wet deposition)
  - +  $A \phi_a v_d C_a$ (dry deposition of particle-phase material)
  - + "fog deposition"
  - "pollutant losses from the lake due to droplet resuspension"
  - +  $A K_{OL} \{ [ (1-\phi_a) (RT/H) C_a ] [ (1-\phi_w) C_w ] \}$ (net deposition of vapor-phase material)
  - + "indirect atmospheric loadings"

Except for the gas constant (R), and the area of the Lake  $(A)^3$ , all of the parameters in the above equations will be time- and location-dependent. That is, at any given time, the following parameters will vary from place to place on a given lake, and, at any given location, each will vary over time:

- (a) the concentration of the pollutant in precipitation  $(C_p)$ , air  $(C_a)$ , and water  $(C_w)$ ;
- (b) the partitioning behavior of the pollutant in the air and water phases, expressed in the above equations as  $\phi_a$  and  $\phi_w$ , the particle-associated fractions in the air and water phases, respectively.
- (c) meteorological variables, such as precipitation rate, temperature (which appears directly in the equations, and, which influences H), and wind speed (which influences  $K_{OL}$  and  $v_d$  and droplet resuspension phenomena);

Ambient measurements cannot obviously be made at every location in the air and water near the surface of a given lake. Thus, one issue that arises in applying the above methodology is the extent to which a given set of measurements "captures" enough of the spatial variations to allow an accurate estimate for a given lake or lake portion. For example, if measurements at only one location are made and used to estimate the net atmospheric deposition to a given lake, the question obviously arises

<sup>&</sup>lt;sup>3</sup>. Obviously, the area of the lake or lake subsection being considered can change, but, the *magnitude* and *rate* of these changes are relatively small compared to the changes in essentially every other parameter involved.

as to how representative of the "average" the measurements are. The same questions arise even when multiple measurement locations are used.

Moreover, while meteorological measurements at a given site can be made more or less continuously, measurements of chemical concentrations at a given site tend to be made only periodically. Thus, when measurements at only specific times are used, an analogous question arises regarding the extent to which the measurements of any parameter are representative enough to construct accurate time-averages.

Thus, the degree of accuracy of the above methodology will depend in detail on the representativeness of the measurements.

In Table 2, below, the parameters which are typically used to estimate the atmospheric loading to the Great Lakes (or any lake, for that matter) are presented, along with a note about how the parameters are obtained.

This report will discuss various aspects of the loading equations above and the parameters summarized below, including:

- the extent to which ongoing measurement programs provide data for loading estimates;
- strengths and weaknesses (i.e., challenges) in making estimates of atmospheric loadings; and
- estimated atmospheric loadings to the Great Lakes.

	Table 2. Parameters Typically Used to Estimate the Net Atmospheric Deposition to a Given Lake or Lake Area (all the parameters below will vary in time and space; thus, averages are used)					
	Parameter	How Obtained (in typical situation)				
n of Gas and Pollutant	Concentration of the Pollutant in Precipitation	Measured				
Wet Deposition of Gas and Particle Phase Pollutant	Precipitation Rate	Measured				
llutant	Concentration of the Pollutant in the Air Near the Lake Surface	Measured				
Dry Deposition of Particle-Phase Pollutant	Vapor/Particle Partitioning Characteristics	Sometimes measured, sometimes estimated. Estimates depend on physical/chemical properties of pollutant, temperature, the nature of the atmospheric aerosol, and the degree to which vapor/particle equilibrium is achieved.				
Dry De Particle	Dry Deposition Velocity of Particle- Associated Pollutant	Typically estimated; often a constant value is assumed				
	Concentration of the Pollutant in the Air Near the Lake Surface	Measured				
on Flux Pollutant	Vapor/Particle Partitioning Characteristics	Sometimes measured, sometimes estimated. Estimates depend on physical/chemical properties of pollutant, temperature, the nature of the atmospheric aerosol, and the degree to which vapor/particle equilibrium is achieved.				
oosition lase Poll	Concentration of Pollutant Truly Dissolved in the Water Near the Lake Surface	Measured, or estimated from the total concentration of the pollutant measured in the water				
Net Dry Depositi of Vapor Phase	Henry's Law Constant	Based on existing laboratory measurements; temperature dependent				
Net I of Va	Temperature	Measured				
	Air-Water Mass Transfer Coefficient	Estimated, using correlation-based semi-empirical theories derived from experimental measurements. Correlations are often based on the wind speed, measured at a given height above the surface.				

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## 3. MONITORING of AMBIENT AIR and PRECIPITATION for PERSISTENT TOXIC SUBSTANCES in the GREAT LAKES BASIN

There are a number of air and/or precipitation monitoring programs for persistent toxic substances in the Great Lakes region.

The most notable is the Integrated Atmospheric Deposition Network (IADN). IADN was established by the 1987 revision to the Great Lakes Water Quality Agreement, and involves coordination among a number of Federal and State/Provincial agencies in the United States and Canada. A summary of this program is given in the IADN Quality Assurance Program Plan (QAPP) (Environment Canada *et al.*, 1994), and by Sweet *et al.* (1995) and Hoff *et al.* (1996). The program will be briefly summarized here.

Air and precipitation samples are collected at three U.S. and two Canadian master stations. There are two satellite stations in the U.S., at which air and precipitation samples are collected. There are twelve satellite stations operated by one or more Canadian federal or provincial agencies (1 of these is in Minnesota; the rest are in Canada). At the four of the Canadian-managed stations operated by OMOEE, air and precipitation samples are collected. At the seven Canadian-managed satellite stations operated by Environment Canada's Ecosystem Health Division (EHD), precipitation samples are collected. At the satellite station at Egbert operated by Environment Canada's Ecosystem Health Division (EHD), precipitation samples are collected. At the satellite station at Egbert operated by Environment Canada's Centre for Atmospheric Research Experiments (CARE), air samples are collected. Different compounds are monitored in various portions of the IADN program.

Several other monitoring programs in the Great Lakes region were identified in this analysis. The IADN program and these other programs are summarized in Tables 3-11, below.

Table 3 gives a list of the monitoring programs identified, with the number of sites in different proximity ranges to the Great Lakes. The one-letter code denoting each program is used in Tables 4-11.

Table 4 shows the universe of monitoring sites identified in the programs shown in Table 3. The 2-3 character codes denoting the locations are used in Tables 5-11. Also, the codes describing "site character", e.g., R = Rural, are used in tables 7-11.

In assembling the universe of monitoring sites, all sites identified in any State or Province adjoining a Great Lake were included, with Quebec, Manitoba, and Vermont added. Obviously, all things being equal, sites closer to a given Lake will probably be more representative than a site further away. However, the Great Lakes region is so large, sites on the order of 100-200 km away from a given Lake cannot obviously be said to be irrelevant.

Tables 5 and 6 give a summary of the coverage of BVES compounds in the monitoring programs operated by Canadian and U.S. agencies, respectively.

Tables 7 through 11 give a summary for each Great Lake of the coverage of BVES compounds in monitoring locations at the monitoring sites in the vicinity of the Lake.

	Table 3. Air and Precipitation	Monitoring Program	is in the Great Lakes Reg	ion Identified in this A	nalysi	5		
c o	Program	Sponsor or Coordinator	Phase(s) Sampled	BVES Compounds Included	# of sites (*)			
d e					0-10 km	10-100 km	> 100 km	
	U.S. IADN Program							
u	U.S. IADN Sites EPA (IN Univ) vapor, part, precip many							
	Canadian IADN Program							
с	Air Monitoring at Master Sites	EC AES	vapor, part	many	2	0	0	
0	OMOEE IADN Satellite Sites	OMOEE	vapor, part, precip	many	3	1	0	
е	IADN Satellite site at Egbert	EC AES	vapor, part	many	0	1	0	
g	Great Lakes Precip Network	EC EHD	precip	many	8	2	0	
n	IADN precip at Pt. Petre	EC NWRI	precip	many	1	0	0	
	Toxaphene Monitoring in the Gre	at Lakes Region						
b	Research at Pt. Petre	EC	vapor, part	Toxaphene	1	0	0	
h	Research at Eagle Harbor	Indiana Univ.	vapor, part	Toxaphene	1	0	0	
	National Air Pollution Surveilland	e Network, Environr	nent Canada					
t	sites measuring PAH's	EC	vapor + part	PAH's	4	1	4	
t	sites measuring COA substances	EC	vapor + part	OCS, DNP, HCB,PCP	4	1	0	
t	sites measuring VOC's	EC	vapor	VOC's	7	4	11	
t	sites measuring PCDD/F's	EC	vapor + part	PCDD/F	4	1	4	
t	sites measuring metals	EC	particulate	total Cd, Pb, Hg	3	2	5	
	Air Toxics Monitoring Program, C	Ontario Ministry of E	nvironment and Energy					
а	sites measuring PAH's	OMOEE	vapor + part	PAH's	5	2	1	
у	sites measuring VOC's	OMOEE	vapor	VOC's	10	4	1	
р	sites measuring PCDD/F's	OMOEE	vapor + part	PCDD/F	1	1	0	
р	sites measuring PCDD/F's	OMOEE	precipitation (Dorset)	PCDD/F	0	1	0	
	Mercury Monitoring Programs							
d	Mercury Deposition Network	various	precipitation	total mercury	1	5	6	
k	Mercury monitoring (CAMNet)	EC	vapor	total mercury	1	1	2	
s	Mercury: Dorset	OMOEE	vapor, precip	total Hg	0	1	0	
q	Univ Mich Air Quality Laboratory	Univ. Michigan	(?) vapor, part, precip	total mercury (?)	0	1	0	
f	Univ Mich Air Quality Laboratory	"trust fund"	(?) vapor, part, precip	total mercury (?)	1	0	0	
I	Lake Champlain study	(?)	(?) vapor, part, precip	total mercury (?)	0	0	1	
r	Mercury research (1995-1996)	EPA	vapor, part	total mercury	5	1	4	
	Additional Monitoring Programs	in the Great Lakes R	legion					
m	Air toxics monitoring	Michigan DNR	vapor + particulate	ongoing (?)	1	3	0	
v	Haz. Air Contam. Monitoring	Vermont ANR	vapor + particulate	many	0	0	5	
w	Green Bay Urban Air Toxics	Wisconsin DNR	vapor + particulate	many	1	0	0	
ŀ	f information was available, the numbe ‹m of a Great Lake, and the number of <u>Great Lakes, with Manitoba, Quebec, a</u>	sites greater than 100		,				

Table 4. Air and Precipitation Monitoring Sites in the Great Lakes Region Identified in this Analysis (a)											
Site Code		State or	Program(s) at Site	Lat	Long	site type	distance from Lake	orientation to Lake			
<u>(b)</u>	Site Name	Province	(c)	(d)	(d)	(e)	(km) (f)	(g)			
	Si	tes in the	Vicinity of	f Lake Si	uperior	1	1				
EH	Eagle Harbor	MI	uhfr	47.46	88.15	R	0.1	s			
BR	Brule River	WI	u d	46.75	91.61	R	0.4	s			
Sb	Sibley	ON	g	48.50	88.68	(R)	~ 1	n			
WR	Wolf Ridge	MN	0	47.41	91.26	(R)	~ 1	n			
ТВ	Thunder Bay	ON	У	48.40	89.25	U	~ 1	n			
SSM	(2 sites) Sault Ste. Marie	ON	а у	46.65	84.35	U	~ 1	е			
TL	Turkey Lakes	ON	g	47.03	84.38	(R)	~ 25	е			
Fb	Fernberg	MN	d	47.95	91.50	(R)	70	w			
TrL	Trout Lake	WI	d	46.05	89.65	(R)	70	S			
PR	Popple River	WI	d	45.80	88.40	(R)	100	s			
MF	Marcell Expt Forest	MN	d	47.53	93.47	(R)	150	w			
CC	Cedar Creek	MN	r	45.40	93.30	(R)	180	sw			
CR	Camp Ripley	MN	d	46.25	94.50	(R)	200	sw			
Lt	Lamberton	MN	d	44.24	95.30	(R)	380	sw			
Wp1	(65 Ellen St.) Winnipeg	MB	t	49.90	97.15	UC	530	nw			
W p2	(301 W eston St.) W innipeg	MB	t	49.80	97.10	UC	530	nw			
	Sit	es in the	Vicinity of	Lake Mi	chigan						
SBD	Sleeping Bear Dunes	MI	u r	44.76	86.06	R	1	е			
Ch	IIT - Chicago	IL	u	41.83	87.62	U	1.5	w			
GBy	Green Bay	WI	w	44.50	88.00	U	~ 1	w			
SH	South Haven	MI	m	42.40	86.30	(R)	~ 1	е			
Pe	Pellston	MI	m	45.55	84.80	(R)	20	е			
LG	Lake Geneva	WI	d	42.58	88.50	(R)	60	w			
Во	Bondville	IL	dr	40.05	88.37	(R)	200	sw			
WM	W ildcat Mountain	WI	r	43.70	90.57	(R)	230	w			
Sites in the Vicinity of Lake Huron											
BI	Burnt Island	ON	сg	45.83	82.95	R	~ 1	island			
GB	Grand Bend	ON	o g	43.33	81.75	(R)	~ 1	е			
Sa	Sarnia	ON	t	42.98	82.40	UR	~ 1	S			
Dv	Deckerville	MI	m	43.50	82.70	(R)	20	w			
Eg	Egbert	ON	e k (t?)	44.26	79.79	(R)	40	se			
Do	Dorset	ON	оѕру	45.20	78.85	R	90	е			
		Sites in t	he Vicinity	of Lake	Erie						
SP	Sturgeon Point	NY	ur	42.69	79.06	(R)	0.1	s			
PS	Port Stanley	ON	0	42.67	81.17	(R)	~ 1	n			
ΡI	Pelee Island	ON	g	41.97	82.52	R	~ 1	island			
RP	Rock Point	ON	g	42.85	79.55	(R)	~ 1	w			
Sm	Simcoe	ON	t	42.88	80.29	R	~ 1	n			
Na	(2 sites) Nanticoke	ON	а у	42.80	80.08	U	~ 1	n			
We	Welland	ON	а	42.98	79.25	U	10	n			
Lw	Longwoods Conservation Area	ON	t	42.90	81.49	R	30	n			
Wd1	(University Ave) Windsor	ON	рау	42.32	83.04	UC	30	n			
Wd2	(College & South St.) Windsor	ON	t	42.29	83.08	U	30	n			
Wd3	Windsor	ON	а у	42.30	83.10	U	30	n			
SC	St. Clair	ON	g	42.38	82.40	(R)	30	n			
Dx	Dexter	MI	qmr	42.35	83.90	(R)	80	w			
SF	Salt Fork Lake	ОН	r	40.10	81.50	(R)	150	s			
HC	Hill Creek St. Park	PA	d	41.83	77.17	(R)	170	se			
AP	Allegheny Portage Nat'l Hist. Site	PA	d	40.33	78.50	(R)	240	s			

Code (b)         Site Name         or Province         at Site (c)         or (d)         type (d)         from Lake (e)         trope (km) (f)           Sites in the Vicinity of Lake Ontario           PP         Pt. Petre         ON         c n g b k t r         43.83         77.15         R         ~ 0.1           Bu         Burlington         ON         g         43.38         79.85         U         ~ 1            To1         (Evans and Arnold Ave) Toronto         ON         t         43.62         79.53         UI         ~ 1            To2         (Junction Triangle) Toronto         ON         t         43.66         79.39         UC         ~ 1            To3         (Downtown) Toronto         ON         t         43.66         79.39         UC         ~ 1           MZ         (Metro Zoo) Toronto         ON         g         43.87         79.19         (U)         ~ 1           Ha1         Hamilton         ON         py         43.26         79.86         U         ~ 1           Ha2-5         (4 additional sites) Hamilton         ON         t         43.26         79.86         UC         ~ 1           St <th>entation o Lake (g) n w w n n n w w w w w w w w n n</th>	entation o Lake (g) n w w n n n w w w w w w w w n n											
Sites in the Vicinity of Lake Ontario           PP         Pt. Petre         ON         cngbktr         43.83         77.15         R         ~0.1           Bu         Burlington         ON         g         43.38         79.85         U         ~1           To1         (Evans and Arnold Ave) Toronto         ON         t         43.62         79.53         UI         ~1           To2         (Junction Triangle) Toronto         ON         t         43.66         79.39         UC         ~1           To3         (Downtown) Toronto         ON         t         43.66         79.39         UC         ~1           MZ         (Metro Zoo) Toronto         ON         g         43.87         79.19         (U)         ~1           Ha1         Hamilton         ON         py         43.26         79.86         U         ~1           Ha2-5         (4 additional sites) Hamilton         ON         y         43.26         79.86         U         ~1           Ha6         Hamilton         ON         t         43.98         79.27         UR         30           Lo         London         ON         t         43.98         79.27 <t< td=""><td>n w n n n w w w w</td></t<>	n w n n n w w w w											
PP         Pt. Petre         ON         c n g b k t r         43.83         77.15         R         ~ 0.1           Bu         Burlington         ON         g         43.38         79.85         U         ~ 1           To1         (Evans and Arnold Ave) Toronto         ON         t         43.62         79.53         UI         ~ 1           To2         (Junction Triangle) Toronto         ON         t         43.67         79.45         UC         ~ 1           To3         (Downtown) Toronto         ON         t         43.66         79.39         UC         ~ 1           To3         (Downtown) Toronto         ON         t         43.66         79.39         UC         ~ 1           MZ         (Metro Zoo) Toronto         ON         g         43.87         79.19         (U)         ~ 1           Ha1         Hamilton         ON         py         43.26         79.86         U         ~ 1           Ha2-5         (4 additional sites) Hamilton         ON         y         43.26         79.86         UC         ~ 1           Ha6         Hamilton         ON         t         43.98         79.27         UR         30	w n n n w w w w n											
Burlington         ON         g         43.38         79.85         U         ~1         w           (Evans and Arnold Ave) Toronto         ON         t         43.62         79.53         UI         ~1         n           (Junction Triangle) Toronto         ON         t         43.67         79.45         UC         ~1         n           (Junction Triangle) Toronto         ON         t         43.66         79.39         UC         ~1         n           (Metro Zoo) Toronto         ON         g         43.87         79.19         (U)         ~1         n           (Metro Zoo) Toronto         ON         y         43.26         79.86         U         ~1         w           5         (4 additional sites) Hamilton         ON         y         43.26         79.86         UC         ~1         w           5         (4 additional sites) Hamilton         ON         t         43.26         79.86         UC         ~1         w           6         Hamilton         ON         t         43.26         79.86         UC         ~1         w           6         Cornwall         ON         t         43.26         79.86         UC												
To1         (Evans and Arnold Ave) Toronto         ON         t         43.62         79.53         UI         ~ 1           To2         (Junction Triangle) Toronto         ON         t         43.67         79.45         UC         ~ 1           To3         (Downtown) Toronto         ON         t         43.66         79.39         UC         ~ 1           MZ         (Metro Zoo) Toronto         ON         g         43.87         79.19         (U)         ~ 1           Ha1         Hamilton         ON         py         43.26         79.86         U         ~ 1           Ha2-5         (4 additional sites) Hamilton         ON         y         43.26         79.86         U         ~ 1           Ha6         Hamilton         ON         y         43.26         79.86         UC         ~ 1           St         Stouffville         ON         t         43.98         79.27         UR         30           Lo         London         ON         y         43.00         81.25         U         40           Co         Cornwall         ON         a y         45.05         74.70         U         150           Ot1         (R	n n n w w w w											
To2         (Junction Triangle) Toronto         ON         t         43.67         79.45         UC         ~ 1           To3         (Downtown) Toronto         ON         t         43.66         79.39         UC         ~ 1           MZ         (Metro Zoo) Toronto         ON         g         43.87         79.19         (U)         ~ 1           Ha1         Hamilton         ON         py         43.26         79.86         U         ~ 1           Ha2-5         (4 additional sites) Hamilton         ON         y         43.26         79.86         U         ~ 1           Ha6         Hamilton         ON         t         43.98         79.27         UR         30           Lo         London         ON         y         43.00         81.25         U         40           Co         Cornwall         ON         ay         45.05         74.70         U         150           Ot1         (Rideau & Wurtenburg)Ottawa         ON         t         45.43         75.68         UC         150	n n w w w w											
To3         (Downtown) Toronto         ON         t         43.66         79.39         UC         ~ 1           MZ         (Metro Zoo) Toronto         ON         g         43.87         79.19         (U)         ~ 1           Ha1         Hamilton         ON         p.y         43.26         79.86         U         ~ 1           Ha2-5         (4 additional sites) Hamilton         ON         y         43.26         79.86         U         ~ 1           Ha6         Hamilton         ON         t         43.26         79.86         UC         ~ 1           St         Stouffville         ON         t         43.26         79.86         UC         ~ 1           Lo         London         ON         t         43.26         79.86         UC         ~ 1           St         Stouffville         ON         t         43.98         79.27         UR         30           Lo         London         ON         y         43.00         81.25         U         40           Co         Cornwall         ON         a y         45.05         74.70         U         150           Ot1         (Rideau & Wurtenburg)Ottawa <t< td=""><td>n n w w w n</td></t<>	n n w w w n											
MZ         (Metro Zoo) Toronto         ON         g         43.87         79.19         (U)         ~1           Ha1         Hamilton         ON         p.y         43.26         79.86         U         ~1           Ha2-5         (4 additional sites) Hamilton         ON         y         43.26         79.86         U         ~1           Ha6         Hamilton         ON         y         43.26         79.86         U         ~1           St         Stouffville         ON         t         43.26         79.86         UC         ~1           Lo         London         ON         t         43.98         79.27         UR         30           Lo         London         ON         y         43.00         81.25         U         40           Co         Cornwall         ON         a y         45.05         74.70         U         150           Ot1         (Rideau & Wurtenburg) Ottawa         ON         t         45.43         75.68         UC         150	n w w w n											
Ha1         Hamilton         ON         p y         43.26         79.86         U         ~ 1           Ha2-5         (4 additional sites) Hamilton         ON         y         43.26         79.86         U         ~ 1           Ha6         Hamilton         ON         y         43.26         79.86         UC         ~ 1           St         Stouffville         ON         t         43.26         79.86         UC         ~ 1           Lo         London         ON         t         43.98         79.27         UR         30           Lo         London         ON         y         43.00         81.25         U         40           Co         Cornwall         ON         a y         45.05         74.70         U         150           Ot1         (Rideau & Wurtenburg) Ottawa         ON         t         45.43         75.68         UC         150           Ot2         (88 Slater) Ottawa         ON         t         45.42         75.70         UC         150	w w w n											
Ha2-5         (4 additional sites) Hamilton         ON         y         43.26         79.86         U         ~ 1           Ha6         Hamilton         ON         t         43.26         79.86         UC         ~ 1           St         Stouffville         ON         t         43.26         79.86         UC         ~ 1           Lo         London         ON         t         43.98         79.27         UR         30           Lo         London         ON         y         43.00         81.25         U         40           Co         Cornwall         ON         a y         45.05         74.70         U         150           Ot1         (Rideau & Wurtenburg) Ottawa         ON         t         45.43         75.68         UC         150           Ot2         (88 Slater) Ottawa         ON         t         45.42         75.70         UC         150	w w n											
Ha6         Hamilton         ON         t         43.26         79.86         UC         ~1           St         Stouffville         ON         t         43.98         79.27         UR         30           Lo         London         ON         y         43.00         81.25         U         40           Co         Cornwall         ON         a y         45.05         74.70         U         150           Ot1         (Rideau & Wurtenburg) Ottawa         ON         t         45.43         75.68         UC         150           Ot2         (88 Slater) Ottawa         ON         t         45.42         75.70         UC         150	w n											
St         Stouffville         ON         t         43.98         79.27         UR         30           Lo         London         ON         y         43.00         81.25         U         40           Co         Cornwall         ON         a y         45.05         74.70         U         150           Ot1         (Rideau & Wurtenburg) Ottawa         ON         t         45.43         75.68         UC         150	n											
Lo         London         ON         y         43.00         81.25         U         40           Co         Cornwall         ON         a y         45.05         74.70         U         150           Ot1         (Rideau & Wurtenburg) Ottawa         ON         t         45.43         75.68         UC         150           Ot2         (88 Slater) Ottawa         ON         t         45.42         75.70         UC         150												
Co         Cornwall         ON         a y         45.05         74.70         U         150           Ot1         (Rideau & Wurtenburg) Ottawa         ON         t         45.43         75.68         UC         150           Ot2         (88 Slater) Ottawa         ON         t         45.42         75.70         UC         150	n											
Ot1         (Rideau & Wurtenburg) Ottawa         ON         t         45.43         75.68         UC         150           Ot2         (88 Slater) Ottawa         ON         t         45.42         75.70         UC         150												
Ot2         (88 Slater) Ottawa         ON         t         45.42         75.70         UC         150												
	-											
Mo6         (7650 Charteauneuf, Anjou) Montréal         QU         t         45.60         73.56         UR         280           Az         Il Azermenting         QU         t         45.60         73.40         D         200	ne											
As         I'Assomption         QU         t         45.82         73.43         R         300           MS         Mt         Sutton         QU         t         45.09         72.09         D         200	ne											
MS         Mt. Sutton         QU         t         45.08         72.68         R         300           Bb         Brattleboro         VT         y         42.84         72.56         U         300	ne											
	ne											
Fr         St. Francoise         QU         t         46.02         71.93         R         400           QQ         0         0         0         71.03         R         400	ne											
QC         Québec City         QU         t         46.82         71.22         UI         500           Jo         Jonguière         QU         t         48.44         71.20         UI         620	ne											
	ne											
a         Sites Identified in the "Great Lakes Region"         Includes all identified sites in any State or Province adjoining one or more Great L with sites in Manitoba, Quebec, and Vermont added. This table does not include any additional sites used in L. Mich. Mass Balance St the AEOLOS project; information about these was not available for this study.												
b Site Code These codes are used in other tables, as well												
c Programs at Site See Table 3 for program code descriptions. With only a few exceptions, are sites/programs are "active" as of April 1997. Former sites/programs are generally not included.												
d Latitude and Longitude In some cases, these are somewhat approximate.												
e Site Type Approximate Character of Site (all subject to confirmation and checking): Codes in parentheses are guess-timates and should be checked. R = Rural; U = Urban; UI = Urban Industrial; UC = Urban Commercial; UR = Urban Residential												
f Distance from Lake (km) Many of the sites are also in somewhat close proximity to more than one Great La The distance given is the approximate distance to the closest Great Lake. An "~" indicates that precise distance from Lake was not determined in this study, but, the approximate value is given.	,											
g Orientation to Lake Approximate direction from Lake to Site, i.e., "sw" = site is southwest of stated La	ke											

Tabl	e 5.	Monitori	ng of BV	ES Compo	ounds in A	Ambient A	ir and Pre	cipitation i	n the Grea	at Lakes R	egion: Ca	anadian	Program	<b>S</b> (a,b)	
Site Category	y>		Master Sites	3		Satellite Sites		Nation	al Air Pollution	Surveillance N	etwork (NAPS)	(c)	OM	OEE Toxics Mor	itoring
program	ı>	IADN	IADN	IADN		IADN		voc	PAH	PCDD/F	COA	PART	PAH	voc	PCDD/F
sponsor / coordir	nator	EC AES	EC EHD	EC NWRI	EC AES	OMOEE	EC EHD			EC			OMOEE	OMOEE	OMOEE
site	s>	BI, PP	BI, PP	РР	Eg	Do, WR. PS, GB	Sb, TL. GB, SC. PI, RP. Bu	PP, To2-3, St, Eg, Ha6,Sm, Lw,Sa,Wd2	PP, To2, Ha6, Sm, W d2	PP, To2, Ha6, Sm, Wd2	PP, To2, Ha6, Sm, Wd2	PP, To1, To3, W d2	SSM(2), W d1&3, Na(2), W e,Co	SSM(2), TB W d1&3, Lo Na(2), Do Ha1-5, Co	Ha1, Wd1 [+precip at Do]
phase(s	)>	vapor part.	precip	precip	vapor part.	vap, part. precip	precip	vapor	combined vapor+part	combined vapor+part	combined vapor+part	part	combined vap+part	vapor	combined vapor+part
Compound or Group	Lvl		Notes: (a) A	An "x" = compou	nd is measure	d in given prog	ram. (b) see Ta	bles 3, 4 and Lis	st of Abbreviatio	ons. (c) The NA	APS program al	so includes	sites in Quebe	ec and Manitoba	
MERCURY and MERCURY C	омро	UNDS													
Elemental Mercury (Hg°)	I														
DivalentHg (e.g., HgCl <sub>2</sub> )	I														
Monomethyl Mercury	I														
Total Gaseous Mercury	I	k: PP			k	s: Do									
Particulate Mercury	I											x			
Total Mercury	1					s: Do (pre)									
OTHER METALS / ORGANON	IETAL	LICS													
Alkylated Lead	I														
Total Cadmium (Cd)	П	x	x	x	x	x	x					x			
Individual Cd Species	II														
Tributyltin Compounds	Ш														
ORGANOCHLORINE BIOCID	ES					-	-		-	-				-	
Aldrin	I	x	x	x	x	x	x								
Dieldrin	I	x	x	x	x	x	x								
Chlordane	I	α,γ,tn,ox	α,γ	α,γ,tn	α,γ,tn,ox	α,γ,tn,ox	α,γ								
DDT / DDD / DDE	I	x	x	x	x	x	x								
Endrin	II	x	x	x	x		x								
Heptachlor	Ш	x	x	x	x	x	x								
Heptachlor Epoxide	Ш	×	x	x	x	x	x								
Hexachlorocyclohexanes	II	α,β,δ,γ	α,γ	α,γ	α,β,δ,γ	α,β,γ	α,γ								
Methoxychlor	Ш	x	x	x	x	x	x								
Mirex	I	×		x	x	x									
Pentachlorophenol	Ш										x				
Toxaphene	I	b: PP													

INDUSTRIAL / MISCELLANEOUS															
4-Bromophenyl Phenyl Ether															
3,3'-Dichlorobenzidene															
Hexachloro-1,3-Butadiene	11		<u> </u>					x							
4,4'-Methylene bis(2-Chloroaniline)	II														
Octachlorostyrene	1					x					x				
CHLOROBENZENES				1							I	<u> </u>			
1,4-dichlorobenzene	П		x	x			x	x						x	
Tetrachlorobenzenes	П		x	x		x	x								
Pentachlorobenzene	Ш		x	x		x	x								
Hexachlorobenzene	I	x	×	x	x	x	x				x				
POLYCHLORINATED DIBEN	ZO-P-D	IOXINS & DIB	ENZOFURA	NS (PCDD/F'S)	and POLYCH	LORINATED B	IPHENYLS (PC	B'S)		·		·:		·	·
PCDD/F's	I					p:Do(pre)				x					x
PC B's	I	x		x	x	x									
POLYCYCLIC AROMATIC HY	DROC	ARBONS	•							•				•	
Acenaphthene	II-a	x	x	x	x		x		x				x		
Acenaphthylene	II-a	x	x	x	x		x		x				x		
Anthracene	П	x			x				x				x		
Benz (a) Anthracene	II	x		x	x	x			x				x		
Benzo (b) Fluoranthene	II-a	x		x	x	х			x				x		
Benzo (j) Fluoranthene	ll-a														
Benzo (k) Fluoranthene	II-a	x			x	x			x				x		
Benzo (a) Pyrene	I	x		x	x	х			x				x		
Benzo (e) Pyrene	ll-a	x			x				x				x		
Benzo (g,h,i ) Perylene	II	x			x	х			x				x		
Chrysene	ll-a	x		x	x	x			x				x		
Dibenz(a,h)Anthracene	ll-a	x			x	x			x				x		
Dinitropyrenes	II										x				
Fluoranthene	ll-a	x	×	x	x	x	x		x				x		
Fluorene	ll-a	x	×	x	x		x		x				x		
Indeno(1,2,3-cd)Pyrene	ll-a	x			x	x			x				x		
Naphthalene	ll-a							x						x	
Phenanthrene	II	x		x	x	x			x				x		
Perylene	II								x				x		
Pyrene	ll-a	x	x	x	x	х	x		x				x		

1	Table	6. Monitoring of BVE	S Compounds in Am	bient Air and Precipit	ation in the Great Lak	es Region: U.S. Prog	rams (a,b,c)	
Site Catego	ry>	Master Sites	Satelli te Sites	MichiganDNR	Wisconsin DNR	Mercury	Mercury	Vermont
progra	m>	IADN	IADN	air toxics	air toxics	MDN	HgR	HACMP
sponsor / coordi	inator	EPA	EPA	Mich DNR	Wisc DNR	various	EPA (?)	Vermont ANR
site	es>	EH, SBD, SP	BR, Ch	SH, Pe, Dv, Dx (ongoing ?)	GBy (moved to Wisconsin Rapids in June 1997)	MF, Fb,CR, Lt, AP, HC, PR, TrL, LG	EH, SBD, SP, BI, PP, Bo, WM, SF, Dx, CC (1995-1996 only)	BV, Wn, Rt, Un, Bb (sites 250-300 km from Lk Ontario)
phase(s	s)>	vapor part. precip	vapor part. precip	combined (?) vapor+part	combined vapor+part	precip	vapor part	vapor+part
Compound or Group	LM	Notes:	(a) "x" = compound is measured in	given program (b) see Tables 3,	4 & List of Abbrev. (c) Table does r	not include info. regarding L Mich. N	lass Balance Study or AEOLOS pr	oject
MERCURY and MERCURY COMPOUNDS								
Elemental Mercury (Hg°)	1							
Divalent Hg (e.g., HgCl <sub>2</sub> )	I							
Monomethyl Mercury	I							
Total Gaseous Mercury	I	r; f: EH		q: Dx			x	Un: LCS
Particulate Mercury	1	r; f: EH					x	Un: LCS
Total Mercury	I		d: BR			х		
OTHER METALS / ORGANOMETALLICS								
Alkylated Lead	1							
Total Cadmium (Cd)	П	x	x	x	x			X?
Individual Cd Species	П							
Tributyltin Compounds	П							
ORGANOCHLORINE BIOCIDES								
Aldrin	I							
Dieldrin	1	x	x	x	x			
Chlordane	I	α,γ,tn	α,γ,tn		x			
DDT / DDD / DDE	1	x	x	x	DDT			
Endrin	II							
Heptachlor	П							
Heptachlor Epoxide	П							
Hexachlorocyclohexanes	II	α,γ	α,γ	α,γ	Υ			
Methoxychlor	II							
Mirex	1							
Pentachlorophenol	II							
Toxaphene	I	h: research						

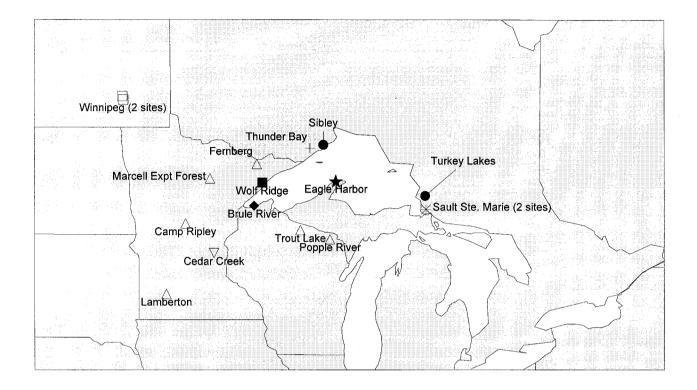
INDUSTRIAL / MISCELLANEOUS							
4-Bromophenyl Phenyl Ether	Ш						
3,3'-Dichlorobenzidene	Ш						
Hexachloro-1,3-Butadiene	Ш				x		x
4,4'-Methylene bis(2-C hlor oanil ine)	Ш						
Octachlorostyrene	I						
CHLOROBENZENES						 	
1,4-dichlorobenzene	П				x		x
Tetr achlorobenz enes	Ш						
Pentachlorobenzene	Ш						
Hexachlorobenzene	I	x	x	x			
POLYCHLORINATED DIB ENZO-P-DIOXINS &	DIBENZ	OFURANS (PCDD/F'S) and PO	LYCHLORINATED BIPHENYLS	(PCB'S)			
PCDD/F's	I						x
PCB's	I	x	x	x	"total pcb's"		x
POLYCYCLIC AROMATIC HYDROCARBONS						 	
Acenaphthene	II-a	x	x				x
Acenaphthylene	II-a	x	x	x			x
Anthracene	Ш	x	x	x			x
Benz (a) Anthracene	II	x	x		x		x
Benzo (b) Fluor anthene	II-a	x	x				x
Benzo (j) Fluoranthene	II-a						
Benzo (k) Fluoranthene	II-a	x	x			 	x
Benzo (a) Pyrene	I	x	x	x	x	 	x
Benzo (e) Pyrene	II-a	x	x				
Benzo (g,h,i ) Perylene	Ш	x	x				x
Chrysene	II-a	x	x		x		x
Di benz(a,h)Anthracene	II-a	x	x			 	x
Dinitropyrenes	II					 	
Fluoranthene	II-a	x	x	x	x		x
Fluorene	II-a	x	x	x			x
Indeno(1,2,3-cd)Pyrene	II-a	x	x				x
Naphthal ene	II-a			x	x	 	x
Phenanthrene	Ш	x	x	×	x		x
Perylene	11						
Pyrene	II-a	x	x	x	x		x

					ir Monitorin	a				Drooin	tation Monit	oring	
Compound or Compound Group	Level	_				Ī	Ac				tation Monit		Ac
	<u>e</u>	Eagle Harbor	Brule River	WolfRidge	Thunder Bay	Sault Ste. Marie (2 sites)	Additional Sites	Eagle Harbor	Brule River	WolfRidge	Sibley	Turkey Lakes	Additional Sites
distance from Lake (km)>		0.1	0.4	~ 1	~ 1	~ 1	CC = 180 Wp1,2 = 530	0.1	0.4	~ 1	~ 1	~ 25	70 - 380 km
character of site>		R	R	(R)	U	U		R	R	(R)	(R)	(R)	
MERCURY and MERCURY COMPOUN	IDS												
Elemental Mercury (Hg°)	1												
Divalent Hg (e.g., HgCl <sub>2</sub> )	I												
Monomethyl Mercury	1												
Total Gaseous Mercury	1	fr					r: CC						
Particulate Mercury	T	fr					r:CC; t:Wp1						
Total Mercury	1								d				d: Fb, TrL, PR, MF, CR, Lt; r: CC
OTHER METALS / ORGANOMETALLIC	s												
Alkylated Lead	I												
Total Cadmium (Cd)	П	u	u	o			t: Wp1	u	u	o	g	g	
Individual Cd Species	П												
Tributyltin Compounds	П												
ORGANOCHLORINE BIOCIDES	1	-	1	1	1	1			1	1	1	1	
Aldrin	1			о						о	g	g	
Dieldrin	I	u	u	o				u	u	0	g	g	
Chlordane	1	u	u	o				u	u	0	g	g	
DDT / DDD / DDE	1	u	u	o				u	u	0	g	g	
Endrin	П										g	g	
Heptachlor	П			0						0	g	g	
Heptachlor Epoxide	Ш			0						0	g	g	
Hexachlorocyclohexanes	Ш	u	u	о				u	u	o	g	g	
Methoxychlor	П			0						0	g	g	
Mirex	I			o						0			
Pentachlorophenol	П												
Toxaphene	1	h						Ī					
INDUSTRIAL / MISCELLANEOUS			·	·	·	ı			J				
4-Bromophenyl Phenyl Ether	П												
3,3'-Dichlorobenzidene	П												
Hexachloro-1,3-Butadiene	Ш						t: Wp1						
4,4'-Methylene bis (2-C hlor oanil ine)	II												

Table 7. Summa	ary of A	mbient		-		-	of BVES Subst ns, see tables 3 and 4)		n the Vio	inity of	Lake Su	perior	
				Ai	r Monitorin	g				Precipi	tation Monit	toring	
Compound or Compound Group	Level	Eagle Harbor	Brule River	WolfRidge	Thunder Bay	Sault Ste. Marie (2 sites)	Additional Sites	Eagle Harbor	Brule River	WolfRidge	Sibley	Turkey Lakes	Additional Sites
distance from Lake (km)>		0.1	0.4	~ 1	~ 1	~ 1	CC = 180 Wp1,2 = 530	0.1	0.4	~ 1	~ 1	~ 25	70 - 380 km
character of site>		R	R	(R)	U	U		R	R	(R)	(R)	(R)	
CHLOROBENZENES													
1,4-dichlorobenzene	П				У	У	t: Wp1				g	g	
Tetr achl orobenz enes	П			o						o	g	g	
Pentachlorobenzene	П			o						0	g	g	
Hexachlorobenzene	I	u	u	o				u	u	o	g	g	
POLYCHLORINATED DIB ENZO-P-DIO	XINS & DI	BENZOFU	IR ANS (PC	DD/F'S) ar	nd POLYC	HLORINAT	ED BIPHENYLS (PC	B'S)	•				
PCDD/F's	I						t: Wp1,Wp2						
PCB's	I	u	u	0				u	u	0			
POLYCYCLIC AROMATIC HYDROCAR	BONS									L			
Acenaphthene	I⊦a	u	u			а	t: Wp1,Wp2	u	u		g	g	
Acenaphthylene	I⊦a	u	u			а	t: Wp1,Wp2	u	u		g	g	
Anthracene	П	u	u	o		а	t: Wp1,Wp2	u	u	o			
Benz (a) Anthracene	П	u	u	o		а	t: Wp1,Wp2	u	u	o			
Benzo (b) Fluoranthene	I⊦a	u	u	o		а	t: Wp1,Wp2	u	u	0			
Benzo (j) Fluoranthene	I⊦a												
Benzo (k) Fluoranthene	I⊦a	u	u	0		а	t: Wp1,Wp2	u	u	o			
Benzo (a) Pyrene	I	u	u	o		а	t: Wp1,Wp2	u	u	o			
Benzo (e) Pyrene	I⊦a	u	u			а	t: Wp1,Wp2	u	u				
Benzo (g,h,i ) Perylene	П	u	u	o		а	t: Wp1,Wp2	u	u	o			
Chrysene	I⊦a			o		а	t: Wp1,Wp2			o			
Di benz(a,h)Anthracene	I⊦a	u	u	o		а	t: Wp1,Wp2	u	u	o			
Dinitropyrenes	П												
Fluoranthene	I⊦a	u	u	o		а	t: Wp1,Wp2	u	u	o	g	g	
Fluorene	I⊦a	u	u			а	t: Wp1,Wp2	u	u		g	g	
Indeno(1,2,3-cd)Pyrene	I⊦a	u	u	o		а	t: Wp1,Wp2	u	u	o			
Naphthalene	I⊦a				У	У	t: Wp1						
Phenanthrene	П	u	u	o		а	t: Wp1,Wp2	u	u	o			
Perylene	П					а	t: Wp1,Wp2						
Pyrene	I⊦a	u	u	o		а	t: Wp1,Wp2	u	u	0	g	g	

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Figure 1. Air and Precipitation Sampling Sites for One or More BVES Compounds in the Vicinity of Lake Superior



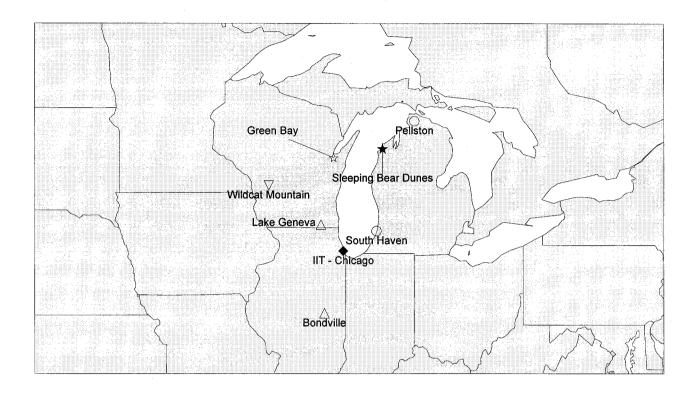
	prograr code (see	m
<u>symbol</u>	(see <u>Tbl 3)</u>	description
*	u	IADN Master Site (air and precipitation) The site in this figure, Eagle Harbor, is a U.S. Site; Toxaphene is being monitored at this site in program "h" Mercury is/was monitored at this site in program "f" and "r", respectively
<b>♦</b>	u	U.S. IADN Satellite Site (air and precipitation) Brule River, is also part of the Mercury Deposition Network, program "d"
۲	g	IADN Satellite Site, Environment Canada EHD (precipitation)
	0	OMOEE IADN Satellite Site (air and precipitation)
	t	Nat'l Air Pollutant Surveillance Network, Envr. Canada (air) both sites in Winnepeg measure PCDD/F's and PAH's one of the sites in Winnepeg measures VOC's and particulate metals
+	у	OMOEE Air Toxics Monitoring, VOC's
*	a,y	OMOEE Air Toxics Monitoring, PAH's and VOC's
$\bigtriangleup$	d	Mercury Deposition Network (precipitation only)
$\bigtriangledown$	r	EPA Mercury Research Project (1995-96) (air and precip.)

Table 8. Summary	of Amb		d Precipitati scriptions of					ity of Lake	Michigan	
				air mo	onitoring			pre	ecipi tati on monito	ing
Compound or Compound Group	Level	Sleeping Bear Dunes	IIT - Chicago	Green Bay (city)	South Haven	Peliston	Additional Sites	Sleeping Bear Dunes	IIT - Chicago	Additional Sites
distance from Lake (km)>		1	1.5	~ 1	~ 1	20				
character of site>		R	U	U	(R)	(R)				
MERCURY and MERCURY COMPOUNDS	6									
Elemental Mercury (Hg°)	I									
Divalent Hg (e.g., HgCl <sub>2</sub> )	I									
Monomethyl Mercury	I									
Total Gaseous Mercury	I	r					r: WM,Bo			
Particulate Mercury	I	r	_	-	_	_	r: WM,Bo			
Total Mercury	I									d: LG, Bo
OTHER METALS / ORGANOMETALLICS	<u> </u>		<u> </u>	<u>.</u>						
Alkylated Lead	I									
Total Cadmium (Cd)	П	u	u	w	m	m		u	u	
Individual Cd Species	П									
Tributyltin Compounds	П									
ORGANOCHLORINE BIOCIDES	! <b>!</b>		<u> </u>							
Aldrin	I									
Dieldrin	I	u	u	w	m	m		u	u	
Chlordane	I	u	u	w				u	u	
DDT / DDD / DDE	I	u	u	w	m	m		u	u	
Endrin	П									
Heptachlor	П									
Heptachlor Epoxide	II									
Hexachloroc yclohexanes	II	u	u	w	m	m		u	u	
Methoxychlor	II				_					
Mirex	I				_					
Pentachlorophenol	II									
Toxaphene	I									
INDUSTRIAL / MISCELLANEOUS				·						
4-Bromophenyl Phenyl Ether	П									
3,3'-Dichlorobenzidene	11									
Hexachloro-1,3-Butadiene	П			w						
4,4'-Methylene bis (2-C hlor oanil ine)	11									
Octachlorostyrene	I									

Table 8. Summary	Table 8. Summary of Ambient Air and Precipitation Monitoring of BVES Substances in the Vicinity of Lake Michigan         (for descriptions of codes in columns, see tables 3 and 4)											
				air mor	nitoring		,	pre	cipi tati on monitor	ing		
Compound or C ompound Group	Level	Sleeping Bear Dunes	IIT - Chicago	Green Bay (city)	South Haven	Peliston	Additional Sites	Sleeping Bear Dunes	IIT - Chicago	Additional Sites		
CHLOROBENZENES												
1,4-dichlorobenzene	Ш											
Tetrachlorobenzenes	II			w		-						
Pentachlorobenzene	II					-						
Hexachlorobenzene	I	u	u		m	m		u	u			
POLYCHLORINATED DIB ENZO-P-DIOXI	NS & DIBE	NZOFURANS	(PCDD/F'S) and	POLYCHLOR	NATED BIPHE	NYLS (PCB'S)						
PCDD/F's	I											
PCB's	I	u	u	w	m	m		u	u			
POLYCYCLIC AROMATIC HYDROCARB	ONS							-				
Acenaphthene	l⊦a	u	u					u	u			
Acenaphthylene	l⊦a	u	u		m	m		u	u			
Anthracene	П	u	u		m	m		u	u			
Benz (a) Anthracene	П	u	u	w				u	u			
Benzo (b) Fluor anthene	l⊦a	u	u					u	u			
Benzo (j) Fluoranthene	l⊦a											
Benzo (k) Fluoranthene	l⊦a	u	u					u	u			
Benzo (a) Pyrene	I	u	u	w	m	m		u	u			
Benzo (e) Pyrene	l⊦a	u	u					u	u			
Benzo (g,h,i ) Perylene	П	u	u					u	u			
Chrysene	l⊦a			w								
Dibenz(a,h)Anthracene	l⊦a	u	u					u	u			
Dinitropyrenes	П											
Fluoranthene	l⊦a	u	u	w	m	m		u	u			
Fluorene	l⊦a	u	u		m	m		u	u			
Indeno(1,2,3-cd)Pyrene	l⊦a	u	u					u	u			
Naphthal ene	l⊦a			w	m	m						
Phenanthrene	П	u	u	w	m	m		u	u			
Perylene	II											
Pyrene	l⊦a	u	u	w	m	m		u	u			

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Figure 2. Air and Precipitation Sampling Sites for One or More BVES Compounds in the Vicinity of Lake Michigan



oumbol	progran code (see	
<u>symbol</u>	<u>Tbl 3)</u>	description
*	u	IADN Master Site (air and precipitation) The site in this figure, Sleeping Bear Dunes, is a U.S. Site; Mercury was monitored at this site in program "r"
	u	U.S. IADN Satellite Site (air and precipitation)
☆	W	Wisconsin Department of Natural Resources
0	m	Michigan Department of Natural Resources (air)
Δ	d	Mercury Deposition Network (precipitation only) Mercury was monitored at Bondville in program "r"
$\nabla$	r	EPA Mercury Research Project (1995-96) (air and precip.)

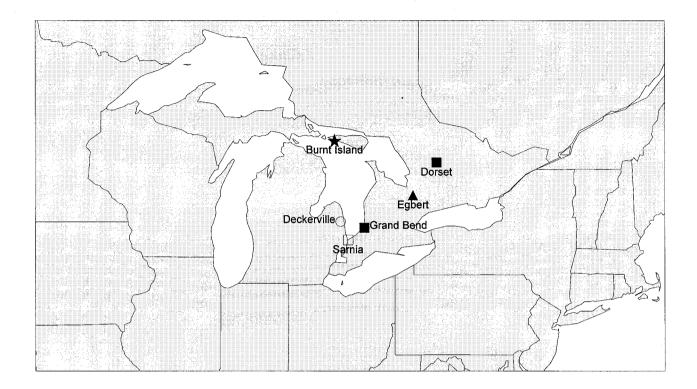
(Note: this map does not include additional sampling locations utilized in the Lake Michigan Mass Balance Study or the AEOLOS project; accounts of these research projects are given by others elsewhere.)

Table 9. Sum	mary	of Ambient Aira (forde:				S Substanc		inity of Lak	e Huron	
				Air Monit	oring			Pre	cipitation Monito	ring
Compound or Compound Group	Level	Burnt Island	Grand Bend	Samia	Dorset	Deckerville	Egbert	Burnt Island	Grand Bend	Dorset
distance from Lake (km)>		~ 1	~ 1	~ 1	90	20	40	~ 1	~ 1	90
character of site>		R	(R)	UR	(R)	(R)	(R)	R	(R)	(R)
MERCURY and MERCURY COMPO	UNDS									
Elemental Mercury (Hg°)	I									
Divalent Hg (e.g., HgCl <sub>2</sub> )	I									
Monomethyl Mercury	I									
Total Gaseous Mercury	I	r k summer 97			s		k			
Particulate Mercury	I	r					t			
Total Mercury	I									s
OTHER METALS / ORGANOMETALL	ICS									
Alkylated Lead	I									
Total Cadmium (Cd)	Ш	с	o		0	m	et	g	g	0
Individual Cd Species	Ш									
Tributyltin Compounds	Ш									
ORGANOCHLORINE BIOCIDES										
Aldrin	I	с	o		0		е	g	g	0
Dieldrin	I	с	o		0	m	е	g	g	o
Chlordane	I	с	о		о		е	g	g	o
DDT / DDD / DDE	I	с	o		0	m	е	g	g	0
Endrin	Ш	с					е	g	g	
Heptachlor	Ш	с	o		0		е	g	g	0
Heptachl or Epoxide	Ш	с	о		o		е	g	g	o
Hexachloroc yclohexanes	Ш	с	0		0	m	е	g	g	0
Methoxychlor	Ш	с	о		о		е	g	g	0
Mirex	I	с	o		o		е			o
Pentachlorophenol	Ш									
Toxaphene	I									
INDUSTRIAL / MISCELLANEOUS							1			
4-Bromophenyl Phenyl Ether	Ш									
3,3'-Di chlorobenzidene	II									
Hexachloro-1,3-Butadiene	Ш			t			t			
4,4'-Methylene bis (2-C hlor oanil ine)	Ш									
Octachlorostyrene	1		0		0					o

Table 9. Sum	mary	of Ambient Air a (for de:			oring of BVE lumns, see T			inity of Lak	e Huron	
				Air Monito	oring			Pro	ecipitation Monito	ring
Compound or Compound Group	Level	Burnt Island	Grand Bend	Samia	Dorset	Deckerville	Egbert	Burnt Island	Grand Bend	Dorset
distance from Lake (km)>		~ 1	~ 1	~ 1	90	20	40	~ 1	~ 1	90
character of site>		R	(R)	UR	(R)	(R)	(R)	R	(R)	(R)
CHLOROBENZENES										
1,4-dichlorobenzene	Ш			t	У		t	g	g	
Tetrachlorobenzenes	Ш		o		o			g	g	o
Pentachlorobenzene	Ш		o		0			g	g	o
Hexachlorobenzene	I	с	o		o	m	е	g	g	0
POLYCHLORINATED DIB ENZO-P-D	IOXINS	& DIBENZOFURANS	S (PCDD/F'S) ar	nd POLYCHLOF	RINATED BIPHE	NYLS (PCB'S)				
PCDD/F's	I									р
PCB's	I	с	0		0		е			o
POLYCYCLIC AROMATIC HYDROC	ARBON	IS							·	
Acenaphthene	II-a	с					е	g	g	
Acenaphthylene	II-a	с				m	е	g	g	
Anthracene	II	с	0		o	m	е			о
Benz (a) Anthracene	II	с	0		o		е			ο
Benzo (b) Fluoranthene	II-a	с	0		0		e			0
Benzo (j) Fluoranthene	II-a									
Benzo (k) Fluoranthene	II-a	с	o		o		е			o
Benzo (a) Pyrene	I	с	о		o	m	е			o
Benzo (e) Pyrene	II-a	с					е			
Benzo (g,h,i ) Per ylene	Ш	с	o		o		е			0
Chrysene	II-a	с	o		o		е			о
Dibenz(a,h)Anthracene	II-a	С	0		0		е			0
Dinitropyrenes	Ш									
Fluoranthene	II-a	с	0		0	m	е	g	g	0
Fluorene	II-a	с				m	е	g	g	
Indeno(1,2,3-cd)Pyrene	II-a	с	0		0		е			0
Naphthalene	II-a			t	У	m	t			
Phenanthrene	Ш	с	0		o	m	е			0
Perylene	Ш									
Pyrene	II-a	с	0		o	m	е	g	g	0

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	prograr code (see	n
<u>symbol</u>	<u>(1000</u> Tbl 3)	description
*	С	IADN Master Site (air and precipitation) The site in this figure, Burnt Island, is a Canadian site; Air monitoring conducted by Atmospheric Environment Service, Envr. Canada Precipitation monitoring by Envr. Canada EHD (program "g") Mercury was monitored at this site in program "r" Monitoring of total gaseous mercury may begin in summer 1997 (program "k")
	e	IADN Satellite Site at Egbert, Ontario, Environment Canada (air) total gaseous mercury is measured here also, program "k" this is also reportedly a NAPS site ("t") where VOC's and particulate metals are measured
<b>.</b> .	0	OMOEE IADN Satellite Site (air and precipitation) At Dorset, total mercury in precipitation and total gaseous mercury are measured (program "s") Also at Dorset, PCDD/F's in precipitation and VOC's in air are measured by OMOEE (programs "p" and "y") At Grand Bend, precipitation monitoring by EHD is also performed (program "g")
	t	Nat'l Air Pollutant Surveillance Network, Envr. Canada (air) at the NAPS site at Sarnia, VOC's are measured
0	m	Michigan Department of Natural Resources (air)

Table 10. Su	d	.,						imns, see T				110		Lune L		
						Air Monitor	ing						Precip	tation Mor	nitoring	
Compound or Compound Group	Level	Sturgeon Point	Port Stanley	Simcoe	Nanticoke (2 sites)	Welland	Windsor (*)	Longwoods	Dexter	Additional Sites	Sturgeon Point	Port Stanley	Pelee Island	RockPoint	St. Clair	Additional Sites
distance from Lake (km)>		0.1	~ 1	~ 1	~ 1	10	30	30	80		0.1	~ 1	~ 1	~ 1	30	
character of site>		(R)	(R)	R	U	U	U	R	(R)		(R)	(R)	R	(R)	(R)	
MERCURY and MERCURY COMPO	UNDS		. ,					1	. ,			. ,		. ,		
Elemental Mercury (Hg°)	I															
Divalent Hg (e.g., HgCl <sub>2</sub> )	1															
Monomethyl Mercury	I															
Total Gaseous Mercury	I	r			I 				qr	r: SF						
Particulate Mercury	I	r					t		q r	r: SF						
Total Mercury	I										r					q:Dx r:Dx,SF d:HC,AP
OTHER METALS / ORGANOMETAL	LICS															
Alkylated Lead	I															
Total Cadmium (Cd)	Ш	u	ο				t		m		u	ο	g	g	g	
Individual Cd Species	Ш															
Tributyltin Compounds	Ш															
ORGANOCHLORINE BIOCIDES			1	1	1	1	1	1	1	1		1	1	1	1	
Aldrin	I		o									o	g	g	g	
Dieldrin	I	u	o						m		u	o	g	g	g	
Chlordane	I	u	o								u	o	g	g	g	
DDT / DDD / DDE	I	u	o						m		u	o	g	g	g	
Endrin	II												g	g	g	
Heptachlor	П		о									o	g	g	g	
Heptachl or Epoxide	Ш		o									o	g	g	g	
Hexachloroc yclohexanes	II	u	o						m		u	o	g	g	g	
Methoxychlor	II		о									о	g	g	g	
Mirex	I		о									о				
Pentachlorophenol	Ш			t			t									
Toxaphene	I															
INDUSTRIAL / MISCELLANEOUS				1	1	1		1	1	1						
4-Bromophenyl Phenyl Ether	II															
3,3'-Di chl orobenz idene	Ш															
Hexachloro-1,3-Butadiene	II			t			t	t								
4,4'-Methylene bis (2-C hlor oanil ine)	II															
Octachlorostyrene	I.		o	t			t				Í	o				

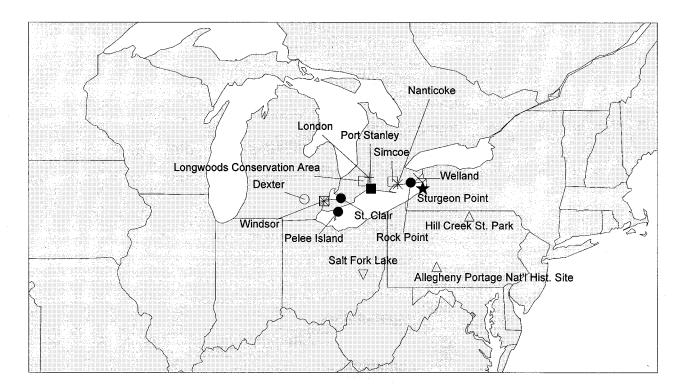
Table 10. Su	mmai	ry of A	mbient		-	oitation ons of cod		-			ces in	the Vic	inity of	Lake E	rie	
					A	vir Monitor	ing						Precip	itation Mo	nitoring	
Compound or C ompound Group	Level	Sturgeon Point	Port Stanley	Simcoe	Nanticoke (2 sites)	Welland	Windsor (*)	Longwoods	Dexter	Additional Sites	Sturgeon Point	Port Stanley	Pelee Island	RockPoint	St. Clair	Additional Sites
distance from Lake (km)>		0.1	~ 1	~ 1	~ 1	10	30	30	80		0.1	~ 1	~ 1	~ 1	30	
character of site>		(R)	(R)	R	U	U	U	R	(R)		(R)	(R)	R	(R)	(R)	
CHLOROBENZENES	11															
1,4-dichlorobenzene	Ш			t	у		t y	t					g	g	g	
Tetrachlorobenzenes	Ш		o									o	g	g	g	
Pentachlorobenzene	Ш		o									о	g	g	g	
Hexachlorobenzene	1	u	о	t			t		m		u	о	g	g	g	
POLYCHLORINATED DIBENZO-P-D	IOXINS	& DIBEN		ANS (PC D	)D/F'S) ar	nd POLYC	HLORIN	Ated Bif	PHENYLS	6 (PC B'S)						
PCDD/F's	I			t			tp									
PCB's	I	u	o						m		u	o				
POLYCYCLIC AROMATIC HYDROC	ARBON	IS														
Acenaphthene	II-a	u		t	а	а	ta				u		g	g	g	
Acenaphthylene	II-a	u		t	а	а	ta		m		u		g	g	g	
Anthracene	Ш	u	o	t	а	а	ta		m		u	о				
Benz (a) Anthracene	Ш	u	o	t	а	а	ta				u	o				
Benzo (b) Fluor anthene	II-a	u	o	t	а	а	ta				u	o				
Benzo (j) Fluoranthene	II-a															
Benzo (k) Fluoranthene	II-a	u	o	t	а	а	ta				u	о				
Benzo (a) Pyrene	I	u	0	t	а	а	ta		m		u	ο				
Benzo (e) Pyrene	II-a	u		t	а	а	ta				u					
Benzo (g,h,i ) Perylene	Ш	u	o	t	а	а	ta				u	0				
Chrysene	II-a		o	t	а	а	ta					o				
Dibenz(a,h)Anthracene	II-a	u	0	t	а	а	ta				u	0				
Dinitropyrenes	Ш															
Fluoranthene	II-a	u	o	t	а	а	ta		m		u	o	g	g	g	
Fluorene	II-a	u		t	а	а	ta		m		u		g	g	g	
Indeno(1,2,3-cd)Pyrene	II-a	u	o	t	а	а	ta				u	0				
Naphthalene	II-a			t	У		t y	t	m							
Phenanthrene	Ш	u	o	t	а	а	ta		m		u	ο				
Perylene	Ш			t	а	а	ta									
Pyrene	ll-a	u	o	t	а	а	ta		m		u	o	g	g	g	

(\*)

NAPS operates one site in Windsor, measuring VOC's, PAH's, PCDD/F's, selected COA substances, and particulate metals. OMOEE operates two sites in Windsor. At both sites, PAH's and VOC's are measured; at the University Avenue site, PCDD/F's are measured in air.

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# Figure 4. Air and Precipitation Sampling Sites for One or More BVES Compounds in the Vicinity of Lake Erie



	prograi code (see	m
<u>symbol</u>	<u> Tbl 3)</u>	description
*	u	IADN Master Site (air and precipitation) The site in this figure, Sturgeon Point, is a U.S. site; Mercury was monitored at this site in program "r" Monitoring of mercury in precipitation may start again soon (program "d")
	0	OMOEE IADN Satellite Site (air and precipitation) At Dorset, total mercury in precipitation and total gaseous mercury are measured (program "s") At Grand Bend, precipitation monitoring by EHD is also performed (program "g")
	t	Nat'l Air Pollutant Surveillance Network, Envr. Canada (air) At the Windsor NAPS site, VOC's, PCDD/F's, PAH's, particulate metals, and selected COA substances are measured; OMOEE also monitors PCDD/F's at a site in Windsor (program "p") At the Longwoods NAPS site, VOC's are measured At the Simcoe NAPS site, VOC's, PCDD/F's, PAH's, & selected COA substances are measured
x	а	OMOEE Air Toxics Monitoring, PAH's (site on this map: Welland, Ontario)
+	у	OMOEE Air Toxics Monitoring, VOC's (site on this map: London, Ontario)
*	a,y	OMOEE Air Toxics Monitoring, PAH's and VOC's (sites on this map: Windsor (2 sites) and Nanticoke (2 sites), Ontario)
0	m	Michigan Department of Natural Resources (air) Mercury is/was being monitored at Dexter in programs "q" and "r", respectively
$\Delta$	d	Mercury Deposition Network (precipitation only)
$\bigtriangledown$	r	EPA Mercury Research Project (1995-96) (air and precip.)

Table 11. S	Gumm	ary of	Ambier	nt Air a				toring of BVES Substances in th olumns, see Tables 3 and 4)	e Vicinity	of Lake	Ontario	
						A	ir Monitorin	g		Pred	pitation Mo	nitoring
Compound or Compound Group	Level	Pt. Petre	Toronto (*)	Hamilton (**)	Stouffville	London	Cornwall	Additional Sites	Pt. Petre	Toronto	Burlington	Additional Sites
distance from Lake (km)>		0.1	~ 1	~ 1	30	40	150	all ≥ 150 km	0.1	~ 1	~ 1	all $\ge 200  \text{km}$
character of site>		R	U	U	UR	U	U		R	U	U	
MERCURY and MERCURY COMPO	UNDS											
Elemental Mercury (Hg°)	I											
Divalent Hg (e.g., HgCl <sub>2</sub> )	I											
Monomethyl Mercury	I											
Total Gaseous Mercury	I	kr						k SA; l: Un				
Particulate Mercury	1	tr	t			ĺ		t: Ot2, Mo2, Mo6,QC; I: Un				
Total Mercury	1					Ĭ			r			k SA;I: Un
OTHER METALS / ORGANOMETALL	ICS						. <u> </u>			·	!	
Alkylated Lead	I											
Total Cadmium (Cd)	11	ct	t					t: Of2,Mo2, Mo6,QC; v. BV, Wn, Rt, Un, Bb	ng	g	g	
Individual Cd Species												
Tributyltin Compounds												
ORGANOCHLORINE BIOCIDES	<u> </u>				<u>.</u>	<u>_</u>	<u> </u>			<u></u>	<u> </u>	
Aldrin	I	с							ng	g	g	
Dieldrin	I	с							ng	g	g	
Chlordane	I	с							ng	g	g	
DDT / DDD / DDE	1	с							ng	g	g	
Endrin	11	с			_				ng	g	g	
Heptachlor	11	с				1			ng	g	g	
Heptachlor Epoxide	11	с							ng	g	g	
Hexachlorocyclohexanes		с	1	<u> </u>					ng	g	g	
Methoxychlor		с							ng	g	g	
Mirex	1	c							n		5	
Pentachlorophenol		t	t	t					-			
Toxaphene		b			<u> </u>				_			
INDUSTRIAL / MISCELLANEOUS	<u> '</u>		<u> </u>	<u> </u>	<u> </u>	<u> </u>				<u> </u>		
4-Bromophenyl Phenyl Ether	Ш											
3,3'-Dichlorobenzidene												
Hexachloro-1,3-Butadiene	11	t	t	t	t			t:Ot1,2,SA, Mo1,2,4,5,As,MS,Fr; v: BV, Wn, Rt, Un, Bb				
4,4'-Methylene bis(2-C hloroanil ine)	11								-	<u> </u>		
Octachlorostyrene	1	t	t	t					-			

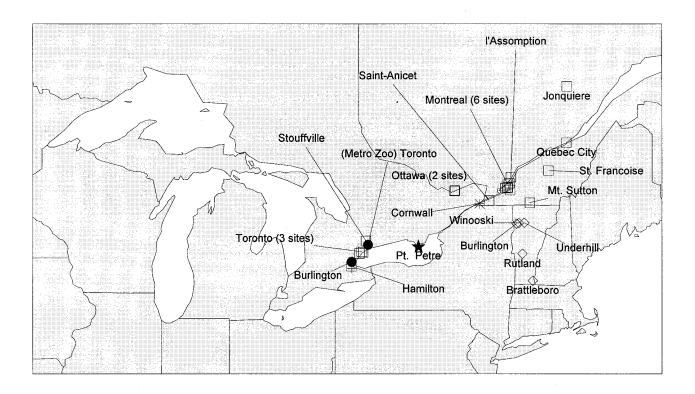
						Ai	r Monitorin	g		Pre	cipitation Mo	nitoring
Compound or Compound Group	Level	Pt. Petre	Toronto (*)	Hamilton (**)	Stouffville	London	Cornwall	Additional Sites	Pt. Petre	Toronto	Burlington	Additional Sites
distance from Lake (km)>		0.1	~ 1	~ 1	30	40	150	all ≥ 150 km	0.1	~ 1	~ 1	all $\ge 200  \text{km}$
character of site>		R	U	U	UR	U	U		R	U	U	
CHLOROBENZENES												
1,4-dichlorobenzene	11	t	t	t y	t	у	У	t:Ot1,2,SA, Mo1,2,4,5,As,MS,Fr; v: BV, Wn, Rt, Un, Bb	n g	g	g	
Tetr achlorobenz enes	Ш								n g	g	g	
Pentachlorobenzene	Ш								n g	g	g	
Hexachlorobenzene	I	ct	t	t					n g	g	g	
POLYCHLORINATED DIBENZO-P-D	IOXINS	& DIBEN	NZOFUR	ANS (PCI	DD/F'S) a	nd POLY	CHLOR IN/	ATED BIPHENYLS (PCB'S)			'	
PCDD/F's	I	t	t	tp				t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb				
PCB's	1	с						v. BV, Wn, Rt, Un, Bb	n			
POLYCYCLIC AROMATIC HYDROC	ARBON	IS									'	
Acenaphthene	l⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	n g	g	g	
Acenaphthylene	l⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	n g	g	g	
Anthracene	11	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb				
Benz (a) Anthracene	Ш	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	n			
Benzo (b) Fluor anthene	l⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	n			
Benzo (j) Fluoranthene	l⊦a											
Benzo (k) Fluoranthene	l⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb				
Benzo (a) Pyrene	I	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	n			
Benzo (e) Pyrene	l⊦a	ct	t	t			а	t: Mo2,Jo				
Benzo (g,h,i ) Perylene	Ш	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb				
Chrysene	I⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	n			
Dibenz(a,h)Anthracene	I⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb				
Dinitropyrenes	Ш											
Fluoranthene	I⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	n g	g	g	
Fluorene	l⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	n g	g	g	
Indeno(1,2,3-cd)Pyrene	l⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb				
Naphthalene	l⊦a	t	t	t y	t	у	У	t:Ot1,2,SA, Mo1,2,4,5,As,MS,Fr; v. BV, Wn, Rt, Un, Bb				
Phenanthrene	Ш	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	n			
Perylene	11	t	t	t			а	t: Mo2,Jo				
Pyrene	I⊦a	ct	t	t			а	t: Mo2,Jo; v: BV, Wn, Rt, Un, Bb	ng	g	g	

(\*) Three NAPS sites in Toronto; not all compounds are measured at all sites (VOC's at all 3; PAH, PCDD/F and COA at 1 site only; particulate metals at 2 sites)

(\*\*) One NAPS site in Hamilton; Five OMOEE air toxics monitoring sites in Hamilton: VOC's measured at all five, PCDD/F in air measured at one site (downtown)

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# Figure 5. Air and Precipitation Sampling Sites for One or More BVES Compounds in the Vicinity of Lake Ontario



	progra code (see	m
<u>symbol</u>	<u>Tbl 3)</u>	description
*	с	IADN Master Site (air and precipitation) The site in this figure, Point Petre, is a Canadian site; Air monitoring is conducted by Atmospheric Environment Service, Envr. Canada ("c") Precipitation monitoring by National Water Research Institute, Envr. Canada ("n") Precipitation monitoring also by Envr. Canada EHD (program "g") Total gaseous mercury monitored by Environment Canada (program "k") Toxaphene monitored by Environment Canada (program "b") Mercury was monitored at this site in program "r"
•	g	IADN Satellite Site, Environment Canada EHD (precipitation)
	t	Nat'l Air Pollutant Surveillance Network, Envr. Canada (air) Toronto (3 sites): VOC's (3 sites); particulate metals (2 sites); PAH's, PCDD/F's and selected COA substances (1 site); Stouffville, St. Anicet, l'Assomption, St. Francoise, Mt. Sutton: VOC's only; Quebec City: particulate metals are measured; Ottawa (2 sites): VOC's measured at both, particulate metals at 1 site Simcoe & Hamilton: VOC's, PCDD/F's, PAH's, & selected COA substances measured; At one site in Hamilton, OMOEE also measures PCDD/F's (program "p") Jonquiere: PAH's and PCDD/F's measured Montreal (six sites): VOC's (4 sites); PAH's and PCDD/F's (1 site); particulate metals (2 sites)
+	У	OMOEE Air Toxics Monitoring, VOC's (sites on this map: Hamilton, Ontario (5 sites))
*	a,y	OMOEE Air Toxics Monitoring, PAH's and VOC's (site on this map: Cornwall, Ontario)
$\diamond$	v	Vermont Agency of Natural Resources (air)

### 4. AMBIENT MONITORING of PERSISTENT TOXIC SUBSTANCES in the WATER of the GREAT LAKES

#### A. Overview of Monitoring Programs

There have been many measurements of toxic compounds in the water of the Great Lakes. These measurements are typically made on cruises of research vessels in which samples of lake water are collected at different locations ("stations") in a given Lake.

Table 12 gives a list of the different research cruises that carried out water measurements for each of the Great Lakes during the period 1986-1996. In some cases, information on 1997 cruises was included. For each cruise, the sponsoring agency is listed followed by the principal investigator, where known, in parentheses. Then, the seasons in which the cruises took place (Sp = Spring, Su = Summer, etc.) are listed, with the number of sampling stations in parentheses. Lastly, there is a reference to a "suite number", which is an abbreviation for the suite of chemical compounds measured in the particulate program. Each suite number corresponds to a column in Table 13 where the compounds measured for that particular cruise are indicated (compounds that were measured are marked with an "x")..

The rows in Table 13 list the BVES compounds as well as several others that have been measured in the water of the Great Lakes. Each of the columns correspond to the specific research projects or cruises that have carried out water measurements in one or more of the Great Lakes between 1986 and 1996. The table is organized by toxic compound groupings: Metals/Organometallics, Organochlorine Biocides, Industrial/Miscellaneous, Chlorobenzenes, Dioxins, PCB's, and PAH's.

In the following, a narrative summary of monitoring efforts will be presented, considering the research done in accordance with the above chemical groupings.

While a substantial effort was made to assemble a comprehensive list of cruises and compounds measured, it is acknowledged that it is likely that one or more efforts may have been missed. It is hoped that in the further review and discussion of this document, any errors of omission or content can be corrected.

As discussed in Section 2, the phase of a given pollutant in the water column may be important for estimating the net direction and rate of gas exchange. When it could be obtained, information regarding the phase of the measurements made — dissolved or associated with suspended sediment — will be presented.

Year	Lake Superior	Lake Huron	Lake Michigan	Lake Erie	Lake Ontario
1997	<ul> <li>EC(L'Italien): Sp &amp; Su, (14 stations), Suite 4;</li> <li>EPA: Sp (6 stations) Suite 6;</li> <li>DS: Sp(?) (5-9 sta.) Suite 11</li> </ul>	• EPA: Sp (6 stations) Suite 6	<ul> <li>EPA: Sp (6 stations) Suite 6;</li> <li>DS: Sp? (5-9 sta.) Suite 11</li> </ul>	• EPA: Sp (6 stations), Suite 6	• EPA: Sp (6 stations) Suite 6
1996	<ul> <li>EC(L'Italien): Sp &amp; Su, (14 stations), Suite 4;</li> <li>EPA: Sp (6 stations) Suite 6;</li> <li>DS: Sp(?) (5-9 sta.) Suite 11</li> </ul>	• EPA: Sp (6 stations) Suite 6	• EPA: Sp (6 stations) Suite 6	• EPA: Sp (6 stations), Suite 6	• EPA: Sp (6 stations) Suite 6
1995			<ul> <li>EPA, LMMBS: 8 cruises (41 stations), Suite 5;</li> <li>DS: Sp? (5-9 sta.) Suite 11</li> </ul>	<ul> <li>EC(L'Italien): Sp,Su,Fa (9 stations), Suite 3</li> </ul>	
1994			<ul> <li>EPA, LMMBS: 8 cruises (41 stations), Suite 5;</li> <li>DS: Sp? (5-9 sta.) Suite 11;</li> <li>Cook &amp; Burkhard: Fall (3 stations) Suite 12</li> </ul>	<ul> <li>EC(L'Italien): Sp,Su,Fa (9 stations), Suite 3</li> </ul>	<ul> <li>US EPA Region 2 (Yeh): Sept. (30 stations) Suite 9</li> </ul>
1993	<ul> <li>EPA: Sp Su (6+ stations) Suite 6;</li> <li>DS: Sp? (5-9 sta.) Suite 11</li> </ul>	• EPA: Sp Su (6+ stations) Suite 6	<ul> <li>EPA: Sp Su (6+ stations) Suite 6;</li> <li>DS: Sp? (5-9 sta.) Suite 11</li> <li>Cleckner et al. (1995): Fall (1 station); mercury</li> </ul>	• EPA: Sp Su (6+ stations) Suite 6	<ul> <li>EC(L'Italien): Sp,Su,Fa (6 stations) Suite 2;</li> <li>EPA: Sp Su (6+ stations) Suite 6</li> </ul>
1992	<ul> <li>EPA: Sp Su (6+ stations) Suite 6;</li> <li>EPA? NOAA? (Eisenreich et al.): Su (5 stations) Suite 7</li> </ul>	• EPA: Sp Su (6+ stations) Suite 6	• EPA: Sp Su (6+ stations) Suite 6	<ul> <li>EPA: Sp Su (6+ stations) Suite 6</li> </ul>	<ul> <li>EC(L'Italien): Sp,Su,Fa, Suite 2, 6 stations;</li> <li>EPA: SP SU (6+stations) Suite 6</li> </ul>
1991	<ul> <li>EPA: Sp Su (6+ stations) Suite 6;</li> <li>DS: Sp? (5-9 sta.) Suite 11</li> </ul>	<ul> <li>EPA: Sp Su (6+ stations) Suite 6;</li> <li>DS: Sp? (5-9 sta.) Suite 11</li> </ul>	<ul> <li>EPA: Sp Su (6+ stations) Suite 6;</li> <li>DS: Sp? (5-9 sta.) Suite 11</li> </ul>	<ul> <li>EPA: Sp Su (6+ stations) Suite 6;</li> <li>DS: Sp? (5-9 sta.) Suite 11</li> </ul>	<ul> <li>EPA: Sp Su (6+ sta.)Suite6;</li> <li>DS: Sp? (5-9 sta.) Suite 11;</li> <li>EC: Su (2 sta.) metals (including cadmium)</li> </ul>
1990	<ul> <li>EPA? NOAA? (Eisenreich et al.): Su (6 stations) Suite 7</li> </ul>	• EPA (McConnell,Bidleman): Su (10 stations) Suite 8	• EPA (McConnell,Bidleman): Su (11 stations) Suite 8	• EPA (McConnell,Bidleman): Su (18 stations) Suite 8	<ul> <li>EC(L'Italien) annual spring cruise, (47 stations) Suite 1;</li> <li>EPA (McConnell,Bidleman): Su (7 stations) Suite 8</li> </ul>
1989			• EPA (McConnell,Bidleman): Sp (3 stations -Green Bay) Suite 8		
1988	<ul> <li>EPA? NOAA? (Eisenreich et al.): Su (5 stations) Suite 7</li> </ul>	• EC(L'Italien): Sp, (63 stations for Hur+Ont) Suite 1			<ul> <li>EC(L'Italien): Sp, (63 stations for Hur+Ont) Suite 1</li> </ul>
1987	• EC(L'Italien): Sp, (46 stations for Sup+Hur) Suite 1	<ul> <li>EC(L'Italien): Sp, (46 stations for Sup+Hur) Suite 1</li> </ul>		<ul> <li>Gill and Bruland (1990) Su, (1 nearshore station only?) mercury</li> </ul>	• Gill and Bruland (1990) Su, (1 nearshore station only?) mercury
1986	<ul> <li>EC(L'Italien): Sp, (96 total stations for 4 lakes) Suite 1;</li> <li>EC IWD (Stevens&amp;Neilson) Sp (19 stations) Suite 10</li> <li>NOAA/EPA? (Eisenreich) Su (6 stations) Suite 7;</li> </ul>	<ul> <li>EC(L'Italien): Sp, (96 total stations for 4 lakes) Suite 1;</li> <li>EC IWD (Stevens&amp;Neilson): Sp (18 stations) Suite 10</li> </ul>		<ul> <li>EC(L'Italien): Sp (96 total stations for 4 lakes) Suite 1;</li> <li>EC IWD (Stevens&amp;Neilson) Sp (21 stations) Suite 10</li> </ul>	<ul> <li>EC(L'Italien): Sp (96 total stations for 4 lakes) Suite 1;</li> <li>EC IWD (Stevens&amp;Neilson): Sp (33 stations) Suite 10</li> </ul>

				Table 13: 0	Compoun	ds Measu	red in Wa	ater Monit	oring Pro	grams in t	he Great	Lakes			
		=		===================================	Suite 2	Suite 3	Suite 4	Suite 5	Suite 6	Suite 7	Suite 8	Suite 9	Suite 10	Suite 11	Suite 12
			-+		Julie 2	Suite 5	Suite 4	Suite 5	Suite 0	Suite	Suite 0	Suite 5	Suite IU	Sultern	(**)
		1	-	EC(L'Italien)	EC(L'Italien)	EC(L'Italien)	EC(L'Italien)	Lake Mich.	EPA	Eisenreich et al.	EPA (Bidleman)	US EPA	EC IWD	Swackhamer	Cook &
		    Le	evel	86-90 (Sup.,	92-93	94-95	96-97	Mass Bal.	Study: 91-97	86,88,90,92	GL Survey	Region II	Stevens	Great Lakes	Burkhard
chemical name			(1,11)	Hur., Erie, Ont.)	Ontario	Erie	Superior	Study (94-95)	Great Lakes	Superior	1989, 1990	Ontario, 94	Great Lakes 86	91-97	Michigan 94
		=	===	=======											
Metals / Organometallic															
		== =							======			======			
admium and cadmium compounds															
alkylated lead compounds			<u>!</u>												
mercury and mercury compounds tributyl tin	000688-75-3		1					x							
======================================			-	 == =================================											
Organochlorine Biocide															
aldrin					X	x	×					x	x		X
				<b>^</b>		~	^					~			~
atrazine	001912-24-9		-					x	x(93)			x			x
DEA (atrazine metabolite)								x							1
DIA (atrazine metabolite)								x							
		=		========					======						
Alpha-Chlordane			L	x	x	x	x					x			x
Gamma-Chlordane	005566-34-7		1	x	x	x	x					x			x
cis-Chlordane			1												x
trans-Chlordane			<u> </u>												x
trans-nonachlor	039765-80-5		+					x				x			x
cis-nonachlor	005103-73-1		 ===	    ==================								x			X
p,p'-DDT				===================================	x	X	×		x(92,93)?				x		
p,p-DD1 p,p'-DDD(p,p'-TDE)	000030-29-3		÷	X    X	x	x	x		x(92,93)?			x	x		x
p,p-000(p,p-102) p,p'-DDE			i t	X	× ×	x	x		x(92,93)?			x	x		x
o,p'-DDT	000789-02-6		÷t	x	x	x	x		x(92,93)?			~	x		x
					~	~	~								
dieldrin	000060-57-1		T	x	x	x	x					x	x		x
endrin	000072-20-8		п	x	x	x	x					x	x		x
endrin aldehyde	007421-93-4		11?		x	x	x								x
	:	=	===	=================											
Apha-Endosulphan	000959-98-8			x	x	x	x						x		x
Beta-Endosulphan	033213-65-9			X	x	x	x						x		x
		=	===	=======					======			======	======	======	
heptachlor	000076-44-8		11	X	x	x	x					x	x		x
heptachlor epoxide	001024-57-3			x	x	x	x					x	x		x
				====================================											
alpha-hexachlorocyclohexane beta-hexachlorocyclohexane	000319-84-6 000319-85-7		11 11	X	x	x	x				x	x	x		x
delta-hexachlorocyclohexane	000319-85-7		"												x
gamma-hexachlorocyclohexane	000058-89-9		"	x	x	x	x				x	x	x		x
mixed hexachlorocyclohexanes			<u> </u>			~	^ 	1			~	~	<u>^</u>		, î
				==========										===========	=========
methoxychlor	000072-43-5		П	x	x	x	x						x?		x
mirex	002385-85-5		Ι	x	x	x	x					x	x		x
photomirex			1?		x	x	x								x
		=		========		=======	=======		======================================		======================================	======	======	===========	=======
pentachlorophenol	000087-86-5														
toxaphene	008001-35-2		1	<u>                                      </u>								x		x	x
		== =	===				===========		==========		=======	============			========
Industrial/Miscellaneou			$\rightarrow$												
4-bromophenyl phenyl ether		== =							======================================				=======	=======	
3,3'-dichlorobenzidene								1							x ?
hexachloro-1,3-butadiene					x	x	x								x
4,4'-methylene bis(2-chloroaniline)	000101-14-4				*	<u>^</u>	Â	1							<u>^</u>
octachlorostyrene					x	x	x					x			x
hexachlorocyclopentadiene	000077-47-4				x	x	x								x
phthalates		1		11		x	x								
Chlorobenzenes		== =													
1,2-dichlorobenzene		_		x	x	X	X						x		
1,3-dichlorobenzene				x	x	x	x						x		
	000106-46-7		п	x	x	x	x	1	1			1	x	1	1

			1	. 1				1	1			1	1		
1,2,3-trichlorobenzen				x	x	x	x					x	X		x
1,2,4-trichlorobenzen				x	x	x	x					x	x		x
1,3,5-trichlorobenzen	e 000108-70-3			X	x	x	x					x	x		x
1,2,3,4-tetrachlorobenzen	e 000634-66-2			x	x	x	x					x	x		x
1,2,3,5-tetrachlorobenzen	e 000634-90-2		i li	1								x			x
1,2,4,5-tetrachlorobenzen				•								x			x
pentachlorobenzen				1	~	~	~					x	~		x
				•	x	x	x						x		
hexachlorobenzen				x	x	x	x					x	x		x
	-	_	_												
Polychlorinated Dibena	zo-P-Dioxin	s & I	Diþ	enzofurans											I
====================================			== =												
	D 001746-01-6	_	_	1		x	x								x
other PCDD/F congener		<u>    </u>		1		^	^								x
··· ··· ··· ··· ··· ··· ··· ··· ··· ··				1											
	-	_	== =		============										
Polychlorinated Bipher	nyls (PCBs)														
====================================		== ===	== =												
All 209 PCB congener	s			1					x						x (all 209?)
A subset of 135 PCB congener		1 1	- li	1								x			· · · · · · · · · · · · · · · · · · ·
		<u>    </u>	- 1.0	1						v/99 00 02)		^		-	
A subset of 82 PCB congener										x(88,90,92)					
A subset of 65 PCB congener		<u>    </u>		1				x		10			-		
A subset of 35 PCB congener		<u>    </u>	- 1.0	<u>  </u>						x(86)				<b></b>	I
	2 053469-21-9			1	X+								x?	<u> </u>	ļ'
Aroclor 125	4 011097-69-1			l	X+								x?		
Aroclor 126	0 011096-82-5			1	X+								x?		
Total PCBs	*	1	- li	x	x+?	X+	x	x?	x?	x?			x		
====================================		== ===									==========	===========			
Polycyclic Aromatic Hy	drocarbon		-												
. siyeyene Aromatic Hy		_	+											+	
			_		======							======			
II dinitropyrenes (mixed				1											l
====================================															
acenaphthen	e 000083-32-9	-	A												x
acenaphthylen	e 000208-96-8	-	A	x(88,90)	x	x	x					x			x
anthracen	e 000120-12-7	1 1		1								x			x
benz [ a ] anthracen						x	x			x(86)		x			x
dibenz [a,h] anthracen						~	~			x(86)		x			x
ll chrysen						x	x			x(86)		x			x
	e 000206-44-0				x	x	x			x(86)		x			x
benzo [ b ] fluoranthene										x(86)		x			x
benzo [ j ] fluoranthene	e 000205-82-3	-	A	1											
benzo [ k ] fluoranthen	e 000207-08-9	-	A							x(86)		x			x
benzo [b and k ] fluoranthen	e	-	A	x(88,90)	x	x	x								
	e 000086-73-7	Ш.	A		x	x	x			x(86)		x			x
	e 000095-13-6			x(88,90)	x										
				============											
naphthalen			- 10	1											
			- 1 "			x	x					x			x
2-chloronaphthalen				(···,··)	x	x	x								x
II 1- methylnaphthalen				x(88,90)	x	x	x								ļ'
II 2- methylnaphthalen	e 000091-57-6			x(88,90)	x	x	x								
1,2,3,4-Tetrahydronaphthalen	e			x(88,90)	x										
====================================		===:	==	=========					=============						
perylen	-			1							1				
benzo [ g,h,i ] perylen				x(88,90)	x	x	x	1		x(86)		x		1	x
dibenzo(ah)perylen		<u>.</u>			<u>^</u>	<u> </u>	~					~		1	
	e 000085-01-8			x(88,90)	x	x	x			x(86)		x		+	x
														+	
	e 000129-00-0				x	x	x			x(86)		x			x
benzo [ a ] pyrene				x(88,90)	x	x	x			x(86)		x		<b></b>	x
ll benzo [ e ] pyrene										x(86)		x		<b></b>	I
indeno [ 1,2,3-c,d ] pyrene	e 000193-39-5	-	A	x(88,90)	x	x	x			x(86)		x		ļ	x
====================================		== ===	== =												
Suspended Sediment	Properties														
===================================	- ·														
Total Suspended Solid										x?		x		+	
				1	x	x		X	x		x		x	+	X
Dissolved Organic Carbon (DOC		<u>  </u>			x	x		x	x	x?	x	x		+	x
Particulate Organic Carbon (POC		1	1		x	x		x	x	x?	x	x			x
Total Organic Carbon (TOC)		1		1				x	x	x?	x?	x		ļ	x
		===:	==												
		1 -													
				(	at particular com	pound was meas	ured in 1986 but	not in the other	years covered in	that column.					
		ses, e	.g.  x	(86) this imples th											
Notes: Where an entry contains a year	within parenthe														I
III	within parenthe an uncertainty a	s to w	heth	er the exact comp	ound listed has t	een measured.	Elimination Strat	tegy (BVFS)· add	itional compound	s measured are a	also included in t	his table			ļ
III         Notes:         Where an entry contains a year         Where the entry is x? there is a         In the column referring to Level	within parenthe an uncertainty a , I and II are des	s to w ignatio	heth ons f	er the exact comp for substances as	ound listed has t specified in the E	een measured. Binational Virtual					also included in t	his table			
III	within parenthe an uncertainty a , I and II are des added to form a	s to w ignatio group	heth ons f o of F	er the exact comp for substances as PAH compounds fo	ound listed has t specified in the E or this analysis; i	een measured. Sinational Virtual it is only one of m	any possible gro	oups of PAH's that	t could be consid		also included in t	his table			
Notes: Where an entry contains a year Where the entry is x? there is a In the column referring to Level II - A refers to PAH compounds * Reference to Total PCBs is u	within parenthe an uncertainty a , I and II are des added to form a usually simply t	s to w ignatio group ie sum	heth ons f o of F n of t	er the exact comp for substances as PAH compounds for the subset of cong	ound listed has t specified in the E or this analysis; i eners measured,	een measured. Sinational Virtual it is only one of n , not a category to	any possible gro be compared b	ups of PAH's that etween measurer	at could be consid nents.		also included in t	his table			
Notes: Where an entry contains a year Where the entry is x? there is a In the column referring to Level II - A refers to PAH compounds	within parenthe an uncertainty a , I and II are des added to form a usually simply t 94, and 95 there	s to w ignatio group le sum was r	heth ons f o of F n of t epor	er the exact comp for substances as PAH compounds for the subset of cong tedly PCB contam	ound listed has t specified in the E or this analysis; i eners measured,	een measured. Sinational Virtual it is only one of n , not a category to	any possible gro be compared b	ups of PAH's that etween measurer	at could be consid nents.		also included in t	his table			

#### i. Metal/Organometallics

Total mercury and methyl mercury were measured in Lake Michigan during 1994 and 1995 as part of the Lake Michigan Mass Balance Study (U.S. EPA 1995; Mason and Sullivan, 1997). This study was carried out over two years and mercury measurements were made during six cruises [June, August, and October/November 1994 and April, August, and September/October 1995], with approximately 20 samples collected per cruise. Determinations of both dissolved and particulate-phase concentrations were made.

Cleckner et al. (1995) report measurements of mercury at a site in Lake Michigan 6.4 km east of Chicago, where the water depth was about 12 meters. Eight samples at depths of 0.3 and 10 meters, and 7 samples of the surface micro-layer were collected over a 4-day period in September 1993 and analyzed for total mercury.

Gill and Bruland (1990) report measurements of particulate and dissolved mercury in Lake Ontario and Lake Erie (collected near the shores, as reported by Mason and Sullivan, 1997). Analysis of the dissolved fraction for total organo-mercury compounds and total reactive mercury was also carried out.

Reports of measurements of other metals in the Great Lakes were found in the literature. For example, measurements of cadmium in Lake Ontario were reported by Nriagu et al. (1993). In this study, ultra-clean techniques were used, and the authors stated that the "preliminary results obtained using the protocols described cast doubt on most of the published data on trace metal concentrations in the Great Lakes." *Nriagu found a dramatic decrease in cadmium with depth, suggesting that atmospheric deposition may be very important for this compound*. High levels of cadmium were also found near the bottom of the lake, presumably released from metal-rich sediments. Cadmium in the Great Lakes ecosystem — including potential human health effects from eating contaminated fish — was recently discussed by Bernier *et al.* (1995).

No data or reports of measurements for tributyltin or alkylated lead in the water of any of the Great Lakes could be identified through searches of the literature or discussion with government regulatory/monitoring personnel familiar with monitoring programs which have been conducted in the Great Lakes.

#### ii. Organochlorine Biocides

Stevens and Nielson (1989) report data from a cruise in spring 1986 for Lakes Superior, Huron, Erie and Ontario, carried out under the auspices of Environment Canada's Inland Waters Directorate. The set of organochlorine compounds measured were: aldrin, DDT and metabolites, dieldrin, endrin,  $\alpha$ - and  $\beta$ -Endosulphan, heptachlor, heptachlor epoxide,  $\alpha$ - and  $\gamma$ -HCHs, methoxychlor, and mirex.

Environment Canada's Inland Waters Directorate has continued to measure a range of compounds in this category (L'Italien 1993, 1996ab, 1997). For all the cruises during the 1986-90 period involving Lake Superior, Huron, Erie and Ontario, as well as the 92-93 study of Ontario, the 94-95 study of Erie, and the 96-97 study of Superior, the following organochlorine biocides have or are being measured: aldrin,  $\alpha$ - and  $\gamma$ -chlordane, DDT and metabolites, dieldrin, endrin,  $\alpha$ - and  $\beta$ -Endosulphan, heptachlor, heptachlor epoxide,  $\alpha$ - and  $\gamma$ -HCHs, methoxychlor, and mirex. In addition, except for the 1986-90 studies, endrin aldehyde and photomirex were also measured.

For 1986-90 Environment Canada work, total water column — dissolved plus particulate — concentrations are reported. In more recent work (1992-95), pollutant concentrations associated with the particulate phase (i.e., suspended sediment) were also attempted. In the studies of Lake Erie (1994-95) and Lake Ontario (1992-93), the levels of suspended sediments were too low to reliably measure particulate-phase concentrations. When measurements could be made, the proportion of many organic pollutants associated with the particulate phase was often on the order of 1% or less. Lake Superior has even lower suspended sediment concentrations than Lake Erie or Ontario, and thus, the fraction associated with suspended sediments may be even lower.

During 1996-97, Environment Canada is conducting 2 cruises (spring and summer) sampling only the surface waters. Sample volumes of 100 liters will be taken, and only the dissolved phase concentrations will be measured (L'Italien, 1997).

Bidleman and McConnell carried out cruises with the EPA in Green Bay in June of 1989 and Lakes Michigan, Huron, Erie and Ontario during August 1990. Organochlorine biocides measured included alpha- and gammahexachlorocyclohexanes. Measurements were reported for the dissolved phase only.

In 1992 and 1993, EPA annual cruises measured DDT and its metabolites for all five lakes and atrazine in 1993. The EPA makes reference to total water column concentrations only (SOLEC, 1994). During the Lake Michigan Mass Balance Study (1994-1995), organochlorine biocides measured included atrazine and metabolites and trans-nonachlor.

In 1994, the US EPA Region II utilized *Lake Guardian* research vessel for a September cruise on Lake Ontario. The following organochlorine compounds were measured: aldrin, atrazine,  $\alpha$ - and  $\gamma$ -chlordane, trans- and cis-nonachlor, p-p'-DDD, p,p'-DDE, dieldrin, endrin, heptachlor, heptachlor epoxide,  $\alpha$ - and  $\gamma$  -HCHs, mirex, and toxaphene. Measurements of pollutant concentrations in the total water-column and suspended sediment phase were made (U.S. EPA Region II, 1994).

#### iii. Industrial/Miscellaneous

For the two-year studies of Lake Ontario (1992-93), Lake Erie (1994-95) and Lake Superior (1996-97), Environment Canada measurements include the following compounds in this category: hexachloro-1,3 butadiene and octachlorostyrene. Hexachlorocyclopentadiene is/was also measured. For the latter two studies, measurements of a number of phthalates are included.

The only other measurement that could be identified for compounds in this pollutant category were those made for octachlorostyrene by the U.S. EPA Region II, during a September 1994 cruise on Lake Ontario (U.S. EPA Region II, 1994).

#### iv. Chlorobenzenes

Stevens and Nielson (Envr. Canada Inland Waters Directorate) carried out an annual spring cruise in 1986 for Lakes Superior, Huron, Erie and Ontario. The set of chlorobenzenes measured were: 1,2-, 1,3-, and 1,4-dichlorobenzene, 1,2,3-, 1,2,4-, and 1,3,5-trichlorobenzene, 1,2,3,4-tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene. Environment Canada has continued to measure the same group of chlorobenzenes as part of their cruises on the Great Lakes (L'Italien, 1993, 1996ab, 1997), including the cruises during the 1986-90 period involving Lakes Superior, Huron, Erie and Ontario, the 1992-93 study of Lake Ontario, the 1994-95 study of Lake Erie, and the 1996-97 study of Lake Superior.

U.S. EPA Region II measured several chlorobenzenes during their September 1994 cruise on Lake Ontario: 1,2,3-, 1,2,4-, and 1,3,5-trichlorobenzene, 1,2,3,4-, 1,2,3,5-, and 1,2,4,5-tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene.

### v. Polychlorinated Dibenzo-P-Dioxins & Dibenzofurans

There were water measurements made for dioxin compounds for 2,3,7,8tetrachloro-dibenzo-*p*-dioxin by Serge L'Italien and Environment Canada for Lake Erie in their 1994-1995 study and in their current study of Lake Superior for 1996 and 1997. In the case of Lake Erie (1994-95), they carried out cruises each year in the Spring, Summer and Fall, each with 9 sampling stations. The current project for Lake Superior involves two cruises a year in the Spring and Summer, each with 14 sampling stations.

Cook and Burkhard measured PCDD/F as an adjunct research project of the Lake Michigan Mass Balance Study in water samples from Lake Michigan in 1995 (Cook, 1997).<sup>4</sup>

<sup>&</sup>lt;sup>4</sup>. Cook and Burkhard report that EPA Method 1613 is being used for the analysis of dioxins and furans in water samples (Cook, 1997). This method includes

#### vi. Polychlorinated Biphenyls (PCBs)

There are a total of 209 different PCB congeners. The detailed lists of all the individual congener measurements have not been included in Table 13, as it was beyond the scope of this analysis to consider congener-specific details in relation to PCB monitoring programs. Instead, the total number of congeners measured is listed.<sup>5</sup>

Stevens and Nielson (Inland Waters Directorate) report PCB measurements in terms of Aroclors 1242, 1254, and 1260. These measurements were part of an annual spring cruise in 1986 for Lakes Superior, Huron, Erie and Ontario. Since 1986, Environment Canada has made measurements of total PCBs. In the case of the 92-93 study of Ontario, there is an explicit reference to the Aroclors 1242, 1254 and 1260, but it is not clear whether this same combination corresponds to references to total PCBs for the other years. Unfortunately, PCB contamination was discovered on the research vessels for the cruises in 1992, 1993, 1994, and 1995, making the PCB measurements questionable (L'Italien, 1997).

PCB measurements were made for all the cruises in which Eisenreich and others were involved for Lake Superior. In 1986 only 35 PCB congeners were measured but for 1988, 1990 and 1992 a subset of 82 congeners were measured. Jeremiason *et al.* (1994) refers to measurements for both the particulate and dissolved fractions for the 1988, 1990 and 1992 data. Baker and Eisenreich (1990) present data from the 1986 cruise; only dissolved-phase measurements are given and no reference is made to the particulate-phase concentrations.

The EPA has conducted annual cruises for all the Great Lakes since 1991 except for 1994-95 when their efforts were concentrated on the Lake Michigan Mass Balance Study. For the years 1991-1993, the EPA conducted both a spring and summer cruise for at least 6 stations per lake. For 1996 and 1997, there was just a spring cruise for six stations on each lake. For all these cruises, measurements have been made for all 209 PCB congeners, except for the Lake Michigan Mass Balance Study (LMMBS) which measured only 65 PCB congeners. Measurements of both dissolved and particulate-phase concentrations were made in the LMMBS.

measurements of all seventeen 2,3,7,8-substituted tetrachloro- through octachlorodibenzo-p-dioxins and dibenzofurans (e.g., Telliard, et al.).

<sup>&</sup>lt;sup>5</sup>. As mentioned as a note to Table 13, reference to "total PCBs" is usually simply the sum of the subset of congeners measured, not the actual total PCB's in the sample. Thus, "total PCB" data from measurement programs analyzing different subsets of the 209 PCB congeners cannot be compared easily.

Finally, U.S. EPA Region II measured a total of 135 PCB congeners during their September 1994 cruise on Lake Ontario.

# vii. Polycyclic Aromatic Hydrocarbons (PAHs)

Environment Canada has been regularly measuring a range of PAH's in the Great Lakes. In 1987, the only PAH measured for the cruises on Lake Superior and Huron was fluoranthene. For the cruises during 1988 and 1990 involving Lake Huron, and Ontario, as well as the 1992-93 study of Ontario, the 1994-95 study of Erie, and the 1996-97 study of Superior, the following PAH's have or are being measured: acenaphthylene, fluoranthene, benzo[b+k] fluoranthene, fluorene, 2-chloronaphthalene, 1-methylnaphthalene, 2-methylnaphthalene, benzo[g,h,i] perylene, phenanthrene, pyrene, benzo[a]pyrene, and indeno[1,2,3-c,d] pyrene. For the cruises during 1988 and 1990 involving Lake Huron, and Ontario, as well as the 92-93 study of Ontario, two additional PAH's were measured: indene, and 1,2,3,4-tetrahydronaphthalene. In the most recent cruises on Lake Erie in 1994-95 and Lake Superior in 1996-97, two additional PAH's are being measured: benz[a]anthracene and chrysene/triphenylene.

Eisenreich (sponsored by the EPA) conducted a cruise on Lake Superior in August of 1986 and measured the following PAH's: benz[a]anthracene, dibenz[a,h]anthracene, chrysene/triphenylene, fluoranthene, benzo[b]fluoranthene, benzo[k]fluoranthene, fluorene, benzo[g,h,i]perylene, phenanthrene, pyrene, benzo[a]pyrene, benzo[e]pyrene, and indeno[1,2,3-c,d]pyrene.

The September 1994 cruise on Lake Ontario by the US EPA Region II measured the following PAH's: acenaphthylene, anthracene, benz [a] anthracene, dibenz [a, h] anthracene, chrysene, fluoranthene, benzo [b] fluoranthene, benzo [k] fluoranthene, fluorene, naphthalene, benzo[g,h,i] perylene, phenanthrene, pyrene, benzo[a] pyrene, benzo[e]pyrene, and indeno[1,2,3-c,d] pyrene.

# viii. Suspended Sediment Properties

These properties are not part of the BVES compound list but are relevant for much of the work involving water measurements, similar to the importance of particulate measurements in interpreting air concentration data. Although, most cruises measure total suspended solids, we were unable to confirm whether all cruises have done so.

The last three entries in Table 13 are dissolved, particulate and total organic carbon, and again, it was not possible to determine during the course of this study whether such measurements had been made or not for some of the cruises. Information on the nature of suspended sediment measurements that were made (or not made) could not be obtained for the Environment Canada cruises and the Eisenreich et al. cruises in Lake Superior.

### B. Summary of Monitoring of Persistent Toxic Substances in the Water of the Great Lakes

The detailed information in Tables 12 and 13 and discussed in the narrative above is summarized in Table 14, below. In this table, the measurement programs for each BVES compound for each Great Lake for the period from 1986 - 1996 are summarized.

It can be noted that for many of the compounds that are being measured in water monitoring programs in the Great Lakes, the measurements are relatively infrequent. This may pose a challenge for the accurate estimate of the direction and rate of gas exchange for those compound for which this phenomenon is relevant. Typically, data from 1 or more years previous must be used when attempting to make such gas-exchange estimates.

each. Abbrev.: LvI = BVES Level; DS	= D. Swad		PA = U.S. Envr. Protection Ag	ted in parentheses,e .g., (14,14) in ency; S&N = Stevens & Nielson; LM ory, Duluth, Mn.		
Compound or Group	Lvi	Lake Superior	Lake Huron	Lake Michigan	Lake Erie	Lake Ontario
METALS / ORGANOMETALL	.ICS					
Total Cadmium	II					EC '91 (2) many different depths sampled (Nriagu etal., 1993)
Individual Cadmium Species	11					
Alkylated Lead	I					
Total Mercury	I			LMMBS '94-95, (6 cruises, ~ 20 samples per cruise) Cleckner '93 (1) [Cleckner et al. (1995)]	Gill and Bruland '87 (1? near shore?) [Gill and Bruland (1990)]	Gill and Bruland '87 (1? near shore?) [Gill and Bruland (1990)]
Individual Mercury Species	1			LMMBS '94-95 (6 cruises, ~ 20 samples per cruise) (methyl mercury)	Gill and Bruland '87 (1? near shore?) [Gill and Bruland (1990)] total dissolved organo- Hg and total dissolved reactive Hg	Gill and Bruland '87 (1? near shore?) [Gill and Bruland (1990)] total dissolved organo- Hg and total dissolved reactive Hg
Tributyltin	11					

### Table 14. Summary of Water Monitoring of BVES Persistent Toxic Substances in the Great Lakes (1986-1996)

Notes: The number of measurement locations or stations for each cruise of a particular study is indicated in parentheses, e.g., (14,14) indicates that there were two cruises with 14 stations each.

Abbrev.: Lvl = BVES Level; DS = D. Swackhamer; EC = Envr. Canada; EPA = U.S. Envr. Protection Agency; S&N = Stevens & Nielson; LMMBS = L. Mich Mass Balance Study;

C&B = Cook and Burkhard, EPA National Health and Environmental Effects Research Laboratory, Duluth, Mn.

Compound or Group	LvI	Lake Superior	Lake Huron	Lake Michigan	Lake Erie	Lake Ontario
ORGANOCHLORINE BIOC	IDES					
Aldrin / Dieldrin	1	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38)	C&B '94 (3)	S&N '86 (21) EC '86 (21) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Chlordane		EC '86 (22) EC '87 (22) EC '96-97 (14,14)	EC '86 (38) EC '87 (38) EC '88 (38)	C&B '94 (3)	EC '86 (21) EC '94-95 (9,9,9)	EC '86,88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
DDT / DDD / DDE	1	S&N '86 (19) EC '86 (22) EC '87 (22) EPA '92-93 (6) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38) EPA '92-93 (6)	EPA '92-93 (6) C&B '94 (3)	S&N '86 (21) EC '86 (21) EPA '92-93 (6) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6) EPA '92-93 (6) EPA Region II '94 (30)
Endrin	11	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38)	C&B '94 (3)	S&N '86 (21) EC '86 (21) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Heptachlor / Heptachlor Epoxide	II	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38)	C&B '94 (3)	S&N '86 (21) EC '86 (21) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Hexachlorocyclohexanes	11	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38) EPA (Bidleman) '90 (10)	EPA (Bidleman) '89 (3- Green Bay) EPA (Bidleman) '90 (11) C&B '94 (3)	S&N '86 (21) EC '86 (21) EPA (Bidleman) '90 (18) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EPA (Bidleman) '90 (7) EC '92-93 (6,6,6) EPA Region II '94 (30)
Methoxychlor	II	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38)	C&B '94 (3)	S&N '86 (21) EC '86 (21) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6)
Mirex	I	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38)	C&B '94 (3)	S&N '86 (21) EC '86 (21) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Pentachlorophenol	П					
Toxaphene	1	DS '91,93,96,97 (~7)	DS '91 (~7)	DS '91,93,94,95,97 (~7) C&B '94 (3)	DS '91 (~7)	DS '91 (~7) EPA Region II '94 (30)

Table 14.	Sumr	nary of Water Monito	ring of BVES Persi	stent Toxic Substand	ces in the Great Lak	<b>es</b> (1986-1996)
each. Abbrev.: LvI = BVES Level; DS	S = D. Swa	cations or stations for each cruise ackhamer; EC = Envr. Canada; EF National Health and Environmen	PA = U.S. Envr. Protection Age	ncy; S&N = Stevens & Nielson; LM		
Compound or Group	Lvi	Lake Superior	Lake Huron	Lake Michigan	Lake Erie	Lake Ontario
INDUSTRIAL / MISCELLANI	EOUS					
4-Brom op he nyl Phenyl Ether	П			C&B '94 (3) (?)		
3,3'-Dichlorobenzidene	П					
Hexachloro-1,3-Butadiene	П	EC '96-97 (14,14)		C&B '94 (3)	EC '94-95 (9,9,9)	EC '92-93 (6,6,6)
4,4'-Methylene bis (2-Chloroaniline)	II					
Octachlorostyrene	I	EC '96-97 (14,14)		C&B '94 (3)	EC '94-95 (9,9,9)	EC '92-93 (6,6,6) EPA Region II '94 (30)
CHLOROBENZENES						
1,4-dichlorobenzene	11	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38)		S&N '86 (21) EC '86 (21) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6)
Tetrachlorobenzenes	II	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38)	C&B '94 (3)	S&N '86 (21) EC '86 (21) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Pentachlorobenzene	II	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38)	C&B '94 (3)	S&N '86 (21) EC '86 (21) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Hexachlorobenzene	I	S&N '86 (19) EC '86 (22) EC '87 (22) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38)	C&B '94 (3)	S&N '86 (21) EC '86 (21) EC '94-95 (9,9,9)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)

Table 14.	Sum	mary of Water Monito	oring of BVES Persi	istent Toxic Substanc	es in the Great Lak	<b>es</b> (1986-1996)
each. Abbrev.: Lvl = BVES Level; D	S = D. Sw		PA = U.S. Envr. Protection Age	ed in parentheses,e .g., (14,14) ind ency; S&N = Stevens & Nielson; LM ry, Duluth, Mn.		
Compound or Group	Lvi	Lake Superior	Lake Huron	Lake Michigan	Lake Erie	Lake Ontario
POLYCHLORINATED DIBE	NZO-P-D	IOXINS & DIBENZOFURANS (	PCDD/FS)			
2,3,7,8-TCDD	I	EC '96-97 (14,14)		C&B '94 (3)	EC '94-95 (9.9.9)	
other PCDD/F congeners	I			C&B '94 (3)		
POLYCHLORINATED BIPH	ENYLS (I	PCB'S)				
PCB's	1	S&N '86 (19) Eisenreich, '86,88,90,92 (5 or 6) EC '86 (22) EC '87 (22) EPA '91-93 (6,6) EPA '96-97 (6) EC '96-97 (14,14)	S&N '86 (18) EC '86 (38) EC '87 (38) EC '88 (38) EPA '91-93 (6,6) EPA '96-97 (6)	EPA '91-93 (6,6) C&B '94 (3) LMMBS '94-95, (8 cruises, 41 stations) EPA '96-97 (6)	S&N '86 (21) EPA '91-93 (6,6) EC '94-95 (9.9.9) EPA '96-97 (6)	S&N '86 (33) EC '86,88,90 (55) EC '92-93 (6,6,6) EPA '91-93 (6,6) EPA Region II '94 (30) EPA '96-97 (6)
POLYCYCLIC AROMATIC	HYDROC	ARBONS				
Acenaphthene	II-a			C&B '94 (3)		
Acenapthylene	II-a	EC '96-97 (14,14)	EC '88 (38)	C&B '94 (3)	EC '94-95 (9.9.9)	EC '88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Anthracene	П			C&B '94 (3)		EPA Region II '94 (30)
Benz [ a ] Anthracene	II	Eisenreich, '86 (6) EC '96-97 (14,14)		C&B '94 (3)	EC '94-95 (9.9.9)	EPA Region II '94 (30)
Benzo [ b ]Fluoranthene	II-a	Eisenreich, '86 (6)		C&B '94 (3)		EPA Region II '94 (30)
Benzo [ j ]Fluoranthene	II-a					
Benzo [ k]Fluoranthene	II-a	Eisenreich, '86 (6)		C&B '94 (3)		EPA Region II '94 (30)
Benzo [ a ] Pyrene	1	Eisenreich, '86 (6) EC '96-97 (14,14)	EC '88 (38)	C&B '94 (3)	EC '94-95 (9.9.9)	EC '88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Benzo [ e ] Pyrene	II-a	Eisenreich, '86 (6)				EPA Region II '94 (30)
Benzo [g,h,i ] Perylene	П	Eisenreich, '86 (6) EC '96-97 (14,14)	EC '88 (38)	C&B '94 (3)	EC '94-95 (9.9.9)	EC '88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)

### Table 14. Summary of Water Monitoring of BVES Persistent Toxic Substances in the Great Lakes (1986-1996)

Notes: The number of measurement locations or stations for each cruise of a particular study is indicated in parentheses, e.g., (14,14) indicates that there were two cruises with 14 stations each.

Abbrev.:	LvI =	BVES Level; DS	5 = D. Swackhamer; EC = Envr. Canada; EPA = U.S. Envr. Protection Ager	ncy; S&N = Stevens & Nielson; LMMBS = L. Mich Mass Balance Study;

C&B = Cook and Burkhard, EPA National Health and Environmental Effects Research Laboratory, Duluth, Mn.

Compound or Group	Lvl	Lake Superior	Lake Huron	Lake Michigan	Lake Erie	Lake Ontario
POLYCYCLIC AROMATIC H	YDROCA	ARBONS (continued)				
Chrysene	II-a	Eisenreich, '86 (6) EC '96-97 (14,14)		C&B '94 (3)	EC '94-95 (9.9.9)	EPA Region II '94 (30)
Dibenz [a,h]Anthracene	II-a	Eisenreich, '86 (6)		C&B '94 (3)		EPA Region II '94 (30)
Dinitropyrenes	П					
Fluoranthene	II-a	EC '87 (22) Eisenreich, '86 (6) EC '96-97 (14,14)	EC '87-88 (38)	C&B '94 (3)	EC '94-95 (9.9.9)	EC '88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Fluorene	II-a	Eisenreich, '86 (6) EC '96-97 (14,14)	EC '88 (38)	C&B '94 (3)	EC '94-95 (9.9.9)	EC '88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Indeno [ 1,2,3-cd ] Pyrene	II-a	Eisenreich, '86 (6) EC '96-97 (14,14)	EC '88 (38)	C&B '94 (3)	EC '94-95 (9.9.9)	EC '88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Naphthalene	II-a	EC '96-97 (14,14)		C&B '94 (3)	EC '94-95 (9.9.9)	EPA Region II '94 (30)
Phenanthrene	II	Eisenreich, '86 (6) EC '96-97 (14,14)	EC '88 (38)	C&B '94 (3)	EC '94-95 (9.9.9)	EC '88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Perylene	П					
Pyrene	II-a	Eisenreich, '86 (6) EC '96-97 (14,14)	EC '88 (38)	C&B '94 (3)	EC '94-95 (9.9.9)	EC '88,90 (55) EC '92-93 (6,6,6) EPA Region II '94 (30)
Other PAH's		EC '87 (22) EC '96-97 (14,14)	EC '88 (38)	C&B '94 (3)	EC '94-95 (9.9.9)	EC '88,90 (55) EC '92-93 (6,6,6)

# 5. SPATIAL REPRESENTATIVENESS of AMBIENT MONITORING DATA for the ESTIMATION of the NET LOADING of PERSISTENT TOXIC SUBSTANCES to the GREAT LAKES

It has long been recognized that a fundamental question in relation to the use of ambient monitoring data to estimate loadings to the Great Lakes is the following:

Can data from one or only a few sampling locations in the proximity of a particular Great Lake be used to accurately estimate deposition to the entire Lake?

This question has particular relevance to the use of the data from the Integrated Atmospheric Deposition Network (IADN), which has one master station for each Great Lake. In estimating atmospheric deposition to each of the Great Lakes using IADN data, data from the one master station per Lake is used to represent the atmospheric concentrations of pollutants for the entire Lake (Hoff et al., 1996).

There have been several recent attempts to characterize spatial variability in air and water concentrations in the Great Lakes.

# A. Lake Michigan Urban Air Toxics Study

In the Lake Michigan Urban Air Toxics Study (LMUATS) (Keeler, 1994; Pirrone *et al.*, 1995ab), ambient air samples were collected at three sites for one month during the summer of 1991:

- (1) on a building roof at Illinois Institute of Technology in Chicago, Illinois;
- (2) onboard a research ship located on Lake Michigan 5-10 km offshore of Chicago;
- (3) at South Haven, MI, a rural site approximately 130 km northeast of Chicago, about 3 km inland from the opposite (eastern) shore of Lake Michigan

In Table 15 below, the BVES pollutants (Table 1) measured in LMUATS are listed.

Additional biocides (trans-nonachlor, atrazine, chloropyrifos, simazine, metolachlor, and alachlor) and PAH's (fluorenone, retene, cyclopenta(c,d)pyrene, and coronene) were also measured in the organics series.

Water concentrations were not measured for any of the organics during the study.

•	or which air measurements were Jan Urban Air Toxics Study
Metals/Organometallics	PCB's
Mercury (total)	8 specific congeners
Lead (total)	homologue group totals, e.g., monochloro-PCB's, dichloro-PCB's, etc.
Organochlorine Biocides	
α-HCH ; γ-HCH	PAH's
aldrin and dieldrin	Naphthalene
mirex	Acenaphthylene
$\alpha$ - and $\gamma$ -chlordane and trans-nonachlor	Acenaphthene
4,4'-DDT, p,p'-DDE, and p,p'-DDD	Fluorene
	Phenanthrene
Industrial Miscellaneous:	Anthracene
hexachlorobutadiene	Fluoranthene
[no data presented in Keeler (1994) or	Pyrene
Pirrone <i>et al.</i> (1995b)]	Benz[a]Anthracene
	Chrysene
Chlorobenzenes	Benzofluoranthenes (mixed)
hexachlorobenzene	Benzo[e]Pyrene
	Benzo[a]Pyrene
1,4-dichlorobenzene	Indeno[1,2,3-c,d ]Pyrene
[no data presented in Keeler	Dibenz[a,h]Anthracene
(1994) or Pirrone <i>et al.</i> (1995b)]	Benzo[g,h,i]Perylene

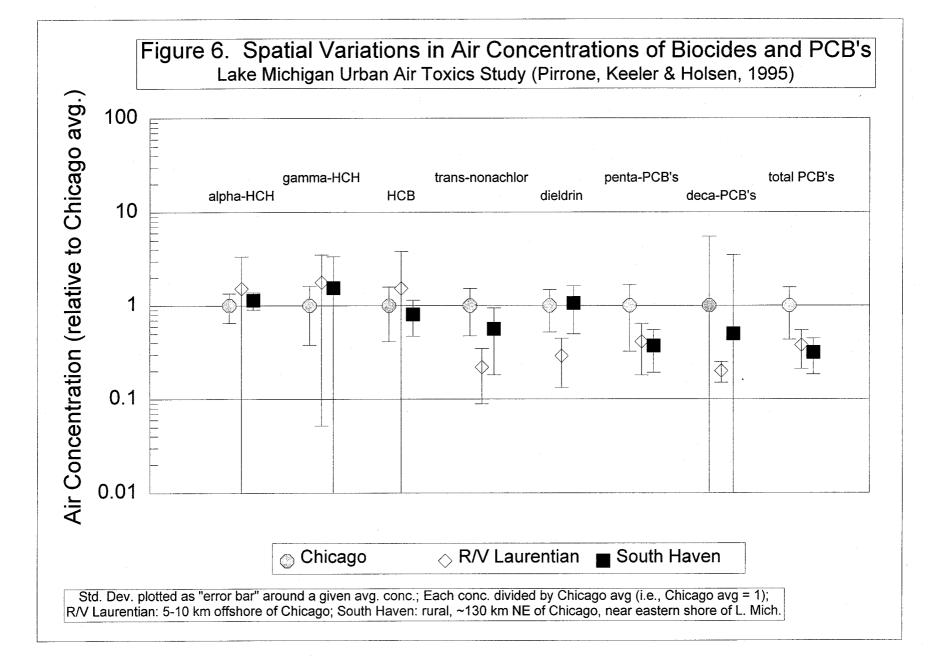
Land-based measurements were made for one month from July 8 to August 9, 1991. Daily 12-hour samples were taken at each land-based site. Over-lake samples 5-10 km offshore of Chicago were taken during two sampling periods (5 days and 3 days in duration). Approximately 17 samples were taken during these periods at the over-lake site. Total (vapor + particle) concentrations of organic compounds were measured.

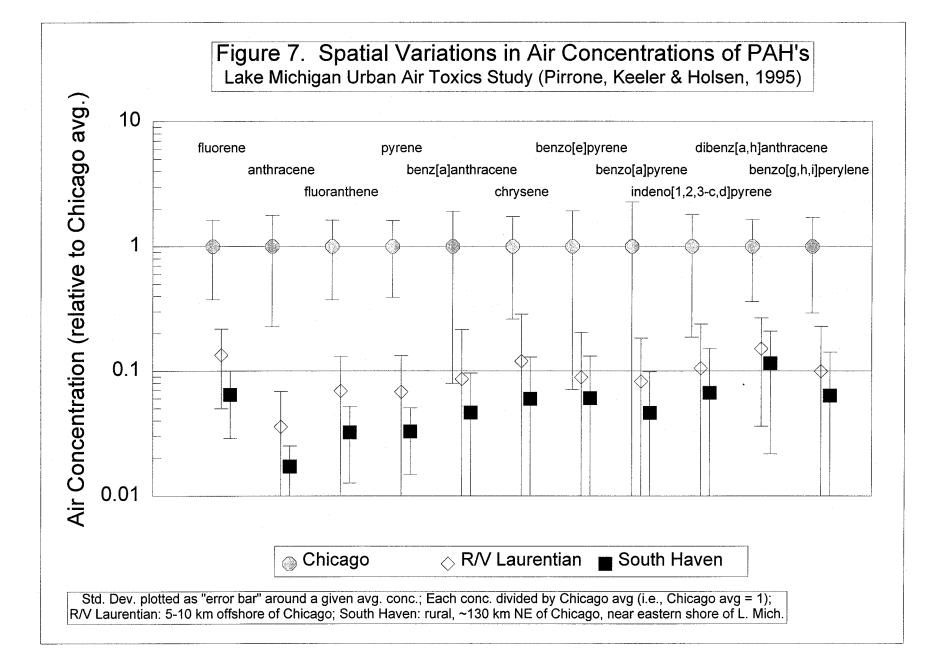
In Figures 6 and 7, below, the average concentrations of biocides, PCB's, and PAH's measured at the three sampling sites are shown.<sup>6</sup> For each pollutant, the concentrations at each site have been normalized (i.e., divided by) the average concentration of the pollutant measured at Chicago. The data presented in Figure 6 show that while there were significant differences in ambient air concentrations

<sup>&</sup>lt;sup>6</sup>. Only a subset of the total number of compounds has been plotted in Figures 2 and 3. The compounds plotted are those for which data were given in Pirrone et al. (1995b). Data for additional compounds is given by Keeler (1994), but these were not included in these figures.

between different sites, there was no consistent trend for organochlorine biocides and HCB, i.e., the relative ordering of sites with respect to concentration changes from compound to compound. For the PCB's (also shown in Figure 6) and PAH's (shown in Figure 7), there was a consistent, relatively dramatic difference between the sites. The site in Chicago consistently had the highest concentration. Also, for each individual PAH and for total PCB's, the over-lake site 5-10 km offshore of Chicago had higher concentrations than the South Haven site on the other side of the Lake. For PCB's and PAH's, the relative magnitude of concentrations was generally consistent with the following pattern:

[Chicago] >> [over-lake, 5-10 km off-shore of Chicago] > [South Haven]





### B. Lake Michigan Mass Balance Study

Results of measurements made in the Lake Michigan Mass Balance Study are being assembled and presented elsewhere, and these data were considered to be outside of the scope of this report. One set of the LMMBS data was obtained in the course of this analysis, however. A brief mention of some of these data will be made here, as they are particularly relevant to the question of spatial variability.

Over-lake air measurements in the northern, central, and southern portions of Lake Michigan during a summer 1994 cruise are summarized in Table 16 (Sweet, 1996).

		ummary of Air M ichigan During \$	-						
Lake Region	<b>Pollutant Concentrations</b> (average values, pg/m <sup>3</sup> , for samples taken at two locations in each region)								
	∑PCB's	DDE	Dieldrin	Benzo(a)Pyrene	Lead				
Northern Region	4,800	5	63	28	8,000				
Central Region	4,600	8	51	27	9,000				
Southern Region	5,100	16	150	180	86,000				
	Ratio	-	Concentration in e Central-North (	the Southern Re Concentration	egion				
	1.1	2.5	2.6	6.5	10.1				

It can be seen from the data in the above table that the over-lake concentrations of the reported pollutants were roughly the same in the north and central portions of Lake Michigan (during the cruise), but that for some pollutants, most notably benzo(a)pyrene and lead, the levels in the air in the southern portion of the Lake were relatively much higher.

As has been observed by many, emissions from the heavily populated urban centers surrounding the southern portion of Lake Michigan (e.g., the Chicago metropolitan area) can exert a strong influence on the levels of pollution in nearby reigons of the Lake. For pollutants associated with combustion processes in urban areas such as benzo(a)pyrene and lead, this influence is clearly seen in the above

data. In the measurements made during this cruise, large spatial variability was not found for PCB's, DDE, or dieldrin in the air above Lake Michigan.

In another part of this study, air and precipitation measurements were made at nine sites in the vicinity of Lake Michigan. These data are summarized in Table 17 (from Sweet, 1996). The data shown are the average of 12 monthly composite samples. As discussed by Clyde Sweet (1996), these data, together with those of Table 16, show several interesting patterns, including the following:

- The highest values of PCB's, BaP, and lead in air and precipitation were found in Chicago IL.
- The highest concentrations of DDE in air and precipitation were found at South Haven MI, suggesting a localized source (e.g., an area of high past use of DDT).
- There was a distinct south-north trend for dieldrin in both air and precipitation (concentrations higher at the southern sites, lower at the northern sites), consistent with the heavy past use of aldrin in agricultural areas south of Lake Michigan (dieldrin in a persistent breakdown product of aldrin).
- The second highest average measured air concentration of PCB's (750 pg/m<sup>3</sup>) was at Beaver Island, a remote island in the northern part of Lake Michigan. The concentration of PCB's was four times higher here than at the other remote sampling site, at Sleeping Bear Dunes (the IADN master station for Lake Michigan). The concentrations of PCB's at Beaver Island in the summer months was particularly high.
- Except for PCB's, concentrations measured at the shoreline sites (Table 17) were similar to those measured at over-lake sites (Table 16). Over-water concentrations were significantly higher at over-water sites than at all shore-line sites except for Chicago.
- The above two results are consistent with a net volatilization of PCB's from Lake Michigan during the summer months, increasing the over-water concentrations and the concentration at Beaver Island (air parcels arriving at Beaver Island spent a relatively long time traversing the water of Lake Michigan). [This result is consistent with that of Hornbuckle *et al.* (1993) who found higher concentrations of PCB's over the water surface of Green Bay than over surrounding land areas.]
- Average concentrations of lead in precipitation was fairly uniform at all sampling sites.
- Average concentrations of BaP in precipitation were generally highest in urban areas, with the concentration at Chicago 10 100 times greater than at any other location.
- Average concentrations of PCB's in precipitation were relatively uniform at the shoreline sampling stations, although the levels measured at Chicago were somewhat higher than any other location measured.
- Based on the measurements, estimating the overall wet deposition to Lake Michigan using data from Sleeping Bear Dunes would seem to be a relatively accurate approach. However, estimating the overall dry deposition of from data at Sleeping Bear Dunes would underestimate the loading, especially for BaP and lead.

Monitoring	7. Summary							Concentra	ations in Air	and Precip				
Location (all sites near the	Orientation Relative to	Orientation West Char Relative to Orientation of San	General Character of Sampling Site	∑PC	(air con CB's		in units of p DE		entrations in drin		n in units of I)Pyrene		ad	
shore except Bondville)	Lake				Sile	air (vapor)	precip.	air (vapor)	precip.	air (vapor)	precip.	air (particle phase)	precip.	air (particle phase)
Manitowoc, W I	central	west	urban	218	5.1	16	0.3	40	1.5	34	1.8	12,000	2,700	
Chiwaukee, W I	south- central	west	urban	282	2.4	13	0.3	69	1.0	63	14	15,000	1,900	
Chicago, IL	south	west	urban	2400	13.0	33	1.0	247	5.8	809	178	38,000	3,300	
Bondville, IL	~200 km sout	h of Chicago	urban	196	1.8	9	0.1	606		20	< 1	8,000	1,400	
Indiana Dunes, IN	south	east	urban	571	1.7	14	0.1	162	1.7	172	15	17,000	2,200	
South Haven, MI	south- central	east	rural	241	5.0	814	2.5	245	2.3	59	3.0	8,000	2,100	
Muskegon, MI	central	east	urban	504	3.2	27	0.3	103	0.2	48	4.9	10,000	2,100	
Sleeping Bear Dunes, MI	north- central	east	remote	183	2.4	14	0.1	40	0.7	9	1.8	2,000	1,500	
Beaver Island, MI	north	center	remote	750	1.7	3	0.3	19	0.5	12	3.4	4,000	1,300	

F

## C. Ambient Air Monitoring of Persistent Toxic Substances Conducted by the Michigan Department of Environmental Quality

Organochlorine compounds in the vapor phase were measured at three sites in the region of the upper Great Lakes during 1990-1991 (Monosmith and Hermanson, 1996), and additional work in the region is continuing (Monosmith, 1997; Hermanson *et al.*, 1997). During the 1990-1991 sampling period, the following compounds were measured, simultaneously, at sites near Grand Traverse Bay, Saginaw Bay, and Sault Ste. Marie, Michigan:

- 25 PCB congeners<sup>7</sup>
- Hexachlorobenzene
- p,p'-DDT and p,p'-DDE
- α-Hexachlorocyclohexane (HCH) and γ-HCH

One 48-hour sample was taken each month, from November 1990 through October 1991, at each of the three sites.

Concentrations of all compounds measured tended to be higher in the summer than in the rest of the year. The sampling site at Sault Ste. Marie had the lowest concentrations of PCB's, while the site at Traverse had the highest concentrations of most PCB's measured. For HCB, DDT/DDE, and the HCH's, there was no consistent pattern to the relative concentrations at the three sites, and for some sampling dates, there were large differences in the concentrations at the three sites.

Additional compounds being measured in the continuing work include 16 different PAH compounds, dieldrin, mirex, and 13 trace metals (not including mercury) (Monosmith 1997).

<sup>&</sup>lt;sup>7</sup>. The 25 congeners chosen for detailed analysis all had relatively high vapor pressures and were expected to exist predominantly in the vapor phase in the atmosphere. The 25 congeners were represented by 14 gas-chromatograph ("GC") peaks; i.e., there were 14 peaks quantified in detail, with each peak either representing a single PCB congener or a group of PCB congeners. ∑PCB, representing the sum of 121 congeners, was also measured.

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## 6. SUMMARY AND CONCLUSIONS

This analysis has considered ambient monitoring programs for a set of persistent toxic substances in the Great Lakes region, and the use of these monitoring data to estimate loadings to the Great Lakes.

The compounds considered were those listed in the Binational Virtual Elimination Strategy (BVES), as shown in Table 1, above.

This analysis focused primarily on the universe of ongoing, systematic, government-sponsored monitoring efforts in the Great Lakes region for air, precipitation, and the water of the Great Lakes. Detailed consideration of the monitoring data themselves or of loading estimates derived from these data was not possible to include given the limited resources available for this analysis.

An overall summary of the coverage of the BVES compounds in the monitoring programs in the Great Lakes region is given in Table 18, below, based on Sections 3 and 4 above.

In considering Table 18, and other issues discussed in this report, the following overall conclusions and observations emerge. Many of the compound-specific issues below are summarized in Table 19, as well.

- Systematic, ambient monitoring programs for air, precipitation, and water exist in the Great Lakes region for many of the BVES compounds. These data can be used to:
  - estimate the overall loading of monitored compounds to the Great Lakes (although uncertainties related to spatial representativeness for some compounds' measurements, e.g., PAH's, is a concern, and, for all of the measured compounds, there are uncertainties in the methodology of estimating the overall loading);
  - provide information on source regions, using back-trajectory modeling;
  - provide validation data for comprehensive atmospheric fate and transport models.
- It would be extremely helpful if a system could be established for making Great Lakes regional air, precipitation, and water monitoring data readily available to the public and to the research community.

- For some of the BVES compounds, there are little or no measurements of ambient air and precipitation concentrations in the Great Lakes region.
- The following BVES compounds are not being monitored comprehensively in air and/or precipitation in the IADN program [see table 19 for additional notes regarding these and other compounds]:
  - mercury speciation (i.e., total mercury is being measured at several locations, but, individual species are not being measured, e.g, the proportion of gaseous mercury that is Hg°, HgCl<sub>2</sub>, HgO, methylmercury, etc.)
  - alkylated lead
  - 4-bromophenyl phenyl ether
  - 3,3'-dichlorobenzidene
  - 1,4-dichlorobenzene
  - PCDD/F's
  - dinitropyrenes
  - hexachloro-1,3-butadiene
  - pentachlorophenol
  - toxaphene
- The addition of 4,4'-methylene bis (2-chloroanaline) and tributyltin compounds to the list of substances monitored might be implemented on a temporary basis, to determine the potential significance of the air pathway to Great Lakes loadings for these compounds.
- Some of the BVES compounds are being monitored in Canada at one or more sites in the Great Lakes region, but not in the U.S. For example, Environment Canada has a recently established a monitoring program for octachlorostyrene, dinitropyrenes, and pentachlorophenol (in conjunction with the COA); no monitoring efforts for these compounds could be found in the U.S. in the Great Lakes region. As another example, the Canadian IADN satellite stations monitor many compounds that are not monitored at any of the U.S. sites, including Endrin, Heptachlor, Methoxychlor, Mirex, and Octachlorostyrene (only at OMOEE IADN satellite sites).
- For some of the BVES compounds, there are limited or no measurements in the water of the Great Lakes. For compounds that exist to a certain extent in the vapor phase in the atmosphere, a lack of water concentration data makes the estimation of loading to the lakes difficult.
- For many of the compounds that are being measured in water monitoring programs in the Great Lakes, the measurements are relatively infrequent. This may pose a challenge for the accurate estimate of the direction and rate of gas

exchange for those compound for which this phenomenon is relevant. Typically, data from 1 or more years previous must be used when attempting to make such gas-exchange estimates. No measurements of any BVES compounds could be identified in Lake Huron within the last 5 years.

- The spatial representativeness of monitoring locations is an issue for at least some of the BVES compounds.
  - For PAH's and other urban-source-associated pollutants, there may be strong influences of urban plumes on the loading to a given Lake, and, the representativeness of rural sampling locations is in question.
  - For PCB's and other pollutants which may be volatilizing from one or more of the Lakes, the representativeness of shoreline stations (as opposed to over-water monitoring locations) is in question.
- In addition to the spatial representativeness issue and the lack of measurements for some compounds, an incomplete understanding and incomplete monitoring of the various mass transfer processes between the atmosphere and the Lakes makes the monitoring-based estimation of the net loadings from the atmosphere to the Great Lakes somewhat uncertain.
- As an example, the deposition velocity for particle-associated material will depend on the particle size distribution, but, chemical specific particle size distributions are infrequently if ever measured for BVES pollutants in the Great Lakes region.
- Fog deposition, droplet resuspension, and indirect atmospheric loading (discussed briefly in Section 2) are not characterized well enough at this time to even attempt to include them in the loading estimates.
- The results of recent research, including the Lake Michigan Mass Balance Study and the AEOLOS research program, are eagerly anticipated. These may serve to significantly increase the understanding of atmospheric loading to the Great Lakes.

Table 18. Summary of Air, Precipitation and Water Monitoring Programs for BVES Substances in the Great Lakes Region													
column#'s (see notes for descriptions)→	2	3	4	5	6	7	8	9	10	11	12	13	14
		V/P	LRT	Air Monitoring: G.L. Region		Precipitation Moni	Precipitation Monitoring: G.L. Region		er Moni	Monitoring: 1992-1			
Compound or Compound Group	Level	Vapor-Particle	Long-Range Air	Integrated Atmos. Dep. Network	Other Programs	Integrated Atmos. Dep. Network	Other Programs	Superior	Michigan	Huron	Erie	Ontario	Loading Estimate Possible?
MERCURY and MERCURY COMPOUND	s											·	
Elemental Mercury (Hg°)	I	v	1	-	-	-	-	-	-	-	-	-	0
Divalent Mercury (e.g., HgCl <sub>2</sub> )	Ι	v	2	-	-	-	-	-	-	-	-	-	0
Monom eth yl Mercury	Ι	v	3	-	-	-	-	-	-	-	-	-	0
Total Gaseous Mercury	I	VV	1-3	-	ksqflr	-	-	-	-	-	-	-	~ 0 (F)
Particulate Mercury	I	PP	2	-	tqflr	-	-	-	-	-	-	-	½ (F)
Total Mercury I		v	1-2	-		-	dsrqflr	-	m	-	-	-	~ 0 (F)
OTHER METALS / ORGANOMETALLICS													
Alkylated Lead	1	(V V)	2	-	-		-	-	-	-	-	-	0
Total Cadmium (Cd)	11	ΡР	2	ceou	t m w v	gnou	-	-	-	-	-	-	1
Individual Cd Species	П	РР	2	-	-		-	-	-	-	-	-	0
TributyItin Compounds	П	v/p	3 - B	-	-		-	-	-	-	-	-	0
ORGANOCHLORINE BIOCIDES													
Aldrin	I	V V	3 - 4	сео	-	gno	-	е	u	-	е	eu	~ 1
Dieldrin	I	V V	3 - 4	ceou	m w	gnou	-	е	u	-	е	eu	~ 1
Chlordane	I			ceou	w	gnou	-	е	u	-	е	eu	~ 1
DDT / DDD / DDE	I	v / p	2	ceou	m w	gnou	-	eu	um	u	eu	eu	1
Endrin	П	V V	3	c e	-	g n	-	е	u	-	е	eu	~ 1
Heptachlor / Heptachlor Epoxide	П	V V	3 - 4	сео	-	gno	-	е	u	-	е	eu	~ 1
Hexachlorocyclohexanes	11	v	1 - 2	ceou	m w	gnou	-	е	u	-	е	eu	~ 1
Methoxychlor	П	v / p	3	сео	-	gno	-	е	u	-	е	е	~ 1
Mirex	1	(V V)	2	сео	-	n o	-	е	u	-	е	eu	~ 1
Pentachlorophenol	11	VV	2	-	t		-	-	-	-	-	-	0
Toxaphene	1	v / p	2	-	b h		-	s	s	-	-	u	~ ½ (C)

INDUSTRIAL / MISCELLANEOUS													
4-Bromophenyl Phenyl Ether	II	VV	3	-	-		-	-	u?	-	-	-	0
3,3'-Dichlorobenzidene	П	v / p	2	-	-		-	-	-	-	-	-	0
Hexachloro-1,3-Butadiene	П	VV	1	-	t w v		-	е	u	-	е	е	~ 1
4,4'-Methylene bis (2-Chloroaniline)	П	v	3 - 4	-	-		-	-	-	-	-	-	0
Octachlorostyrene	I	v	2	0	t	0	-	е	u	-	е	eu	~ 1
CHLOROBENZENES													
1,4-dichlorobenzene	П	V V	2	-	t y w v	g n	-	е	-	-	е	е	~ 1
Tetrachlorobenzenes	II	V V	1	0	-	gno	-	е	u	-	е	eu	~ 1
Pentachlorobenzene	II	VV	1	0	-	gno	-	е	u	-	е	eu	~ 1
Hexachlorobenzene	I	VV	1	ceou	t m	gnou	-	е	u	-	е	eu	~ 1
POLYCHLORINATED DIBENZO-P-DIOXINS & DIBENZOFURANS (PCDD/F'S) and POLYCHLORINATED BIPHENYLS (PCB'S)													
PCDD/F's	T	v / p	2	-	tpv		p (Dorset)	е	u	-	е	-	½ (G)
PCB's	Ι	v / p	2	ceou	m v	nou	-	eu	mu	u	eu	eu	1
POLYCYCLIC AROMATIC HYDROCARBONS													
Acenaphthene	II-a	V V		c e u	t a v	gnu	-	-	u	-	-	-	~ ½ (H)
Acenaphthylene	II-a	V V		ceu	tam v	gnu	-	е	u	-	е	eu	1
Anthracene	П	VV	3	ceu	tam v	u	-	-	u	-	-	u	~ ½ (H)
Benz (a) Anthracene	П	v / p	2	ceou	tawv	nou	-	е	u	-	е	u	~ 1
Benzo (b) Fluoranthene	II-a	v / p		ceou	t a v	nou	-	-	u	-	-	u	~ 1
Benzo (j) Fluoranthene	II-a	v / p		-	-		-	-	-	-	-	-	0
Benzo (k) Fluoranthene	II-a	v / p		ceou	t a v	o u	-	-	u	-	-	u	~ 1
Benzo (a) Pyrene	T	v / p	2	ceou	tam wv	nou	-	е	u	-	е	eu	~ 1
Benzo (e) Pyrene	II-a	v / p		ceu	ta	u	-	-	-	-	-	u	~ ½ (H)
Benzo (g,h,i ) Perylene	П	v / p	2	ceou	t a v	o u	-	е	u	-	е	eu	~ 1
Chrysene	II-a	v / p		ceou	tawv	nou	-	е	u	-	е	u	~ 1
Dibenz (a,h) Anthracene	II-a	ΡΡ		ceou	tav	o u	-	-	u	-	-	u	1
Dinitropyrenes	П	v / p	2	-	t	-	-	-	-	-	-	-	0
Fluoranthene	II-a	v		ceou	tam wv	gnou	-	е	u	-	е	eu	~ 1
Fluorene	II-a	VV		ceou	tamv	gnu	-	е	u	-	е	eu	~ 1
Indeno (1,2,3-cd) Pyrene	II-a	р		ceou	t a v	o u	-	е	u	-	е	eu	~ 1
Naphthalene	II-a	VV		-	t y m w v	-	-	е	u	-	е	u	~ 1/2
Phenanthrene	II	(V V)	3	ceou	tam wv	nou	-	е	u	-	е	eu	~ 1
Perylene	II	v / p	2	-	ta	-	-	-	-	-	-	-	0
Pyrene	II-a	v		ceou	tam wv	gnou	-	е	u	-	е	eu	~ 1

Column-b	y-Column Notes for Table 18.
2	Level I or II compounds [as designated in the Binational Virtual Elimination Strategy (see text)]
3	Vapor/Particle Partitioning Characteristics of Compound(s) ("V/P") (based on Cohen, 1997a)
	<ul> <li>P P = Compound is expected to exist almost entirely in the particle phase in the atmosphere (fraction adsorbed ≥ 98% under all conditions)</li> <li>p = Compound is expected to exist mostly in the particle phase in the atmosphere (fraction adsorbed ≥ 90% under all conditions)</li> <li>v/p = Compound is expected to exist in significant proportions in both the particle phase and the vapor phase as conditions vary</li> <li>v = Compound is expected to exist mostly in the vapor phase in the atmosphere (fraction adsorbed ≤ 10% under all conditions)</li> <li>V V = Compound is expected to exist almost entirely in the vapor phase in the atmosphere (fraction adsorbed ≤ 2% under all conditions)</li> </ul>
4	L.R.T. (Long Range Transport) Potential (Rating)
	This analysis has not included quantitative, integrated modeling in its scope, due to time limitations. Instead, a qualitative approach to the assessment of long-range atmospheric transport has been taken. In this approach, pollutants have been generally categorized as to the relative importance of various fate mechanisms. Based on these considerations, an attempt has been made to qualitatively estimate the atmospheric lifetimes of each of the pollutants considered in this analysis. A "Long Range Air Transport Potential" rating scale of 1-4 is defined as follows:
	• Rating = 1 The pollutant is extemely long-lived in the atmosphere, with an atmospheric lifetime ~ a year or longer; distribution of the pollutant is essentially global.
	• Rating = 2 The pollutant is relatively long-lived in the atmosphere, with atmospheric residence times on the order of at least a week to perhaps several months; long range transport can definitely occur over 1000's to 10,000's of kilometers.
	• Rating = 3 The pollutant is relatively short-lived in the atmosphere, with atmospheric residence times on the order of several hours to a few days; atmospheric transport may occur on regional, mesoscale distances, perhaps of several 100's to perhaps even a 1000 kilometers.
	• Rating = 4 The pollutant is extremely short-lived in the atmosphere, with atmospheric residence times on the order of seconds to minutes to at most an hour or so; with such pollutants, atmospheric transport of emissions will be limited to the local region around the source.
5 - 8	Air and Precipitation Measurements made in the Integrated Atmospheric Deposition Network (IADN) and other programs. See Table 3 for description of codes.
9-13	Any Measurements in Great Lakes Water in the period between 1992 and 1996? e = measurements by Environment Canada; u = measurements by the U.S. Environmental Protection Agency m = measurements in the Lake Michigan Mass Balance Study (sponsored by U.S. EPA); s = sampling conducted by D. Swackhamer et al. (personal communication)
14	Loading Estimate Possible? [ 0 = no; 1 = yes; ½ = somewhat possible]
	• This is a somewhat subjective estimate of the relative possibility of making estimates of loading using existing monitoring data.
	• If the compound is measured at most or all IADN sites (and has some data water data available, if gas exchange may be important) then it is assigned a value of "1", i.e., it is possible to at least try to make an estimate of loadings to the Lakes
	• If the compound is not measured in the air or precipitation in the Great Lakes region, then it is assumed that the possibility of a loading estimate is "0"
	• A possibility of "1/2" suggests that for various reasons, the estimation feasability lies somewhere between the two extremes above.
	• a "~" in front of a value indicates that air/water gas exchange may be an important pathway, but, that water measurements have not been made in all of the Great Lakes. For the lakes in which water measurements are available, rough estimates of loading can be made; for the lakes for which measurement are not available, very rough loading estimates could be made by using, for example, the average water concentrations measured in other lakes.

ADDITION	ADDITIONAL NOTES for Table 18, relating to columns 2,4, and 14 only.				
II-a	For this analysis, several additional PAH's were considered, consisting of the remaining compounds in the EPA's 16-PAH list and the ATSDR 17-PAH list.				
В	TributyItin existence in significant amounts in the atmosphere is uncertain.				
	If present in the atmosphere, it's overall long range transport rating might be on the order of "3".				
с	Measurements of total toxaphene and major congeners are being made at Eagle Harbor and Pt. Petre in a collaborative research project involving Envr Canada & Indiana Univ.				
F	Except for Lake Michigan, mercury has not been measured in the water of the Great Lakes; speciation info of Hg in air/water phases limited; thus, estimates are difficult to make.				
	Total gaseous and particulate-phase Hg were measured at the five IADN master stations during 1995 and 1996 in a research program. This program stopped in December 1996.				
	Sampling at Eagle Harbor is continuing, supported by a trust fund. Sampling at Pt. Petre is being conducted for total gaseous mercury, sponsored by Environment Canada.				
	Sampling for total mercury in precipitation is being conducted at two IADN satellite sites (Brule River, Wisconsin and Dorset, Ontario) as part of the Mercury Deposition Network.				
	The reliability of "particulate mercury" measurements reportedly being made by at least some of the programs is not clear. Thus, the rating of "½" for particulate mercury may be an overestimate.				
G	Vapor- and particulate-phase PCDD/F is being measured in multi-site monitoring programs in Canada, sponsored by Environment Canada and the Ontario Ministry of Environment and Energy, and at several sites in Vermont.				
	These programs were not designed specifically for estimating loading to the Great Lakes, but, there are several sites in the region.				
н	Only water data for one Lake could be found; thus estimates could be made for that Lake.				
	Crude estimates could possibly be made for other Great Lakes, using the water concentrations found in the one Lake, as a first approximation.				

Compound or Group	Air and Precipitation Monitoring Issues	Water Monitoring Issues
<ul> <li>Alkylated Lead</li> <li>4-Bromophenyl Phenyl Ether</li> <li>3,3'-Dichlorobenzidene</li> <li>4,4'-Methylene bis (2-chloroaniline)</li> <li>Tributyltin</li> </ul>	<ul> <li>Not included in any of the air or precipitation monitoring programs identified</li> </ul>	<ul> <li>Not included in any of the water monitoring programs identified</li> </ul>
<ul><li>Pentachlorophenol</li><li>Dinitropyrenes</li><li>Perylene</li></ul>	<ul> <li>Limited air monitoring identified in Canada only</li> <li>No precipitation monitoring</li> </ul>	<ul> <li>Not included in any of the water monitoring programs identified</li> </ul>
• PAH's in general	<ul> <li>Spatial representativeness issue: PAH's are emitted primarily in urban areas.</li> </ul>	<ul> <li>No monitoring in Lake Huron in the las five years</li> </ul>
<ul> <li>PCDD/F (dioxins and furans)</li> </ul>	<ul> <li>Limited number of G reat Lakes monitoring stations in Canada only, near Lakes Erie and Ontario;</li> <li>No monitoring identified near Lakes Superior, Michigan, or Huron;</li> <li>Spation representativeness: monitoring primarily in urban locations, although, e.g., air monitoring at Pt. Petre.</li> <li>Only one site (Dorset) for precipitation monitoring</li> </ul>	<ul> <li>Monitoring by Envr. Canada for 2,3,7,8-TCDD in Lake Erie (1994, 1995) and Lake Superior (1996, 1997);</li> <li>Monitoring by Cook and Burkhard (US EPA) in Lake Michigan in 1994</li> <li>No monitoring in Lake Huron or Lake Ontario in the last five years</li> </ul>
• Mercury	<ul> <li>Limited number of monitoring location;</li> <li>Little or no gas-phase speciation data being collected</li> </ul>	<ul> <li>Systematic measurements only identified for Lake Michigan</li> </ul>
• Toxaphene	<ul> <li>Monitoring only at 2 sites (Eagle Harbor and Pt. Petre)</li> <li>No current measurements in precipitation could be identified</li> </ul>	<ul> <li>No monitoring in Lake Huron or Lake Erie in the last five years</li> </ul>
<ul> <li>Aldrin</li> <li>Endrin</li> <li>Heptachlor &amp; Heptachlor Epoxide</li> <li>Methoxychlor</li> <li>Mirex</li> <li>Octachlorostyrene</li> </ul>	• Measured atsome or all Canadian IADN stations, but not at U.S. sampling sites in the Great Lakes Region	<ul> <li>No monitoring in Lake Huron in the las five years</li> </ul>
• DDT/DDD/DDE	• Spatial representativeness: high concentrations in the air at South Haven — are there other hot spots in the Great Lakes region?	
<ul> <li>Hexachloro-1,3-butadiene</li> </ul>	<ul> <li>Not part of IADN, but, measured in other programs in Can. &amp; U.S.</li> <li>It may be possible to estimate loadings for many of the Lakes;</li> <li>No data near Lake Superior.</li> </ul>	<ul> <li>No monitoring in Lake Huron in the las five years</li> </ul>
<ul><li>1,4-dichlorobenzene</li><li>tetrachlorobenzenes</li><li>pentachlorobenzenes</li></ul>	<ul> <li>Limited air measurements in the Great Lakes region</li> </ul>	<ul> <li>For all, no monitoring in Lake Huron in the last five years</li> <li>For 1,4-DCB, none in Lk. Mich. either</li> </ul>
• PCB's	<ul> <li>Different sets of PCB's being monitored in different programs</li> <li>Since one or more lakes may be volatilizing PCB's, representativeness of shoreline monitoring stations is in question</li> </ul>	<ul> <li>Different sets of PCB's being monitored in different programs</li> </ul>

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