The Transport and Deposition of Persistent Toxic Substances to the Great Lakes

V. Summary

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by

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Preface

This report was commissioned by the IJC International Air Quality Advisory Board. It is the last in a series of five closely related reports prepared for the Board which examine a set of persistent toxic substances identified in the Binational Virtual Elimination Strategy (BVES). The first report deals with the capability of the BVES compounds to be transported long distances in the atmosphere. The second report deals with the status and capabilities of available emissions inventories for BVES compounds. The third and fourth reports deal with modeling the atmospheric transport and deposition of BVES compounds to the Great Lakes and monitoring of these compounds in the Great Lakes region, respectively. This fifth report is a summary of the first four studies.

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, held May 21-22, in Romulus Michigan.

The material presented here was collected and analyzed during the period from October 1996 through May 1997.

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1. Introduction

The extensive bi-national program to improve the environmental quality of the Great Lakes has recently established a challenging policy with respect to a major class of pollutants: the virtual elimination of persistent toxic substances (see Canada-United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes Basin). To this end, the Strategy "...seeks to reduce and virtually eliminate the use, generation or release of persistent toxic substances resulting from human activity." It follows that, to implement this policy, the anthropogenic sources that generate these pollutants must be identified and ranked with respect to their respective contributions to the Great Lakes loadings.

In the case of those pollutants that enter the Great Lakes basin from the air, this becomes an especially challenging task.

First, since the atmospheric transport of such substances can occur over thousands of kilometers, and even globally, the sources that need to be identified and evaluated can number in the thousands.

Second, in order to set priorities for action, the types of sources and the geographic source regions must be ranked with respect to their influence on the total airborne loading of each pollutant to a given lake.

Third, it is important to know how the atmospheric deposition of the pollutants varies over time in different regions of a lake, for such variation will influence their environmental impact and the degree to which ambient monitoring programs — which are necessarily limited in space and time — are representative of the overall process.

Fourth, in order to set overall priorities for action, at the sources, to virtually eliminate the pollutants, it is necessary not only to rank them individually and by region with respect to their loading contribution, but the *total* amount of each pollutant entering the Great Lakes through the atmospheric pathway must also be compared with loadings from other pathways, such as liquid discharges from industrial and municipal sources.

This report is an initial assessment of the current status of the methods that are being developed in response to this challenge and of the information that they have generated.

The recently issued Canada-United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes Basin (Envr. Canada and U.S. EPA, 1996) identified a set of 27 chemicals or chemical groups to be targeted for virtual elimination, including twelve Level I substances or groups and fifteen Level II substances or groups. This initiative is commonly referred to as the Binational Virtual Elimination Strategy (BVES), and in this report, the selected pollutants will be referred to as "BVES compounds." These compounds include:

- Thirteen Level I substances (or groups) consisting of the eleven Critical Pollutants identified by the IJC’s Great Lakes Water Quality Board, plus two additional Critical Pollutants identified by the Lake Superior Lakewide Management Plan (LaMP) and the Lake Ontario Toxics Management Plan (octachlorostyrene and chlordane).

- Fourteen Level II substances (or groups of substances) consisting of pollutants 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 Remedial Action Plan (RAP) processes and the Great Lakes Water Quality Guidance in the United States.

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

Three basic methods have been developed to provide information about atmospheric loadings of persistent toxic substances to the Great Lakes. These are shown schematically in Figure 1. In the first approach, ambient monitoring is used to estimate the overall loading to a given Lake. This approach is discussed in Section 5, below. In the second approach, information about source regions that contribute significantly to observed ambient concentrations is developed by analyzing where the sampled air parcels came from. This approach is discussed, below, in Section 4. In the third approach, a comprehensive atmospheric model is used to attempt to link emission from individual sources or source regions to atmospheric deposition to a given Lake. This approach will also be discussed, below, in Section 4.

Initially, as a screening-level analytical approach, pollutants can be evaluated as to their relative capability to be transported long distances in the atmosphere, i.e., their atmospheric lifetime. Compounds expected to have relatively short atmospheric lifetimes would not be able to be transported long distances by the winds, and so, attention can be primarily focused on local or regional sources. On the other hand, for compounds with relatively long atmospheric lifetimes, distant sources might make significant contributions to the atmospheric loading to a given Lake. A general discussion of atmospheric lifetimes of persistent toxic substances is presented in Section 2.

In addition to helping to qualitatively address questions about the potential, relative importance of local, regional, and distant sources, these basic considerations about the atmospheric behavior of pollutants are integral to several of the above quantitative methodologies. Knowledge about atmospheric deposition phenomena — which, of course, affect atmospheric lifetimes — is used to link ambient measurements to deposition, especially for dry deposition[1] to the Lakes (discussed in Section 5).

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[1]. Dry deposition is that which occurs in the absence of precipitation. Wet deposition occurs as a result of precipitation.

Similarly, the atmospheric fate phenomena considered in estimating atmospheric lifetimes are fundamental components of comprehensive modeling analyses discussed in Section 4.

Emissions inventories, discussed in Section 3, are another fundamental component of efforts to understand the atmospheric loading pathway. They are required as input for comprehensive atmospheric fate and transport models, and, by themselves, yield information about potential sources of atmospheric loadings.

In Section 6, an overall summary of the above analytical components of the atmospheric loading pathway is presented. In this last section, Table 7 is presented as an overall summary.[2]

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[2]. For brevity, summary data will generally be presented only in this final table. In Sections 2 - 5, reference will be made to particular columns in Table 7, and these columns in this table will serve as overall summaries of these previous sections.

Table 1. Compounds and Compound Groups Targeted in the Binational Virtual Elimination Strategy (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 (α,β,δ, and γ) (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

Figure 1. The Atmospheric Deposition Pathway: Conceptual Elements

	Emissions of Pollutants to the Air		Atmospheric Transport and Fate		Ambient Monitoring		Atmospheric Deposition to a Great Lake
SEMI-EMPIRICAL LOADING ESTIMATES	***Atmospheric Deposition to a Given Lake?***						
			atmospheric deposition knowledge used in estimating deposition from ambient measurements (especially for dry deposition)		ambient air and precipitation measurements also: water measurements needed to estimate for vapor phase pollutant	÷	estimated wet and dry atmospheric deposition
			C dry deposition? *C fog deposition?* *C resuspension?*		*C existence of measurements?* *C spatial variations?*		
BACK-TRAJECTORY MODELING	***Source Regions Contributing to Atmospheric Concentrations at a Given Location?***						
	information about source regions contributing to observed measurements	2	back-trajectories estimated from meteorological model	2	ambient air and/or precipitation measurements		
	C how did atmospheric fate and dispersion processes affect transport?		*C what "level" of the atmosphere should be used to estimate back-trajectory?*		*C are there significant variations in measurements over time?*		
COMPRE-HENSIVE ATMOSPHERIC FATE AND TRANSPORT MODELING	***Total Deposition to a Given Lake Arising from All Specified Emissions? Contributions from Each Specified Emission? Relative Importance of Local vs. Regional vs. Continental vs. Global Sources?***						
	Geographically and Temporally Resolved Emissions Inventory	÷	Meteorology; Advection and Dispersion; Physical/Chemical Micro-Environment; Pollutant Behavior in Micro-Envr.	÷	ambient air monitoring data essential for model validation	÷	predicted atmospheric deposition
	C existence of inventory? *C is it complete?* *C quality assurance?* *C confidential?*		*C do emissions from different sources interact?* *C modeling domain?* *C atmospheric fate processes?* *C grasshopper phenomena?*		*C existence of measurements?* *C if predictions don't match measurements, why?*		*C source-receptor relationships?* *C targeted efforts to prevent loadings can be discussed and implemented*

2. The Capability of Persistent Toxic Substances to Be Subjected to Long Range Atmospheric Transport

As a screening-level approach to assess the atmospheric behavior of persistent toxic substances, their relative potential for long-range air transport was evaluated (Cohen, 1997a). A brief summary of the methodologies employed and the principal findings will be presented here. As with all of the analyses discussed here, the principal set of compounds considered were the set of persistent toxic substances identified as Level I or Level II compounds in the Binational Virtual Elimination Strategy (BVES), listed above in Table 1.

First, evidence regarding the *possibility* for atmospheric transport was examined by determining whether a particular substance is in fact emitted into the atmosphere. Direct measurements of emissions and/or evidence of detectable levels of a substance in the atmosphere were considered sufficient evidence for the possibility of atmospheric transport. Such evidence for the possibility of atmospheric transport was found for all of the BVES persistent toxic substances, except perhaps for tributyltin compounds.[3]

As the atmospheric fate of any pollutant is strongly influenced by the extent to which it exists in the vapor phase or as associated with particles (Bidleman, 1988), an attempt was made to estimate the vapor/particle partitioning characteristics of each of the compounds. The basic results of this analysis are shown in Column 3 of Table 7.

Next, an assessment was made, based on a consideration of the phase in which each compound is likely to occur in the atmosphere, of the degree to which each substance will respond to a group of factors that influence its fate in the atmosphere. Factors considered were:

- resistance to destructive reactions in atmosphere;
- resistance to photolytic degradation;
- resistance to gas-phase rainfall washout;
- if and/or when the substance is associated with particles in the

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[3]. Loadings of tributyltin compounds to aquatic ecosystems are generally considered to arise from direct emissions to water (i.e., not as a result of atmospheric deposition). Essentially no information on air emissions or atmospheric concentrations of these compounds could be found. Limited information was found, however, for another alkyl-tin compound, triphenyltin acetate (Stab, et al., 1994). This study was consistent with others that have found volatilization and atmospheric transport to be a significant fate pathway for this compound. Given the chemical similarity, it is possible that tributyltin compounds are emitted to and are transported in the air. While the relative significance of the atmospheric pathway for loading is presumed to be small, it would, of course, be useful to attempt to quantitatively confirm this.

atmosphere, whether the substance is associated with *small* particles.

The influences of these factors were used, in a general way, to estimate the substances' approximate lifetime in the atmosphere. Thus, for example, hexachlorobenzene, which is extremely resistant to all of the factors that tend to remove it from the atmosphere, has an estimated atmospheric lifetime measured in years and atmospheric mobility over global distances. In contrast, a substance very susceptible to photolytic degradation while airborne [e.g., 4,4'-methylene bis (2-chloroaniline)] may have a lifetime measured in minutes and thus very limited atmospheric mobility. The estimated approximate overall atmospheric lifetime reflects the factor(s) which exerts the greatest influence on the removal of the substance from the atmosphere.

Finally, evidence that the substances have been found at distances remote from their points of emission (for example, the occurrence of DDT in the Arctic) was examined.

Based on the above conceptual approach, the BVES persistent toxic substances were classified into four qualitative categories (or "ratings") of atmospheric long-range transport (LRT) potential:

LRT Rating 1: Atmospheric half-life: one year or more
Geographic distribution from sources: global

LRT Rating 2: Atmospheric half-life: one week to a few months
Geographic distribution from sources: 1,000-10,000 km

LRT Rating 3: Atmospheric half-life, a few hours to a few days
Geographic distribution from sources: 100-1,000 km

LRT Rating 4: Atmospheric half-life, seconds to minutes
Geographic distribution from sources: local

The estimated ratings are shown in Column 4 of Table 7.

The following observations and conclusions emerged from this analysis:

Many of the BVES persistent toxic substances appear to be capable of regional, continental and even global transport.

For essentially all compounds, significant uncertainties make accurate, quantitative estimates of atmospheric lifetime difficult. Rates of photolysis and atmospheric reactions (e.g., with hydroxyl radical) were particularly uncertain. For some compounds, even basic physical chemical properties are not well characterized. Moreover, for some compounds, estimates of vapor/particle partitioning in the atmosphere are somewhat uncertain.

For many of the BVES substances, the "grass-hopper" effect will probably serve to increase atmospheric transport distances.[4] It was difficult to quantitatively estimate the effect of this phenomenon on long-range transport potential, other than to state that the approximated lifetimes for some of the pollutants probably represented a lower-bound estimate.

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[4]. In the grass-hopper phenomenon, a pollutant is deposited to the earth's surface, but is then re-emitted. It is believed that some compounds can undergo several — and even many — of such "hops".

3. The Status of Emissions Inventories of Persistent Toxic Substances in the United States and Canada

Emissions inventories are essential for evaluating the role of the atmospheric deposition pathway for the loading of pollutants to the Great Lakes. An analysis of the status and capabilities of emissions inventories of BVES substances was carried out (Cohen 1997b) and will be summarized here.

Through consultation with the IJC International Air Quality Advisory Board, a primary set of emissions inventories was selected for evaluation. These are listed in Table 2.

Emissions inventories are required as inputs to comprehensive atmospheric modeling. Since such modeling (discussed in Section 4) is an important objective, the basic question addressed was the capability of each emissions inventory to serve as an input to such models. There are, of course, many *other* important uses for emissions inventories, and so, the assessments here (which focus on suitability as model inputs) should not necessarily be construed as assessments of the overall usefulness or value of a given inventory.

A set of criteria was developed to evaluate each of the emissions inventories, with particular emphasis on their usefulness to serve as model inputs. These criteria are presented in Table 3.

The overall coverage of different inventories for the BVES persistent toxic substances is summarized in Columns 5 and 6 of Table 7. The following issues and conclusions emerged from this analysis of emissions inventories.

For essentially all of the BVES persistent toxic substances, significant uncertainties in the emissions inventories appear to exist. *While many efforts are underway to improve the quality of emissions inventories, there do not appear to be accurate inventories* <u>*currently*</u> *available at present for most or all of the BVES substances.* The following overall conclusions emerged from this analysis[5]:

! for some compounds, no information about emissions appears to be available;

! for some pollutants, existing inventories have insufficient geographical and/or temporal resolution to be useful for modeling;

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[5]. It must be noted that these conclusions are based upon the information that could be obtained in the course of this research from the relevant government regulatory agencies (e.g., the U.S. EPA, Environment Canada, etc.). It is possible that additional information exists at these or other institutions but, for various reasons, was not identified in this analysis. If such additional information does exist, it is hoped that it will come to light in further discussion.

! for some pollutants, there appears to be no way to assess the accuracy of existing inventories, because of insufficient documentation of inventory details and/or of quality assurance/quality control procedures;

! for some compounds emitted by some sources in certain source regions, available inventory information appears to be confidential and unavailable to the public, independent researchers, or even government researchers who might want to explicitly calculate *and present* source-receptor relationships for specific emissions sources.

There are, of course, ongoing efforts to remedy many of these problems. Moreover, the above findings reflect only the information made available to us for this analysis. Additional information about existing inventories may come to light in further discussion.

The issue of inventory confidentiality emerged as a potential problem for several inventories. It appears that source-specific information in several inventories will be kept confidential, to protect the privacy of the emitters, and therefore will not be available to the public or independent researchers. Moreover source-receptor relationships will be unable to be presented even by those who had access to the data. Confidentiality may also have a deleterious effect on the *quality* of inventories. Emissions inventories are notoriously prone to inaccuracies, but errors are more readily detected if the data are widely available. Finally, the science and practice of atmospheric modeling of persistent toxic substances may be affected by confidentiality. Since emissions inventories constitute one of the fundamental inputs to atmospheric models, open scientific discussion about such models will be hindered to the extent that this input cannot be revealed.

Another issue that emerged is that for most compounds, there has been insufficient source testing to accurately estimate emissions. For many source classes for many of the BVES pollutants, uncertainties in emissions factors make even crude estimates of emissions difficult *even* if emissions sources could be accurately characterized with respect to their processes, throughputs, and pollution control equipment.

Table 2. Emissions Inventories Considered or Identified in this Analysis

[note: some details about coverage of BVES compounds in these inventories are included in Table 7; additional details are provided in Cohen (1997b)]

Emissions Inventory	Region	Notes
Great Lakes Regional Toxics Inventory Consortium of Groups, including the Great Lakes Commission, the U.S. EPA, others; Contact: Carol Ratza, Great Lakes Commission, Ann Arbor, Michigan	Great Lakes Region	An initial inventory, the **Southwest Lake Michigan Urban Air Toxics Emissions Inventory** has been prepared, including area sources in the Chicago, Milwaukee, and Gary Urban Areas. A broader, more comprehensive inventory is being developed and is expected to be available soon. Many details will be included in the inventory, although some facility-specific details will not be provided by participating agencies because of confidentiality policies.
U.S. EPA National Toxics Inventory, Version 2.0 (U.S. EPA, August 1996)	U.S.	This inventory contains geographically resolved emissions estimates for a range of compounds. For this analysis, only county-totals for different source classes were available; facility-specific details or documentation were not available, although it is possible that they may be available in the future.
National Mercury Report to Congress (U.S. EPA (1996b)	U.S.	National estimates and some geographically resolved information.
U.S. Clean Air Act, Section 112(c)(6) Draft Inventory (U.S. EPA, 1996a)	U.S.	National emissions estimates only.
Locating and Estimating Air Emissions from Sources of ___ ("L&E" Documents published by the U.S. EPA)	U.S.	Information on emissions factors given; in recent publications, national emissions estimates are given.
Toxics Release Inventory (TRI) U.S. EPA	U.S.	The 1993 version of this inventory was used as a base inventory for the U.S. EPA National Toxics Inventory.
Dioxin Inventory Project U.S. EPA, National Center for Environmental Assessment; contacts include: Duane Winters & David Cleverly, USEPA Washington D.C.	U.S.	This inventory is not yet available; it may be available sometime in 1997. It is expected to contain geographically resolved emissions estimates, including facility-specific information.

Table 2. Emissions Inventories Considered or Identified in this Analysis

[note: some details about coverage of BVES compounds in these inventories are included in Table 7; additional details are provided in Cohen (1997b)]

Emissions Inventory	Region	Notes
Environment Canada inventories for selected heavy metals & persistent organic pollutants (Environment Canada, 1996)	Canada	Inventories for cadmium and mercury contain substantial geographical resolution, but, much of the information is confidential. Similar spatially-resolved inventories for PAH's may also exist. Inventories for most other organic pollutants are currently available, generally, on a province-wide basis.
National Pollutant Release Inventory Environment Canada	Canada	Information from this inventory was used in the inventories prepared by Environment Canada (1996) for this analysis.
Strategic Options Process (SOP) for the Reduction of Toxics Environment Canada	Canada	Information from this inventory was used in the inventories prepared by Environment Canada (1996) for this analysis.
Canadian Dioxin Emissions Inventory (Federal/Provincial Task Force on PCDD/F's) Co-Chair: Raouf Morcos, Environment Canada	Canada	This inventory is currently being developed and is expected to be available sometime in 1998. Preliminary information from this inventory effort was used in the inventories prepared by Environment Canada (1996) for this analysis.
ORTECH Biocide Emissions Inventories (Scholtz, 1997)	Canada and the U.S. (see note at right)	These inventories, prepared for Environment Canada, may be available in some form soon. For some compounds, *global* emissions estimates are being attempted.
ORTECH Inventory for Ontario and Eastern North America (Johnson et al., 1992)	Ontario and Eastern North America	Overall emissions estimates for a range of pollutants are included. The details of the estimates, included in Appendices to this report, are confidential and are not available.
Inventories being developed by the Ontario Ministry of Environment and Energy (OMOEE) Contact: Peter Wong	Ontario (only?)	This inventory is under development and is not yet available.
Canada Ontario Agreement (COA) — Sources, Releases, and Loadings for COA Substances (Putnam, 1995; Brown and Thornton, 1996)	Ontario	As with essentially all the inventories considered, this inventory is undergoing continual development. It contains different levels of geographical resolution for different compounds; some have facility-specific resolution; some compounds have only province-total estimates.
Commission for Environmental Cooperation Inventories including the North American Pollutant Release Inventory and other projects (NAFTA Commission for Environmental Cooperation)	Canada, U.S. and Mexico	Inventories for one or more compounds are being developed. These inventories are not available at this time. Emissions inventory information may be available in the future.

Table 3. Criteria For Evaluating Air Emissions Inventories In Relation to Their Potential Use as Inputs to Atmospheric Fate and Transport Models

1	**Inventory should be comprehensive, i.e., all potential source classes should be included.** ● Omissions in the inventory should be acknowledged
2	**Inventory should be complete, in its treatment of each source class.** ● All emissions sources for a given source class should be included; omissions should be acknowledged. ● What is level of confidence? e.g., there may be more confidence in an inventory based on frequent, thorough, inspections by trained regulators, etc.
3	**Geographical resolution should be as high as possible.**
4	**Temporal resolution should be as high as possible.**
5	**Estimates should be based on actual measurements to the largest extent possible.** ● Statistical summaries and ranges of the measurements should be provided
6	**Documentation of the use of emissions factors should be provided, and if they are used, adequate information should be provided about them.** ● Inventory should state when emissions factors are used ● Basis of emissions factor should be clearly described ● Uncertainties and ranges should be given ● Match between source and emissions factor basis should be justified
7	**Details on key characteristics of each source should be provided.** ● Throughput; Air Pollution Control Equipment ● Process information especially relevant to emissions
8	**Details about the emissions should be provided.** ● Form of pollutant in emission (vapor, particle, particle size) ● Temperature, height, velocity of the emissions
9	**Data should be available in different useful forms.** ● Both summary-level and facility-level data are useful
10	**Inventory should be publicly available (including facility-specific details).** ● Public and independent researchers have a right to know ● Quality control/assurance: the more people that critically evaluate an inventory, the more likely that errors will be detected

4. Modeling the Atmospheric Transport and Deposition of Persistent Toxic Substances to the Great Lakes

In atmospheric modeling of pollutants, an attempt is made to estimate the transport and fate of pollutants which are emitted to the air. An analysis of past, ongoing, and expected government-initiated modeling efforts for BVES compounds was conducted (Cohen 1997c) and will be summarized here.

The following might describe an ideal modeling analysis:

- the emissions of all sources of a given pollutant are well characterized (geographical resolution; temporal resolution; etc.);

- the atmospheric fate and transport of these emissions is accurately modeled, including accurate characterizations of meteorological processes, micro-chemical (e.g., reaction with hydroxyl radical) and micro-physical processes (e.g., vapor/particle partitioning), and deposition processes (and possible re-emission processes, e.g., the grasshopper effect).

- the total concentrations and deposition to a given receptor are predicted (and compare well with measured values), and, the contributions to the total loading at a given receptor from individual sources and/or source regions are quantified.

There have been many different approaches to modeling the atmospheric fate and transport of air pollutants. Most can be classified as utilizing either a Lagrangian or Eulerian framework.

In a Lagrangian model, pollutant fate and transport is mathematically characterized relative to the trajectory of air parcels. The coordinate system for pollutant dispersion moves along with a given air parcel. Lagrangian models are typically used in attempts to estimate the impact of a given source on downwind receptors. When multiple emissions sources are involved, calculations can be made for one source at a time, or, for all sources together. An advantage of many Lagrangian models is that they can require less computational resources than Eulerian approaches, all things being equal. There are difficulties in using Lagrangian models for simulations in which complex atmospheric chemistry must be considered and in which the interaction of emissions from different sources is important. In Lagrangian-based models, it is possible to consider emissions from multiple sources at the same time. When this is done, however, the process of keeping track of source-receptor relationships becomes numerically difficult.

In the Eulerian approach, pollutant fate and transport is estimated everywhere in the modeling domain relative to a fixed coordinate system. Pollutants from all sources being considered are mixed together into grid cells which cover the modeling domain. During each time step in an Eulerian model, the movement and dispersion of the pollutant is modeled by estimating the behavior of the pollutant *within* a given grid cell and estimating the movement of pollution *from each cell to adjoining cells*. In contrast to Lagrangian models, this type of framework is ideal for situations where the interaction of emissions from different sources is important. Perhaps the classical example of this situation is the problem of estimating concentrations of tropospheric ozone. In this situation, emitted volatile organic compounds (VOC's) and nitrogen oxides emitted from many disperse sources are mixed together in the atmosphere and participate in a complex set of chemical and photochemical reactions. Ozone is formed during these reactions. To predict the concentration of ozone in such a situation, it is necessary to simulate behavior of the pollutants from all significant sources *at the same time*.

The mathematical formulations of deposition processes used in Lagrangian and Eulerian models are often very similar, and in some cases are essentially identical.

Some general statements can be made about modeling the atmospheric behavior of most or all of the BVES compounds. First, ambient atmospheric concentrations of most of the BVES pollutants are typically at most on the order of one part-per-million and in many cases are much smaller.[6] These concentrations are so dilute that, in general, the interactions of BVES pollutants *emitted from different sources* will not be significant. Thus, for most or all of the BVES compounds, it may be possible to model their fate and transport with Lagrangian as well as Eulerian approaches.

There is an important caveat to the above statement, however. It regards the problem of simulating the interaction of pollutants at the earth's surface, especially in regard to vapor-phase dry deposition and re-emission processes. In these processes, the transfer of pollutant between the atmosphere and the surface is governed by the degree to which the instantaneous local situation departs from thermodynamic equilibrium. The degree of departure from equilibrium is dependent on the *total* concentration of the pollutant in the local atmosphere *and* the *total* concentration of pollutant in (and/or on) the particular materials at the earth's surface at the location of interest. The net direction of transfer will depend on the relative concentrations in the two phases. The rate of this net transfer will depend on the degree of thermodynamic imbalance and the motion, diffusion and mixing processes in *each* of the two phases (e.g., the rate of turbulent mixing and diffusion of pollutant in the each phase, etc). For example, the instantaneous rate of deposition of vapor-phase pollutant to a Lake at a given time and location depends not only on the concentration of the pollutant in the gas phase of the atmosphere immediately above the water surface; it also depends on the concentration of the same pollutant in the water near the surface.

The implication of the above considerations for modeling the fate and transport of persistent toxic pollutants is the following. *If* the compound's re-emission from the earth's surface after being deposited is an important process (e.g., re-emission from a

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[6]. Because of their extreme toxicity, persistence and bioaccumulative properties, many of the compounds considered can exert significant toxicological and environmental effects even at very low concentrations.

water body or a terrestrial surface), then, it is likely that an estimate of the net direction of the transfer and the rate of the transfer between the atmosphere and the earth's surface will depend on the *total* instantaneous concentrations of the pollutant in the atmosphere above the surface and at surface itself. This means that past deposition and re-emission and the impacts of *other* sources may affect the fate and transport behavior impact of any given source. In a sense, this complicated situation is similar to the situation described above in modeling the problem of tropospheric ozone. In both situations, an accurate simulation only appears to be possible when all sources are considered together. Finally, these re-emissions situations, it may be necessary to couple the atmospheric model with a model of the surface media (e.g., surface water, soil, etc.). The problem of incorporating the above "grasshopper effect" into air pollution models remains a significant challenge. This topic is discussed in a series of articles by Wania and Mackay (1993ab, 1995, 1996).

An overall summary of the general components of many modeling systems that attempt to simulate the atmospheric fate and transport of pollutants is given in Table 4.

The objective of this analysis was to consider models that have, will, or could attempt to estimate the atmospheric transport and fate of persistent toxic substances (i.e., the BVES compounds listed in Table 1) emitted to the air, with particular emphasis on deposition to the Great Lakes basin.[7] The analysis was limited primarily to modeling efforts undertaken by government agencies in the U.S. and Canada, as identified through: (a) discussions with members of the International Air Quality Advisory Board (which includes representatives from the USEPA, NOAA, Environment Canada, Ontario Ministry of the Environment, and other agencies and institutions); (b) discussions with government researchers engaged in modeling work; and (c) review of the literature.[8] A list (and summary) of the modeling analyses that were considered is given in Table 5.

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[7]. Modeling efforts that have analyzed acid deposition, formation of tropospheric ozone, global warming, and stratospheric ozone depletion were not considered, unless the model used has *also* been applied to one or more of the compounds considered in this analysis, or, if such application is anticipated.

[8]. For the most part, non-governmental modeling efforts have not been included, and this is recognized as a limitation of the analysis. An attempt was made, however, through review of the literature, to include particularly relevant modeling analyses that have attempted to link air emissions sources to atmospheric deposition to the Great Lakes for one of the BVES persistent toxic substances. A few such efforts that could be found are included.

Table 4. General Components of Many Modeling Systems for Simulation of the Atmospheric Fate and Transport of Persistent Toxic Substances

Emissions Inventory

- geographical and temporal resolution of emissions
- speciation of pollutant emitted
- physical form of pollutant when emitted (vapor and/or particle?; particle size distribution?; physical and chemical characteristics of emitted particles?)
- emissions conditions (height, temperature, velocity, etc.)
- if re-emissions of previously deposited material are important, then they must be considered

Characterization of the Dynamic Meteorological Environment

- details of three-dimensional wind movement at many different scales can be important (large scale motions, mesoscale dynamics, short-range phenomena, small-scale turbulent mixing)
- characterization of the dispersion and diffusion of the emitted pollutant in the atmosphere over all relevant length scales

Characterization of the Spatially & Temporally Varying Micro-Physical and Micro-Chemical Environment

- temperature; humidity
- clouds; precipitation
- physical & chemical nature of the atmospheric aerosol
- concentration of hydroxyl radical & other reactants
- intensity & spectral distribution of electromagnetic radiation
- detailed characteristics of the earth's surface

Characterization of the Interaction of the Pollutant Being Modeled with the Micro-Physical and Micro-Chemical Environment

- vapor/particle partitioning
- partitioning to cloud water; precipitation
- reaction rates with hydroxyl radical and other reactants in vapor phase, particle phase
- rates of droplet phase reactions
- rate of photolytic transformation
- deposition and re-emission phenomenon

Model Output

- predicted atmospheric concentration (from all sources) at a given locations at given times
- predicted deposition (from all sources) to a given receptor at a given time at a given location
- source-receptor relationships, e.g., the amount of deposition at a given receptor at a given time attributable to the contribution of a given source's emissions

Tests of Model Validity

- comparison of predicted concentrations and deposition with measured values

Table 5. Summary of Modeling Analyses Considered

#	Model	Reference(s) and/or principal contact(s)	Compounds Considered	Source Regions	Receptor Regions	Type of Results Available *(or Expected to be Available)*
Eulerian Approaches						
1	Global Chemical Transport Model	Pudykiewicz and Koziol, 1997; Pudykiewicz and Dastoor, 1996	α-HCH, γ-HCH	global oceans; future work will include terrestrial emissions	global, including, but not limited to the Arctic	predicted atmospheric concentrations now available; *deposition results available or soon to be available*
2	REMSAD	Axelrad, 1997; Guthrie et al., 1995; SAI, 1996	PCDD/F, Mercury, Cadmium, Polcyclic Organic Matter, Atrazine	U.S. (planned); possibly others	Great Lakes, Chesapeake Bay, possibly other receptors	*results for concentrations and deposition expected to be available in 1-2 years*
3	ADOM	Bloxam et al., 1991; Bloxam, 1997; Petersen et al., 1996	Mercury	Eastern U.S. and Canada	Eastern U.S. and Canada	concentrations and deposition; *additional results expected*
4	RAMS (Meteorological)	McQueen et al., 1997a, 1997b; Valigura et al., 1996	Nitric Acid	(not modeled)	Chesapeake Bay	detailed estimates for dry deposition
5	RADM	Dennis, 1997	Nitrogen compounds	Eastern U.S. and Canada	Chesapeake Bay and Watershed	concentrations; deposition; source-receptor relationships
6	MODELS-3	Novak et al., 1995 Dennis et al., 1996 Bullock, 1997a	Atrazine (+ others)	U.S. and Canada	Lake Michigan (+ others)	*concentrations; deposition; source receptor relationships (?)*

Table 5. Summary of Modeling Analyses Considered

#	**Model**	**Reference(s) and/or principal contact(s)**	**Compounds Considered**	**Source Regions**	**Receptor Regions**	**Type of Results Available *(or Expected to be Available)***
Lagrangian Approaches						
7	RELMAP	Clark, 1992ab	Cadmium (a)	U.S.	Lake Michigan	Deposition to Lake Michigan
8		Bullock et al., 1997b; U.S. EPA 1996c	Mercury	U.S.	U.S.	Atm. Conc. & Deposition in U.S.
9		Bullock 1997ac; Cleverly 1997	PCDD/F	U.S.	U.S.	Atm. Conc. & Deposition in U.S.
10	ASTRAP	Voldner and Schroeder, 1989	Toxaphene	U.S.	Great Lakes	Total predicted deposition; a few source-receptor relationships presented
11		Shannon and Voldner, 1995	Mercury	Eastern U.S. and Canada	Great Lakes	Total predicted deposition
12		Voldner et al., 1993	Lead	U.S. and Canada	Great Lakes	Total predicted deposition
13	HYSPLIT	Cohen et al., 1995	PCDD/F and HCB	U.S. and Canada	Great Lakes	Atmospheric concentrations and predicted deposition; source receptor relationships
14		CBNS, Queens College, ongoing work	PCDD/F	U.S. and Canada	Dairy Farms in Wisconsin and Vermont	*Atmospheric concentrations and predicted deposition; source receptor relationships*
15		CBNS, Queens College, ongoing work	Endocrine-Disrupting Biocides	U.S. and Canada	Drinking water reservoirs in the midwest and northeast	

Table 5. Summary of Modeling Analyses Considered

#	Model	Reference(s) and/or principal contact(s)	Compounds Considered	Source Regions	Receptor Regions	Type of Results Available *(or Expected to be Available)*
Receptor-Oriented Approaches						
16	Back-Trajectory Approach	Valigura et al., 1996	NO2	U.S. and Canada	Chesapeake Bay	Information about source regions contributing to observed concentrations at the receptor location
17		Hoyer et al., 1995	Mercury	U.S. and Canada	Michigan	
18		Blanchard, Hopper, and Hoff, 1997	Cadmium (a)	U.S. and Canada	northern shore of Lake Ontario	
19		Gao, et al., 1996	Cadmium (a)	U.S. and Canada	Dorset Ontario	
20		Gatz and Prospero, 1996	dust	U.S. and Canada	Illinois	
21		Hoff et al., 1992ab	PCB's and organochlorine biocides	U.S. and Canada	Egbert Ontario	
22	Hybrid-Receptor Modeling Approach	Keeler, 1994; Pirrone et al., 1995b	Semivolatile Organic Compounds	Great Lakes region	Lake Michigan	deposition arising from hypothetical "virtual" sources that could explain observed atmospheric concentrations at particular locations.
23		Keeler, 1994; Pirrone et al., 1995a	Trace Elements		Lake Michigan	
24		Keeler and Pirrone, 1996	Cadmium (a)		Lake Erie	
25		Pirrone and Keeler, 1996a	Cadmium, Mercury		Rouge River Watershed	

Notes: (a) Other heavy metals also considered; the only BVES compound included, however, was cadmium

The following observations and conclusions emerged from this analysis.

A. Available Results

The ultimate outputs of most models are the total predicted atmospheric concentration (from all sources) and deposition at given locations at given times arising from emissions input into the model. Several comprehensive modeling analyses were found which present estimates for one or more of the BVES pollutants of the *total* deposition to the Great Lakes arising from emissions from sources in the U.S. and Canada.

Most models are capable of determining source-receptor relationships, e.g., the amount of deposition at a given receptor at a given time attributable to the contribution of a given source's emissions. Lagrangian-based models are particularly suited for generating such results. In some cases, however, Eulerian models can be configured to develop such estimates (e.g., by treating emissions of a given chemical from a given source as a different "species" than emissions of the same chemical from a different source); at the present, however, this is not generally done. There is one example discussed in this report involving the use of an Eulerian model (RADM) to determine source-receptor relationships for nitrogen deposition to the Chesapeake Bay (Dennis, 1997).

With one exception[9], no currently available comprehensive source-receptor results for BVES pollutants could be identified. As with the emissions inventory programs described above, modeling efforts are an active area of research and development. All of the ongoing efforts identified are expected to provide critically useful information in the future.

Some of the reasons for the paucity of available results for source-receptor appear to include the following:

! keeping track of source-receptor relationships can require substantial computational resources; thus, many modeling exercises do not attempt to do so;

! in some cases, there may be a reluctance to specify source-receptor relationships because of actual or perceived uncertainties in the emissions inventory;

! in some cases, there may be a hesitance to specify source-receptor relationships as a way of avoiding an emphasis on the environmental effect of a particular source.

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[9]. A comprehensive but preliminary source-receptor analysis for PCDD/F transport and deposition to the Great Lakes has been carried out (Cohen et al., 1995).

B. Application of Models to BVES Pollutants

With varying degrees of accuracy, each of the models considered in this analysis is or could be capable of simulating the atmospheric fate and transport of most or all of the BVES persistent toxic substances. However, for most of these pollutants, there are significant uncertainties in basic physical-chemical properties, chemical reaction rates (and concentrations of reactants), rates of atmospheric photolysis, vapor/particle partitioning phenomena, and/or wet and dry deposition processes.

These uncertainties will limit the accuracy with which any of the models considered here (or any other model) can predict atmospheric fate and transport of the BVES persistent toxic substances.

Research to develop greater understanding in these uncertain areas should be continued and supported at the highest possible level. In these efforts, laboratory and field measurements should play a very important role.

Accurate air pollution models are only really possible to create after these basic underlying phenomena are well understood. Models can be thought of as the scientific culmination of the process of attempting to understand environmental processes. For a given phenomenon, the course of scientific knowledge might progress through the following steps: (1) measurements are made; (2) theories are developed to explain the measurements; (3) the theories are tested for their predictive ability. Only after a large body of experimental and theoretical scientific work has been performed which leads to a relatively complete understanding of a particular phenomenon can such a process be included in any meaningfully accurate way in an air pollution model. In this sense, models can be seen as an overall test of the scientific understanding of the environment and the behavior of a given pollutant within it.

Thus, given an accurate emissions inventory, *if* the scientific understanding of atmospheric dynamics, pollutant dispersion, physical/chemical transformations and deposition processes were known exactly, it would be possible to accurately predict concentrations and deposition at any desired receptor. We may perhaps be closest to this ideal with the fate and transport of atmospheric sulfur and nitrogen pollutants. However, for most or all of the BVES persistent toxic substances, the scientific community is in the early stages of attempting to construct comprehensive models of atmospheric fate. Future modeling efforts will hopefully be able to incorporate additional scientific findings regarding fundamental atmospheric phenomena.

C. Availability of Emissions Inventories

As discussed above in Section 3, for most or all BVES substances, transparent, complete, geographically and temporally resolved inventories are not currently available.

Thus, many feel that it may be premature to attempt to model most or all of the

BVES persistent toxic substances at this time because of the lack of accurate emissions inventories. If an analysis is based on an inaccurate inventory, successful validation against real-world measurements would be unlikely. Thus, the question of whether an emissions inventory is accurate or not is a very serious one, in regards to its potential use as an input to an air pollution model, as the answer may well determine the success or failure a given effort. It is often very difficult to determine the causes of model failure, and in such situations, it is crucial to have a sense of the level of confidence one should have in the accuracy of the inventory. In this regard, information about quality assurance and quality control approaches taken to assess the validity of the inventory would be extremely helpful.

D. Modeling of Meteorological Processes

Relative to the uncertainties discussed above, many model architects consider the meteorological aspects of the simulation as the most well understood. This is not to say that improvements are not needed in this area. For example, the characterization of clouds and precipitation is believed to need improvement (e.g., McQueen *et al.*, 1996).

Different models treat meteorological and dispersion phenomenon with different approaches; there are differences in horizontal and vertical resolution (grid size) and in the numerical and physical sophistication of the simulations. In the future, it may be possible to compare the predictions of different models (as discussed below). These comparisons might yield information on the relative accuracy of different approaches. In particular, it would be useful to gain insights into the tradeoffs in accuracy among models of varying computational intensity.

E. Model Evaluation

The physical and chemical situation which atmospheric fate and transport models are attempting to simulate is extremely complex, and the scientific understanding of many important processes is somewhat limited. Thus, a range of assumptions and simplifications are made. Comparison of model results against real-world measurements help assess the validity of these approximations. In these overall model evaluation tests, predicted concentrations and deposition are compared against real-world measurements at specific locations at specific times. Typically, there are often very little data against which to compare model predictions. Yet, these tests of model validity must be made.

In these comparisons, a fundamental question sometimes arises: *If model predictions do not match measurements, is the discrepancy the result of inaccuracies in the simulation or in the emissions inventory?* With many uncertainties in different model components, it can be difficult or impossible to determine the causes for poor model performance.

For many of the BVES compounds, *a particularly valuable data set for model validation is that which has been and will be provided by the IADN ambient monitoring* *program.* Data from this and other monitoring programs will be invaluable to the continuing evolution of the emerging science of comprehensive atmospheric modeling of persistent toxic substances. In general, it is important to recognize that while monitoring programs are very useful even by themselves, their value is enhanced when used to test the validity of modeling analyses. To this end, it is useful to consider how existing monitoring programs can be used to facilitate model validation and how new programs might be designed.

Finally, as with any numerical simulation, there are tradeoffs between the resolution and sophistication of the calculation and the computational resources required to carry out the analysis. Some of the models described here can only practically be run in mainframe super-computer environments; some can be run on a personal computer. A useful exercise that might be carried out in the future for some or all of the BVES pollutants would be side-by-side model analyses, using the same emissions inventories and validation data.[10]

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[10]. This type of inter-comparison analysis has been carried out in the past; example of model inter-comparison studies include those for: (a) global distribution of radionuclides emitted from the Chernobyl accident (Klug *et al.*, 1992); (b) sulfur transport and deposition in the U.S. and Canada (Clark *et al.*,1987,1989); (c) transport of atmospheric tracers in the U.S. (Carhart *et al.*, 1989); and (d) transport of tetroons (tracer balloons) in the U.S. (Clarke *et al.*, 1983).

5. Ambient Monitoring of Persistent Toxic Substances in the Great Lakes Region, and the Use of These Data to Estimate the Atmospheric Loading to the Great Lakes

An analysis has been conducted which summarizes ambient monitoring in the Great Lakes region and the use of these data to estimate the net atmospheric loading to the Great Lakes (Cohen and Cooney, 1997). A summary of this analysis is presented here.

A. Methodology Used to Estimate Loadings

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 the *wet deposition pathway* is estimated from the precipitation rate and the concentration of a given 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 a given pollutant in the air above the lake, *and*, as discussed below, for the fraction of a given pollutant that exists in the *vapor phase* (as opposed to the fraction that exists associated with *particles* in the atmosphere), the loading estimate also depends on the concentration of the 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. 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 basic atmospheric pathway loading equation, commonly employed in these estimates, is presented in the box on the next page.

In Table 6, the parameters in this equation, which are necessary to estimate the atmospheric loading to the Great Lakes (or any lake, for that matter) are presented, along with information about how the parameters are typically obtained.

Except for the gas constant (R), and the area of the Lake (A)[11], 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:

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[11]. 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.

Conventional Approach Used in Estimating Loading to a Given Lake From the Atmospheric Pathway Based on Ambient Measurements

L_{total} = $A\ R_p\ C_p$
(wet deposition)

\+ $A\ \ddot{o}_a\ v_d\ C_a$
(dry deposition of particle-phase material)

\+ $A\ K_{OL}\ \{[\ (1-\ddot{o}_a)\ (RT/H)\ C_a\ ]\ -\ [\ (1-\ddot{o}_w)\ C_w\ ]\}$
(net deposition of vapor-phase material)

\+ [fog deposition]

\- [pollutant losses from the lake due to droplet resuspension]

\+ [indirect atmospheric loadings]

The symbols in the above equation are defined as follows
(with one possible set of consistent units):

A = area of lake (m^2)

C_p = concentration of pollutant in precipitation (g/m^3)

C_a = total concentration of pollutant in atmosphere (g/m^3)

C_w = total concentration of the pollutant in the Lake near the surface *(g/m3)*

H = Henry's Law coefficient for the pollutant $(Pa\text{-}m^3/mole)$

K_{OL} = mass transfer coefficient for air-water gas exchange (m/yr)
(Note: 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.)

L_{total} = total loading to a given Lake due to the atmospheric pathway *(g/year)*

R = gas constant $(Pa\text{-}m^3\ /\ mol\ \text{-}^oK)$

R_p = precipitation rate *(m/year)*

v_d = dry deposition velocity *(m/year)* (generally an assumed value)

T = temperature (^oK)

$\ddot{o}_a$ = fraction of pollutant in air associated with atmospheric particles *(dimensionless)*

$\ddot{o}_w$ = fraction of pollutant in water attached to suspended sediment *(dimensionless)*

Table 6. Parameters Typically Used to Estimate the Net Atmospheric Deposition to a Given Lake or Lake Area

(note: all the parameters below will vary in time and space; thus, averages are typically used)

	Parameter	How Obtained (in typical situation)
Wet Deposition of Gas and Particle Phase Pollutant	Concentration of the Pollutant in Precipitation	Measured
	Precipitation Rate	Measured
Dry Deposition of Particle-Phase Pollutant	Concentration of the Pollutant in the Air Near the Lake Surface	Measured
	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 Deposition Velocity of Particle-Associated Pollutant	Typically estimated; often a constant value is assumed
Net Dry Deposition Flux of Vapor Phase Pollutant	Concentration of the Pollutant in the Air Near the Lake Surface	Measured
	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.
	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
	Henry's Law Constant	Based on existing laboratory measurements; temperature dependent
	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.

(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 $\ddot{o}_a$ and $\ddot{o}_w$, 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);

Obviously, ambient measurements cannot 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 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.

B. Very Uncertain Terms in the Loading Equation

Three of the terms in the above “loading equation” are expressed qualitatively only:

! fog deposition
! pollutant losses from the lake due to droplet resuspension
! indirect atmospheric loadings

At this time, conceptual “terms” are included in the loading equation to indicate that is known that these factors will influence the magnitude of the net loading, but, that it is not generally possible at this time to make quantitative estimates. These phenomena will be briefly discussed below.

i. Pollutant Losses from the Lake Due to Droplet Resuspension

While it is somewhat poorly understood at the present, water droplets can be “ejected” from the lake, especially as a result of the breaking of waves. While some of these droplets would be 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 manner.

ii. 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. Moreover, fog deposition is not generally measured in precipitation samplers.

iii. Indirect Atmospheric Loadings

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 make even semi-quantitative estimates at this time.

C. The Capability of Monitoring Programs to Estimate Loading

It must be noted that the analysis (Cohen and Cooney, 1997) was somewhat limited, as the following information was not available for consideration at the time this analysis was conducted:

! Data sets from the Integrated Atmospheric Deposition Network and from other monitoring programs in the Great Lakes region;

! Much of the data and publications developed in the AEOLOS (Atmospheric Exchange Over Lakes and Oceans) research program;

! Essentially all of the data collected and being analyzed for the Lake Michigan Mass Balance Study;

- Atmospheric Deposition to the Great Lakes and Coastal Waters, a book published by the Society of Environmental Toxicology and Chemistry.

The following observations and conclusions emerged from the review of monitoring programs that was carried out.

First, the inclusion (or lack of inclusion) of the BVES compounds in air and water monitoring programs is summarized in Columns 10-16 in Table 7. It can be seen that many of the BVES compounds are being monitored, but some are not. The following BVES compounds are apparently not being monitored systematically in the Great Lakes region:[12]

- alkylated lead
- 4-bromophenyl phenyl ether
- 3,3-dichlorobenzidene
- 4,4'-methylene bis (2-chloroaniline)
- tetrachlorobenzenes and pentachlorobenzene[13]

Thus, obviously, current air monitoring programs do not appear to provide data for estimating the overall loading to the Great Lakes for the above BVES compounds.

PCDD/F's are not included in the IADN program, but, some ambient air measurements are being made in the Great Lakes region. These measurement programs have not been designed with Great Lakes atmospheric loading estimates in mind, however.[14]

Monitoring programs for BVES compounds in the water of the Great Lakes, necessary to make accurate estimates of the net atmospheric loadings (or volatilization) of vapor-phase substances, are more limited.

Except for PCB's and DDT, there have apparently been no water measurements in Lake Huron in the last five years (Column 14 of Table 7).

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[12]. Comprehensive Information about air monitoring programs *other* than the Integrated Atmospheric Deposition Network could not be obtained in time to be included in this draft. These additional programs include those sponsored by state/provincial agencies in Wisconsin, Michigan, Vermont, Ontario, and Quebec.

[13]. Monitoring of these compounds in precipitation is being done in conjunction with the Great Lakes Precipitation Network, carried out by the Canadian Centre for Inland Waters, Environment Canada. Ambient air measurements for these compounds, however, do not appear to be included in any of the monitoring programs identified.

[14]. Most of the current sampling locations for PCDD/F's in the Great Lakes region are in urban areas; the representativeness of these urban samples for lake-wide deposition estimates is, of course, an issue.

For several of the BVES compounds in the other Lakes, there have generally been one or two years in which measurements have been made in the past five years. The compounds included in various water monitoring programs in these other Lakes are summarized in Table 7 (Columns 12, 13, 15, 16). The fact that water measurements are not being made each year means that vapor-phase dry deposition loading estimates must sometimes be made with water concentration data that is one or more years old.

There appear to have been no measurements in any of the Great Lakes in the past five years for the following BVES compounds:

- alkylated lead
- cadmium or cadmium compounds
- mercury (except for measurements in Lake Michigan)
- tributyltin compounds
- pentachlorophenol
- 4-bromophenyl phenyl ether
- 3,3-dichlorobenzidene
- 4,4'-methylene bis (2-chloroaniline)
- dinitropyrenes

Except for cadmium, all of the above compounds exist to a certain extent in the vapor phase in the atmosphere, and so, the lack of water concentration measurements make estimates of overall loading difficult.[15] Cadmium is being measured in the air, and so, estimates of its overall loading to the Great Lakes can be attempted.[16]

For the BVES compounds that are being measured in the air and/or water phases, the issue of spatial representativeness remains a key concern. The relative importance of urban plumes has been the subject of several recent investigations, including the AEOLOS project and the Lake Michigan Mass Balance. Results from these studies are being presented elsewhere by others. It will be briefly noted here (based on Sweet, 1996) that for *some* pollutants, e.g., PAH's, it appears that there are substantially higher over-lake concentrations and deposition downwind of major urban centers (e.g., Chicago). For these pollutants, the concentrations measured at the IADN monitoring stations — which, for all of the Great Lakes except Lake Ontario, are in fairly rural locations — may not be representative of "average" over-lake conditions. For many of the BVES pollutants, however, there do not appear to be dramatic variations in

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[15] . As noted above, there are apparently no ambient air measurements being made in the Great Lakes region for some of the compounds in the above list anyway.

[16] . Cadmium is believed to exist predominantly in the particulate phase in the atmosphere. Therefore, loading estimates based on precipitation and particle-phase deposition alone are appropriate. Consideration of gas-exchange with the lake's surface — for which water column measurements would be required — is not necessary.

air concentrations throughout the Great Lakes region, and so, the IADN measurements would seem to be providing adequate data. The results of the AEOLOS project and the Lake Michigan Mass Balance Study are eagerly anticipated as they are expected to provide substantial insight into this question.

It should be noted that comprehensive modeling of BVES compounds might be of great utility in providing additional insight into the representativeness of ambient monitoring data. Tests of the validity of the modeling could be made by comparing the predicted concentrations at the IADN sites against the actual measurements at those sampling locations. If the modeling methodology appeared valid, it could be used to estimate the spatial and temporal variability of concentrations throughout the region. These results would provide crucial information regarding the representativeness of existing measurements.

The accuracy of loading estimates to the lakes — even if ambient air and water measurements are being made *and* they were representative — is somewhat limited by uncertainties in the terms of the loading equation.

As mentioned above, three of the terms — fog deposition, droplet resuspension, and indirect atmospheric loading resulting from atmospheric deposition to the watershed — are not currently understood sufficiently to include them quantitatively in the loading estimates.

Uncertainties exist in the other terms as well. One uncertainty, for example, concerns the estimate of dry deposition velocity of particulate-phase pollutant. This will depend, of course, on many factors, including the particle size distribution of the pollutant being considered. Particle-size distributions are not generally measured in existing programs -- much less *pollutant-specific* particle size distributions. Thus, the specification of the particulate dry-deposition term in the loading equation is somewhat uncertain.

Finally, it would be useful if ambient monitoring data in the Great Lakes region could be made more readily available to all.

6. Summary

In Table 7, below, an overall summary of information about the atmospheric transport and deposition of the BVES persistent toxics substances to the Great Lakes is presented.

Overall, the key points that have emerged from this analysis include the following:

- Many of the BVES compounds appear to be capable of long range atmospheric transport.

- *Currently*, for most BVES compounds, there are either (a) no emissions inventory information available in the U.S. or Canada, or, (b) the information that is available is incomplete and/or presented with minimal geographical resolution and/or of uncertain reliability. Thus, for most of the BVES compounds, there do not appear to be currently available inventories that could be used as a reliable inputs to comprehensive atmospheric fate and transport models.

- Moreover, it appears, at least currently, that the limited availability of emissions testing data for many source classes of BVES pollutants represents a major impediment to developing accurate inventories.

- Substantial work is underway to improve and develop emissions inventories for many (but not all) of the BVES compounds. However, the degree to which the new inventories will be "transparent" (i.e., with full details made available) is unclear, as many inventories appear to be governed by policies that require facility-specific information to be kept confidential.

- The use of atmospheric models to trace the movement of persistent toxic substances from emissions sources to receptors is essential to understand the problem of pollution of the Great Lakes through the atmospheric pathway. The elucidation of source-receptor relationships is a necessary pre-requisite to developing and implementing approaches to reducing or virtually eliminating such pollutant input to the Great Lakes.

- There have been a number of worthwhile modeling analyses conducted to date, and there are a number of promising modeling efforts currently ongoing. Nevertheless, the current state of knowledge and ability of such efforts is somewhat limited due to the following factors:

  - uncertainties in emissions inventories (as noted above)

  - uncertainties in pollutant-specific atmospheric fate processes;

  - for some of the BVES pollutants, there are very few or no ambient measurements against which to test the overall validity of models.

  - the grasshopper effect may be important for many of the listed compounds, and its inclusion into atmospheric fate and transport models remains a significant challenge.

- Most modeling analyses done to date and most of those underway (that could be identified in this analysis) appear to be focused on estimating the *total* impact from all sources combined, and do not appear to be focused on elucidating specific source-receptor relationships.

- 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 evaluation data for comprehensive atmospheric fate and transport models.

- It would be helpful if a system could be established for making Great Lakes regional monitoring data readily available to the public and to the research community.

- For some of the BVES compounds, there appear to be no systematic, ambient monitoring programs in the air and/or water of the Great Lakes region, and so assessment of the significance of the atmospheric deposition pathway is difficult.

Table 7. Summary of Information About the Atmospheric Transport and Deposition of Persistent Toxic Substances to the Great Lakes

column#'s (see notes for descriptions)→	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
		V/P	LRT	Geographically Resolved Emissions Inventories		Existing Modeling Results or Planned Analyses			Air & Precipitation Monitoring in G.L. Region		Water Monitoring in the Great Lakes: Any Samples 1992-1996?					
Compound or Compound Group	Level	Vapor-Particle Partitioning	Long-Range Air	Inventory Which May Exist or May Soon Exist	Inventory Suitable for	receptor-oriented	comprehensive	source-	Integrated Atmos. Dep. Network	Other Programs	Superior	Michigan	Huron	Erie	Ontario	Loading Estimate Possible?
MERCURY and MERCURY COMPOUNDS																
Elemental Mercury (Hg^o)	I	v	1	-	-	-	2,3,8,11	-	-	-	-	-	-	-	-	0
Divalent Mercury (e.g., $HgCl_2$)	I	v	2	-	-	-	2,3,8,11	-	-	-	-	-	-	-	-	0
Monomethyl Mercury	I	v	3	-	-	-	-	-	-	-	-	-	-	-	-	0
Total Gaseous Mercury	I	V V	1-3	-	-	-	-	-	J	CFUL	-	-	-	-	-	~ 0 (F)
Particulate Mercury	I	PP	2	-	-	-	2,3,8,11	-	J	TFUL	-	-	-	-	-	1
Total Mercury	I	v	1-2	NCLAGE	NE	17,23, 25	2,3,8,11	-	J	DTFUL	-	m	-	-	-	~ 0 (F)
OTHER METALS / ORGANOMETALLICS																
Alkylated Lead	I	(V V)	2	NCLAG	-	-	-	-	total Pb	-	-	-	-	-	-	0
Cadmium & Cadmium Compounds	II	P P	2	NLEAG	NE	18,19, 24,25	2,7	-	total Cd	TWMG	-	-	-	-	-	1
Tributyltin Compounds	II	v/p	3 - B	A	-	-	-	-	-	-	-	-	-	-	-	0
ORGANOCHLORINE BIOCIDES																
Aldrin / Dieldrin	I	V V	3 - 4	EO	-	22	-	-	A	GWM	e	-	-	e	eu	~ 1
Chlordane	I			(check)	-	21	-	-	A (α,γ)	GW	e	-	-	e	eu	~ 1
DDT / DDD / DDE	I	v / p	2	EO	-	21	-	-	A	GWM	eu	m	u	eu	eu	1
Endrin	II	V V	3	EO	-	-	-	-	A'	G	e	-	-	e	eu	~ 1
Heptachlor / Heptachlor Epoxide	II	V V	3 - 4	NG	-	-	-	-	A'	G	e	-	-	e	eu	~ 1
Hexachlorocyclohexanes	II	v	1 - 2	nEOB	B	21,22	1	-	A (α,γ)	GM	e	-	-	e	eu	~ 1
Methoxychlor	II	v / p	3	NOG	-	-	-	-	A'	G	e	-	-	e	e	~ 1
Mirex	I	(V V)	2	EO	-	-	-	-	A'	-	e	-	-	e	eu	~ 1
Pentachlorophenol	II	V V	2	NEAG	-	-	-	-	-	O	-	-	-	-	-	0
Toxaphene	I	v / p	2	EO	-	21	10	-	C	-	s	s	-	-	u	~ 1

Table 7. Summary of Information About the Atmospheric Transport and Deposition of Persistent Toxic Substances to the Great Lakes

column#'s (see notes for descriptions)→	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17
		V/P	LRT	Geographically Resolved Emissions Inventories		Existing Modeling Results or Planned Analyses			Air & Precipitation Monitoring in G.L. Region		Water Monitoring in the Great Lakes: Any Samples 1992-1996?					
Compound or Compound Group	Level	Vapor-Particle Partitioning	Long-Range Air	Inventory Which May Exist or May Soon Exist	Inventory Suitable for	receptor-oriented	comprehensive	source-	Integrated Atmos. Dep. Network	Other Programs	Superior	Michigan	Huron	Erie	Ontario	Loading Estimate Possible?
INDUSTRIAL / MISCELLANEOUS																
4-Bromophenyl Phenyl Ether	II	V V	3	-	-	-	-	-	-	-	-	-	-	-	-	0
3,3'-Dichlorobenzidene	II	v / p	2	NA	-	-	-	-	-	-	-	-	-	-	-	0
Hexachloro-1,3-Butadiene	II	V V	1	NG	N	-	-	-	-	NV	e	-	-	e	e	~ 1
4,4'-Methylene bis (2-Chloroaniline)	II	v	3 - 4	NA	-	-	-	-	-	-	-	-	-	-	-	0
Octachlorostyrene	I	v	2	A	-	-	-	-	A'	O	e	-	-	e	eu	~ 1
CHLOROBENZENES																
1,4-dichlorobenzene	II	V V	2	NLA	N	-	-	-	-	GNWV	e	-	-	e	e	~ 1
Tetrachlorobenzenes	II	V V	1	nL	-	-	-	-	A'	G	e	-	-	e	eu	~ 1
Pentachlorobenzene	II	V V	1	nL	-	-	-	-	A'	G	e	-	-	e	eu	~ 1
Hexachlorobenzene	I	V V	1	NCLEAG	N	22	-	13	A	GOWM	e	-	-	e	eu	~ 1
POLYCHLORINATED DIBENZO-P-DIOXINS & DIBENZOFURANS (PCDD/F'S) and POLYCHLORINATED BIPHENYLS (PCB'S)																
PCDD/F's	I	v / p	2	NCLEOAGFR	N	-	2,9	13,14	-	BPV	e	u	-	e	-	½ (G)
PCB's	I	v / p	2	NCLEOG	-	21,22	-	-	A (E)	MV	eu	mu	u	eu	eu	1
POLYCYCLIC AROMATIC HYDROCARBONS																
Anthracene	II	V V	3	NcleOAG	N	22	-	-	A	KV	-	-	-	-	u	½ (H)
Benz (a) Anthracene	II	v / p	2	NcleOG	N	22	-	-	A	KVW	e	u	-	e	u	~ 1
Benzo (a) Pyrene	I	p	2	NcleOAG	N	22	-	-	A	KVW	e	u	-	e	eu	~ 1
Benzo (g,h,i) Perylene	II	p	2	NcleOG	N	-	-	-	A	KV	e	u	-	e	eu	~ 1
Dinitropyrenes	II	v / p	2	OA	-	-	-	-	-	O	-	-	-	-	-	0
Phenanthrene	II	(V V)	3	NcleO	N	-	-	-	A	KWV	e	u	-	e	eu	~ 1
Perylene	II	p	2	eO	-	-	-	-	-	K	-	-	-	-	-	0
PAH's as a group	II-a	v / p	2	NCLEOAG	N	-	2	-	A	KVWM	e	u	-	e	eu	~ 1

Column-by-Column Notes for Table 7.

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) P P = Compound is expected to exist almost entirely in the particle phase in the atmosphere (fraction adsorbed ≥ 98% under all conditions) p = Compound is expected to exist mostly in the particle phase in the atmosphere (fraction adsorbed ≥ 90% under all conditions) v/p = Compound is expected to exist in significant proportions in both the particle phase and the vapor phase as conditions vary v = Compound is expected to exist mostly in the vapor phase in the atmosphere (fraction adsorbed ≤ 10% under all conditions) V V = Compound is expected to exist almost entirely in the vapor phase in the atmosphere (fraction adsorbed ≤ 2% under all conditions)
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	**Abbreviations for Inventories:** N = USEPA National Toxics Inventory, Version 2.0 C = USEPA Clean Air Act Section 112(c)(6) H = USEPA National Mercury (Hg) Study L = USEPA document series: Locating and Estimating Air Emissions from Sources of... E = Inventories based on Environment Canada emissions databases and information, prepared for this analysis by David Niemi/Marc Deslauriers of Environment Canada O = Ontario and Eastern North America Inventory prepared by Ortech for OMOEE A = Canada Ontario Agreement Inventory (initially prepared by David Putnam; recently updated by Envr. Canada and the Ontario Ministry of Environment and Energy G = Great Lakes Regional Toxics Inventory B = ORTECH/Environment Canada Biocide Inventories F = Canadian PCDD/F Inventory to be Developed by Federal Provincial Task Force on PCDD/F R = U.S. PCDD/F Inventory being Developed in Conjunction with Dioxin Reassessment (contact: David Cleverly, EPA) Note: if the compound is included as part of a group in the inventory, then it is listed as a small case letter corresponding to the above inventory codes (e.g., the USEPA National Toxics Inventory includes a general group entitled hexachlorocyclohexanes; thus, each particular congener is listed in this table with an "n", instead of an "N")
6	Inventories with relatively complete emissions estimates, with geographical resolution *smaller* than the province, state, or federal level, *that could be obtained in this analysis*.
7-9	Modeling analyses with available results, or with results expected; numbers in columns refer to numbers assigned to modeling analyses in Table 5.
10	The IADN program consists of 5 master stations, plus ~14 satellite stations, in the Great Lakes region. Air and precipitation concentrations of the indicated compounds are measured at each site. A = measured in program; A' = measured at some or all Canadian IADN sites, but not at U.S. IADN sites; for additional notes, see below.
11	**Other air and precipitation monitoring programs.** If information was available, the number of monitoring sites within 0-10 km of a Great Lake, the number of sites between 10-100 km of a Great Lake, and the number of sites in States/Provinces adjoining the Great Lakes (Manitoba and Quebec were also included) are listed.

Code	Program	Sponsor	Phase Sampled	Compounds	0-10 km	10-100 km	> 100 km
D	Mercury Deposition Network	various	precipitation	total mercury	1	5	6
K	Toxics Sampling Network-PAH sites	Envr Canada	vapor + particulate	PAH's only	4	1	4
O	Toxics Sampling Network-COA sites	Envr Canada	vapor + particulate	OCS, DNP, HCB, PCP	4	1	0
N	Toxics Sampling Network-VOC sites	Envr Canada	vapor	VOC's only	7	4	11

	B	Toxics Sampling Network-PCDD/F	Envr Canada	vapor + particulate	PCDD/F only	4	1	4
	T	Toxics Sampling Network-metals	Envr Canada	particulate	incl. total Cd, Pb, Hg	3	2	5
	C	CAMNet	Envr Canada	vapor (+precip @>100km site)	total Hg	1	1	1
	G	Great Lakes Precipitation Network Can. Centre for Inland Waters	Envr Canada	precipitation	many compounds	7	2	0
	P	PCDD/F Sampling	OMOEE	vapor + particulate	PCDD/F only	1	1	0
	M	"air toxics monitoring"	Michigan DNR	vapor + particulate	ongoing (?)	1	3	0
	L	"Lake Champlain Study"	?	vapor, particulate, precip. (?)	mercury	0	0	1
	V	Haz. Air Contam. Monitoring Prog.	Vermont ANR	vapor + particulate	many compounds	0	0	5
	W	Green Bay Urban Air Toxics Monitor.	Wisconsin DNR	vapor + particulate	many compounds	1	0	0
	U	Air Quality Laboratory monitoring	Univ. Michigan	vapor, particulate, precip. (?)	mercury	0	1	0
	F	Air Quality Laboratory monitoring	"trust fund"	vapor, particulate, precip. (?)	mercury	1	0	0
12-16	**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)							
17	**Loading Estimate Possible?** [0 = no; 1 = yes; ½ = somewhat possible] "~ 1" 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.							

ADDITIONAL NOTES for Table 7, relating to columns 2,4, 10, and 12-17 only. *Note: these letter-coded notes are not relevant to column 5 or 11.*

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.
B	Tributyltin 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".
C	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.
D	Other PAH's included in the IADN program include coronene and retene
E	Congener specific measurements and homologue group totals
F	Except for Lake Mich., 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.
G	Vapor- and particulate-phase PCDD/F is being measured in multi-site monitoring programs in Canada, sponsored by Envr. 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.
H	Only data for Lake Ontario could be found; thus estimates could be made for that Lake. Crude estimates could possibly be made for the other Great Lakes, using the water concentrations found in Lake Ontario, as a first approximation.
J	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.

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