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Agency

Office of Air Quality
Planning and Standards
Research Triangle Park, NC 27711

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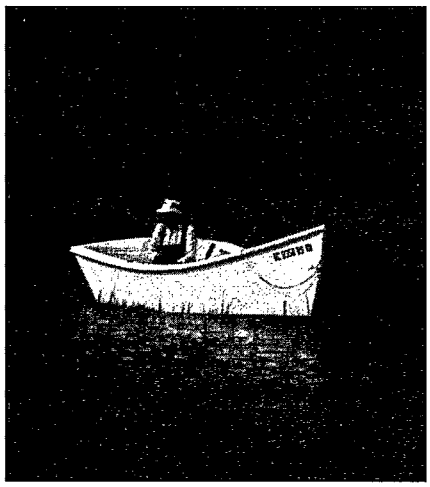
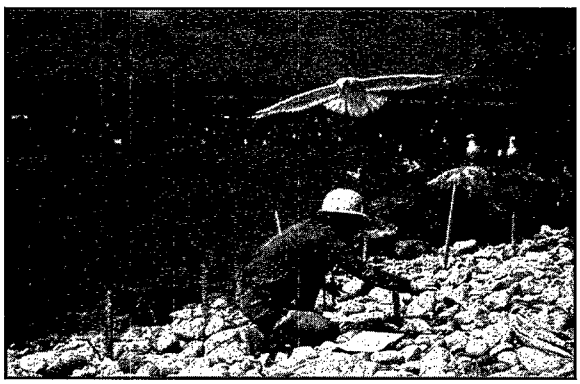
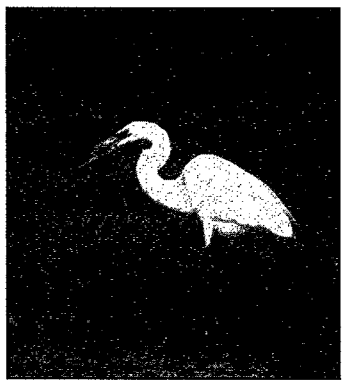
May 1994

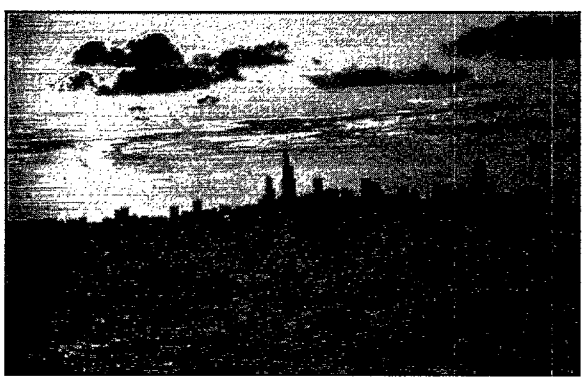
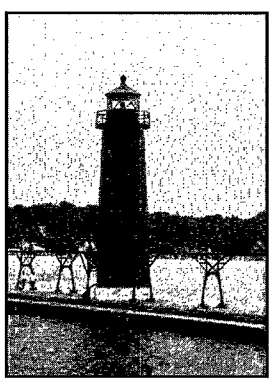
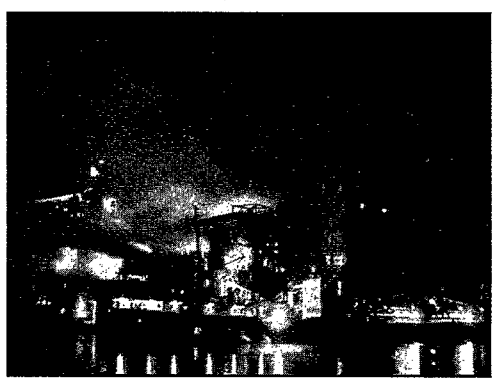
EPA

Deposition of Air Pollutants to the Great Waters

First Report to Congress



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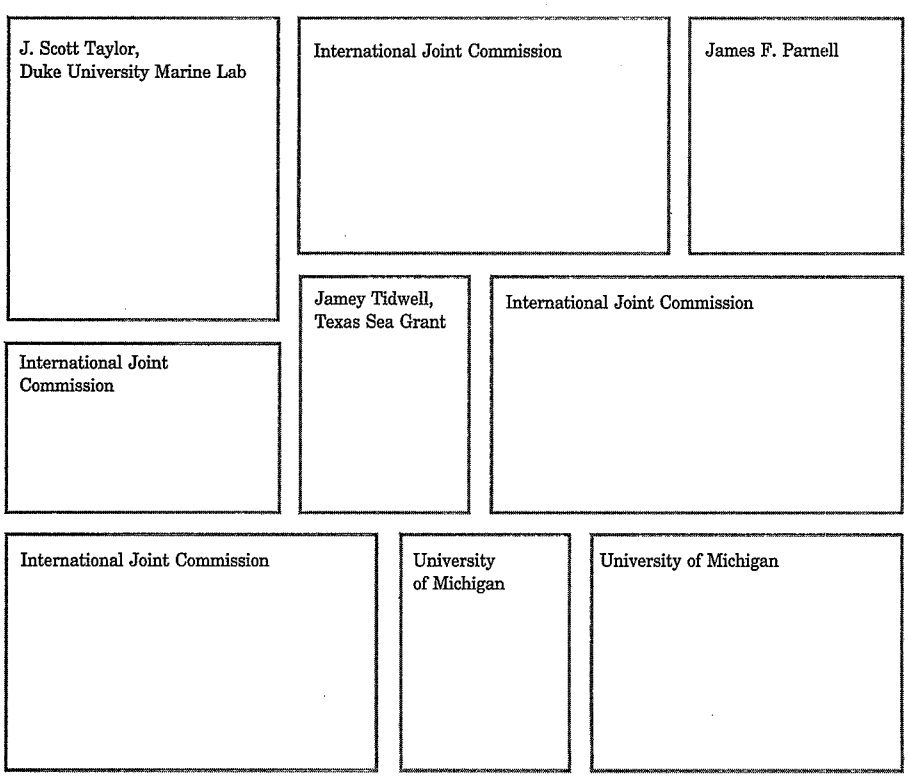

Copies of the first Report to Congress, *Deposition of Air Pollutants to the Great Waters*, can be obtained, as supplies permit, from the Library Services Offices (MD-35), U.S. Environmental Protection Agency, Research Triangle Park, North Carolina 27711, or, for a nominal fee, from the National Technical Information Service, 5285 Port Royal Road, Springfield, Virginia 22161, phone: 1-800-553-NTIS or 703-487-4650.

Information in this Report to Congress has been derived mainly from three detailed background reports prepared by committees of leading independent scientists. These committees were convened by EPA to summarize the current state of scientific knowledge on atmospheric deposition to the Great Waters. Unless otherwise referenced, all scientific information in this report is drawn from the three technical contractor reports:

1. *Relative Atmospheric Loadings of Toxic Contaminants and Nitrogen to the Great Waters*. 1993. Describes the mass balance approach for determining inputs into surface water. Discusses waterbody-specific mass balance calculations for several pollutants.

2. *Identification of Sources Contributing to the Contamination of the Great Waters by Toxic Compounds*. 1993. Describes techniques for source identification. Discusses the importance of local, regional, and distant sources for atmospheric deposition to the Great Waters.

3. *Exposure and Effects of Airborne Contamination*. 1992. Provides a detailed summary of the ecological and human health effects of selected air pollutants of concern for deposition to the Great Waters.

Copies of these reports can be obtained by writing:

Office of Air Quality Planning and Standards
Pollutant Assessment Branch (MD-13)
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

Attention: Great Waters Documents

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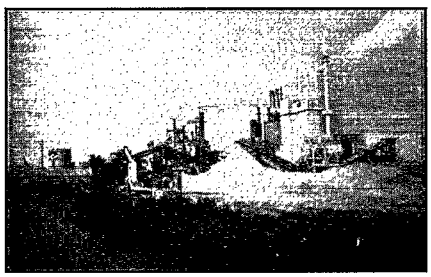

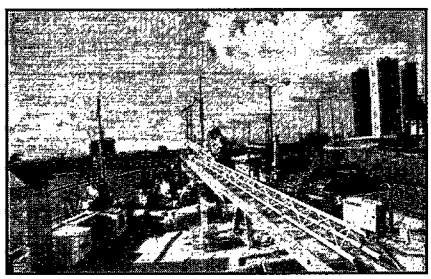

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Abbreviations and Acronyms

AAs	Assistant Administrators
ACTs	Achievable control technology documents
ANPR	Advance notice of proposed rulemaking
AWQC	Ambient water quality criterion or criteria
BCCs	Bioaccumulative chemicals of concern
CWA	Clean Water Act
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
EPA	U.S. Environmental Protection Agency
FIFRA	Federal Insecticide, Fungicide, and Rodenticide Act
g	Gram
GLWQA	Great Lakes Water Quality Agreement
GLWQB	Great Lakes Water Quality Board
GLWQO	Great Lakes Water Quality Objective
HAP	Hazardous air pollutant
HCH	Hexachlorocyclohexane
IJC	International Joint Commission
kg	Kilogram
L	Liter
LQER	Lesser-quantity emission rates
MACT	Maximum achievable control technology
MCL	Maximum contaminant level
m^3	Cubic meter
mg	Milligram
NEP	National Estuary Program
NERRS	National Estuarine Research Reserve System
ng	Nanogram
NOAA	National Oceanic and Atmospheric Administration
NO_x	Oxides of nitrogen
NPDES	National Pollutant Discharge Elimination System
PAH	Polycyclic aromatic hydrocarbon
PCB	Polychlorinated biphenyl
pGLWQC	Proposed Great Lakes water quality criteria
POM	Polycyclic organic matter
ppb	Parts per billion
ppm	Parts per million
ppt	Parts per trillion
RAs	Regional Administrators
TCDD	Tetrachlorodibenzo-p-dioxin
TCDF	Tetrachlorodibenzofuran
TSCA	Toxic Substances Control Act
WHO	World Health Organization
yr	Year

Acknowledgments

The EPA would like to acknowledge the significant contribution that was made in the development of this report by the scientific community. The authors of the three background documents, upon which the scientific content of this Report to Congress is based, prepared these documents in a difficult time frame and worked long hours to revise them in response to peer review comments. These author teams were:

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Also, EPA would like to express appreciation to the wider community of scientists, both within and outside the Agency, who participated in the workshop to discuss and evaluate the draft technical background reports and to all of the participating scientists for their efforts in reviewing drafts of Chapter 3 of this Report to Congress.

In addition, many employees throughout the EPA have made significant contributions to this Report to Congress.

James F. Parnell

Executive Summary

Pollutants emitted into the atmosphere are transported various distances and can be deposited to aquatic ecosystems far removed from their original sources. Scientific studies show that atmospheric deposition is often an important factor in the degradation of water quality and the associated adverse health and ecological effects in studied waterbodies. In response to the mounting information indicating that air pollution contributes significantly to water pollution, Congress included section 112(m), referred to as the Great Waters program, in the Clean Air Act, as amended in 1990 (1990 Amendments). This report fulfills the Act's requirement for a Report to Congress 3 years after enactment.

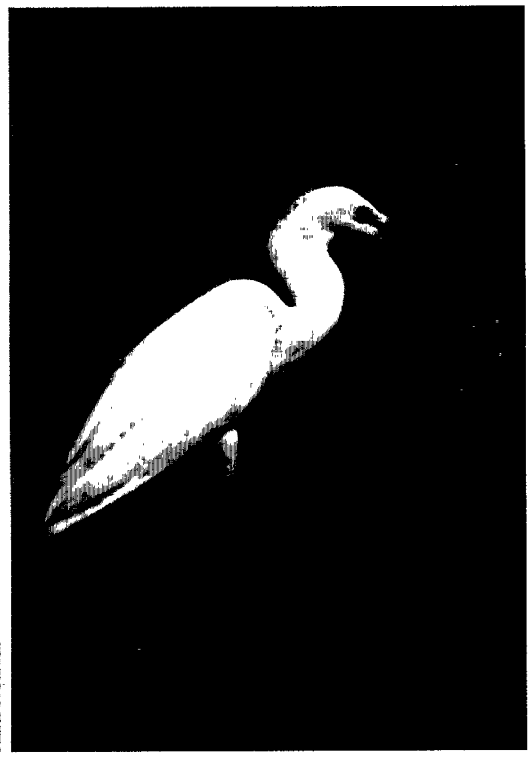
The purpose of the Great Waters program is to evaluate the atmospheric deposition of air pollutants to the Great Lakes, Lake Champlain, Chesapeake Bay, and coastal waters. The report to Congress is to include information on the contribution of atmospheric deposition to pollutant loadings, the environmental or public health effects of such pollution, the source or sources of such pollution, and a description of any regulatory revisions under applicable Federal laws that may be necessary to assure protection of human health and the environment.

The scientific information currently available is summarized in this report, and recommended actions are described.

Water quality conditions in the Great Lakes and many other waterbodies are greatly improved compared to a few decades ago, the result of environmental regulatory programs and public and industrial cleanup efforts addressing primarily waterborne pollution. However, despite the improvements, the Great Waters ecosystems are far from fully recovered, and it is necessary to address the more diffuse sources of pollution, including the air component, in order to attain water quality goals and to ensure protection of human health and the environment.

Pollutants of concern to the Great Waters possess certain common characteristics. They persist in the environment and, thus, can travel great distances, often being deposited and reemitted many times. These pollutants accumulate in the environment, making the potential for exposure to them greater than for pollutants that readily degrade. The potential for long-distance transport is evident by the presence of pollutants in remote, pristine environments such as the Arctic.

Pollutants of concern also accumulate in body tissues and magnify up the food web, with each level accumulating the toxics from its diet and passing the burden along to the animal in the next level of the food



James F. Parnell

web. Top consumers in the food web, usually consumers of large fish, may accumulate chemical concentrations many millions of times greater than the concentrations present in the water. As a result of unsafe concentrations of chemicals in fish, due to biomagnification, fish consumption advisories have been issued in hundreds of waterbodies nationwide, including the Great Lakes. High-risk groups, which fish consumption advisories are established to protect, include breast-feeding mothers because breast-fed babies continue to accumulate from their mothers after birth. For example, they can have PCB levels four times higher than their mothers after 6 to 9 months of breast-feeding. Other groups at high risk are subpopulations such as sport anglers, Native Americans, and the urban poor, who tend to have high fish consumption. EPA and other agencies are addressing this environmental justice issue by examining impacts to higher-risk populations and taking this into consideration in regulating activities.

Significant adverse effects on human health and wildlife have been observed due to exposure, especially through fish consumption, to persistent pollutants that bioaccumulate. Adverse effects range from immune system disease and reproductive problems in wildlife to subtle developmental and neurological impacts on children and fetuses.

Although most of the chemicals of concern are probable human carcinogens, many are also developmental toxicants capable of altering the formation and function of critical body systems and organs. Therefore, the developing embryo and fetus and breast-fed infants are particularly sensitive to these chemicals.

Ecological effects attributable to pollutants of concern are significant and can be subtle or delayed in onset, such as immune function impairment, reproductive problems, and neurological changes—all of which can affect population survival.

Other adverse ecological effects are caused by nitrogen compounds. Nitrogen compounds from atmospheric deposition exacerbate nutrient enrichment (or eutrophication) of coastal waterbodies, which results in impacts that range from nuisance algal blooms to the depletion of oxygen with resultant fish kills.

Studies show that significant portions of loadings to the Great Waters of the pollutants of concern are coming from the atmosphere. For example, 76 to 89 percent of PCBs to Lake Superior and up to 40 percent of nitrogen loadings to the Chesapeake Bay are estimated to come from air pollution. However, insufficient data are available to generalize the atmospheric loadings to all waters. Absolute quantities of deposited pollutants are also important, especially since loadings of even small amounts of pollutants that bioaccumulate can result in significant pollutant burdens in fish.

Pollutants of concern in the Great Waters originate from sources that are local to, as well as distant from, the impacted waters. Transport distances depend on the characteristics of the chemicals and source emissions as well as weather patterns. As such, generalizing source

identification from one waterbody to another would not be accurate. More data are needed to determine sources and source categories affecting the Great Waters.

Uncertainties in current information are significant, and further research is needed to better characterize the most important information for decisionmakers. However, adequate information is available to lead EPA to the conclusion that some actions are justified and necessary at this time. Adverse effects of the chemicals of concern are evident and studies of selected waters show significant proportions of toxic pollution coming from the atmosphere. However, because the linkage between specific sources and subsequent deposition and effects has yet to be demonstrated, the kinds of actions described in this report focus on the chemicals of concern rather than on specific sources.

EPA considered the implications of action and of inaction, while recognizing that section 112(m) of the 1990 Amendments mandates that EPA should act to "prevent" adverse effects and to "assure protection of human health and the environment." EPA's recommendation is that reasonable actions are justified, based on evaluation of the scientific information currently available, and should now be taken and that research should continue. The National Oceanic and Atmospheric Administration (NOAA) concurs with this recommendation.

Most of the actions EPA will undertake focus on utilizing regulatory mechanisms in the Clean Air Act that are intended to address the most hazardous chemicals. EPA believes that the characteristics of toxicity, persistence, and tendency to bioaccumulate warrant special treatment of the Great Waters pollutants of concern and that this is consistent with congressional intent for those regulatory mechanisms and for section 112(m).

The recommendations from the report fall into three strategic themes. First, EPA will continue ongoing efforts to implement section 112 and other sections of the Clean Air Act and use the results of this report in the development of policy that will reduce emissions of Great Waters pollutants of concern. Under this theme, EPA will take actions that include: publishing emission standards affecting important chemicals of concern ahead of schedule, where possible; evaluating the adequacy of control technologies for important pollutants; publishing an advance notice of proposed rulemaking (ANPR) for establishment of lesser-quantity emission rates (LQERs) to define smaller sources to be regulated as "major sources" and evaluating which Great Waters pollutants warrant establishment of an LQER; evaluating which area sources should be regulated with maximum achievable control technology (MACT); and considering appropriate emission levels requiring regulation when sources are modified.

Second, EPA recognizes the need for an integrated multimedia approach to this problem and, therefore, will utilize authorities beyond the Clean Air Act to reduce human and environmental exposure to pollutants of concern. Under this theme, EPA will take actions that

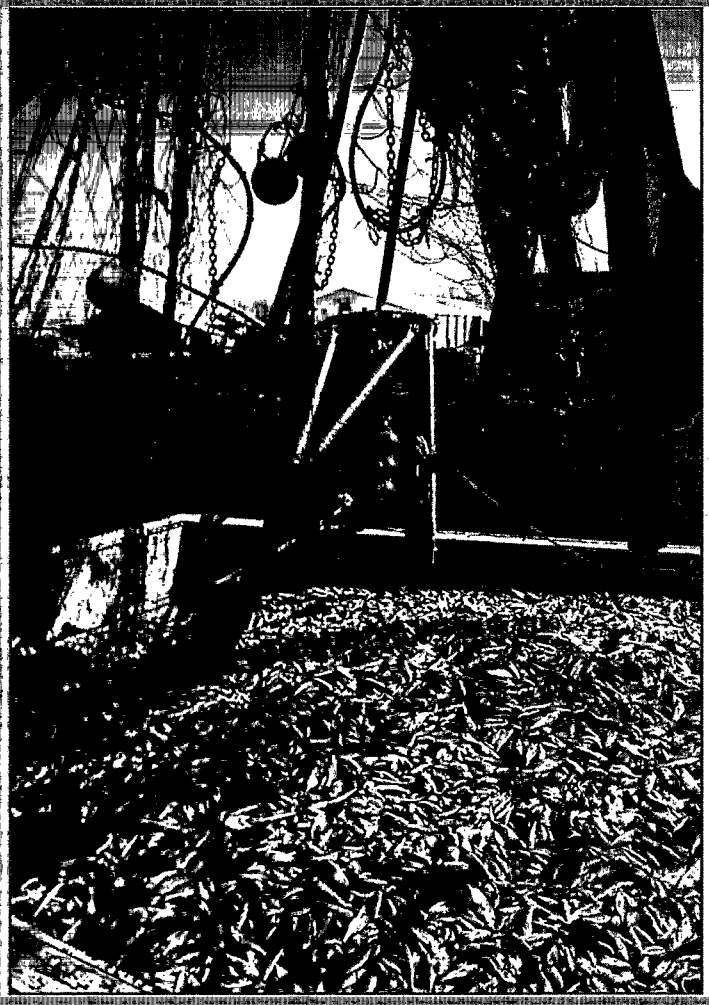
include using the Great Waters Core Project Management Group as a coordinating body to communicate with other offices/agencies. The objectives will be to: coordinate work and especially to identify lead offices to implement recommendations; support changes to the Clean Water Act that address nonwaterborne sources of water pollution; address the exportation of banned pesticides; emphasize pollution prevention efforts to reduce environmental loadings of pollutants of concern; and facilitate information sharing between EPA and other agencies.

Third, EPA will continue to support research activities and will develop and implement a program strategy to define further necessary research. Under this theme, EPA will take actions that include: focusing research planning on a mass-balance approach to determine relative loadings; using an appropriate mix of monitoring, modeling, and emission inventory tasks in conducting mass-balance work; assessing the need for tools to be developed for risk assessment for total exposure to pollutants of concern and for regulatory benefits assessment; and continuing to support ongoing research efforts.

Examples of Effects of Toxic Chemical Pollution in the Great Lakes

- People who regularly consume Great Lakes fish have been found to have higher concentrations of PCBs, DDT, and other toxic chemicals in their body than people who do not.

- Subtle neurological and behavioral effects have been reported in children of women who regularly consumed Lake Michigan fish in the 1970s. These effects are possibly the result of increased concentrations of PCBs in the mother, since PCBs can cross through the placenta and directly affect embryos in the womb.* See page 34 for further discussion of this issue.



Scott Peplow, Ball State University, Muncie, Indiana

- Fish-eating birds, mammals, and reptiles have experienced reproductive problems and a variety of other adverse effects associated with chemical pollution, leading to population declines for many species. Embryo and offspring mortality have been linked to PCBs and other toxic chemicals.

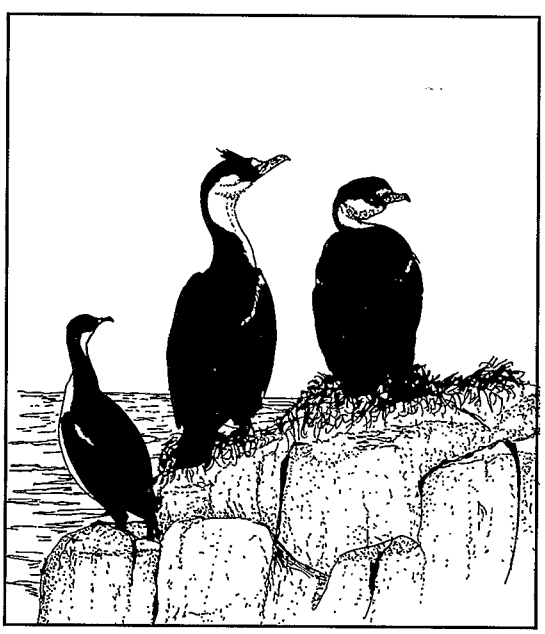
- Damage to fish has been linked to chemical pollution, including effects on hormone function, immune system response, and enzyme activity. Several studies have reported the occurrence of cancer and other toxic effects in bottom-feeding fish in tributaries and harbors of the Great Lakes.

- Commercial fishing has been closed or restricted in portions of all five Great Lakes at various times in recent years because of toxic chemical pollution, and restrictions are still in place for many fish species. Numerous health advisories have been issued to discourage consumption of certain fish caught in the Great Lakes.

*PCB concentrations in fish have decreased since the 1970s. However, fish consumption advisories exist for certain types of fish in specific lakes due to current PCB concentrations in the fish.

Evidence for Long-Distance Atmospheric Transport

- Chemicals from anthropogenic sources (i.e., created by humans) are present in Arctic and Antarctic ecosystems, thousands of miles from likely emission sources.
- Lead and other trace metals have been measured in air and rainfall at remote locations over the Atlantic and Pacific Oceans, thousands of miles from likely sources.
- European research suggests that the major sources of trace metals and persistent organic chemicals deposited from air into the Baltic Sea are located hundreds of miles away.



amounts in the United States since the 1970s, have become widely distributed in the environment and are now, in essence, part of the global "background." These toxic chemicals remain in our environment and continue to cycle between air, water, soil, and biota (living organisms), even after their manufacture, use, or release has stopped.

A number of recent field studies have demonstrated that air pollution is an important contributor to chemical pollution of U.S. lakes and coastal waters. Some of these studies are described below.

- Air and rainfall in the Great Lakes region, the Chesapeake Bay watershed, and other areas have repeatedly been shown to be contaminated with a variety of toxic chemicals. PCBs, for example, are present in the air above all five Great Lakes and are also present (at roughly similar levels) in the air above Chesapeake Bay. PCB levels either currently exceed or have recently exceeded water quality standards in portions of all of the Great Lakes (see appendix B).

- A recent series of studies of Wisconsin lakes indicate that the air is a major contributor of mercury to these lakes and that modest increases in atmospheric deposition of mercury could lead directly to higher levels of mercury in fish. These studies are in broad agreement with research on mercury deposition to Swedish lakes.

- Studies of fish from Siskiwit Lake–a small lake on an island in northern Lake Superior that is isolated from most human influences – have shown contamination with PCBs, toxaphene, and other pesticides, which have no known sources on the island. Toxaphene, a pesticide banned in the United States in 1982, had limited use in the Lake Superior region but was used heavily in the southeastern U.S. Cotton Belt from the late 1960s to the mid-1970s. This use pattern indicates that toxaphene was probably transported by air from the Southeast to the Great Lakes region. Airborne levels of toxaphene are highest in the southeastern U.S. and decline with distance as one moves toward the Great Lakes and north Atlantic regions.[1]

- It is likely that other pesticides present in the Great Lakes, including DDT, are transported long distances by air, from their sources to the Great Lakes region.[2] Based on the amount and chemical form of DDT present in core samples from peat bogs in the Great Lakes region, new releases of DDT are apparent and may be originating from sources outside the United States, possibly Mexico and Central America.[2] Atmospheric deposition of DDT, toxaphene, hexachlorobenzene, and PCB levels in the Great Lakes region, as measured in peat cores, are

University of Michigan

consistent with the U.S. production and use history of these chemicals.[3]

- Long-term sampling of precipitation falling onto coastal waters near Lewes, Delaware, indicates that concentrations of most trace metals in rainfall have been fairly constant in recent years and are far greater than concentrations in most surface waters. A notable exception is the concentration of lead in rainfall, which has declined since the major reduction in use of leaded gasoline in the United States in the mid-1980s.[4]

- Various forms of nitrogen, a nutrient that can cause undesirable effects in coastal marine waterbodies when present in excessive amounts, have been measured in rain falling on Chesapeake Bay and its watershed. A significant fraction of the total nitrogen entering Chesapeake Bay (28 to 40 percent) and several other estuaries is believed to come from atmospheric deposition.

Recent monitoring studies conclude that the air is supplying approximately 77-89, 63, and 58 percent of the PCBs *currently* entering Lakes Superior, Huron, and Michigan, respectively. Atmospheric input accounts for more than 95 percent of the lead entering these waterbodies. Overall, scientists estimate that 35 to 50 percent of current yearly inputs of a variety of toxic chemicals to the Great Lakes may be from the air. Similar studies indicate that atmospheric deposition is an important source of metals, polycyclic aromatic hydrocarbons (PAHs, a subgroup of polycyclic organic matter), PCBs, and nitrogen compounds to Chesapeake Bay. Monitoring studies, along with direct measurements of pollutants entering waterbodies in precipitation, show that air pollution is not just a theoretical source of the toxic chemicals present in large lakes and coastal waters but is, in some cases, a significant contributor to the overall amount of pollution entering waterbodies (see Tables 7 and 8, page 55, and Figure 11, page 52).

The Clean Air Act establishes research, reporting, and regulatory requirements related to atmospheric deposition to the Great Waters, including a mandate that EPA, in cooperation with the National Oceanic and Atmospheric Administration, submit a Report to Congress in 1993 and every 2 years thereafter.

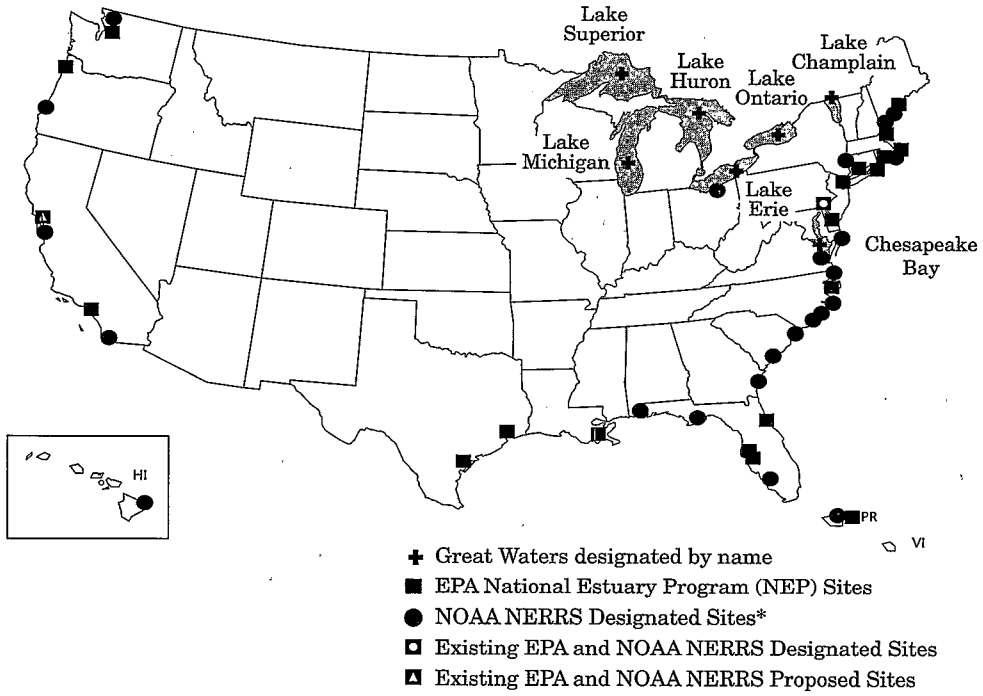
Section 112(m) of the Clean Air Act, as Amended in 1990

Section 112(m) of the 1990 Amendments establishes research and reporting requirements related to atmospheric deposition of hazardous air pollutants* to the Great Lakes, Lake Champlain, Chesapeake Bay, and coastal waters (defined in the statute to include coastal waters in the National Estuary Program or the National Estuarine Research

*Section 112(m), and therefore this Report to Congress, does *not* address acid rain. This report addresses pollutants from the section 112(b) list of 189 hazardous air pollutants, along with two other pollutants of concern.

Reserve System). Figure 1 shows the locations of currently designated Great Waters.

This report focuses primarily on the Great Lakes because much of the available scientific information on deposition of toxic air pollutants is from studies of the Great Lakes. Where data were available, studies of the Chesapeake Bay are cited. The Chesapeake Bay is one of the only coastal waterbodies for which data exist on atmospheric deposition, loadings, and effects of nitrogen and toxic air pollutants. Continuing research, building on available information, particularly from the Great Lakes, is examining whether information from the Great Lakes and Chesapeake Bay is applicable to other Great Waters.



*NOAA=National Oceanic and Atmospheric Administration; NERRS=National Estuarine Research Reserve System

Figure 1. Locations of designated Great Waters.

Section 112(m) directs EPA, in cooperation with the National Oceanic and Atmospheric Administration (NOAA), to assess the extent of atmospheric deposition of hazardous air pollutants to the Great Waters. As part of this assessment, EPA is to monitor atmospheric deposition, investigate sources and deposition rates, conduct research to improve monitoring methods and to determine relative loadings, evaluate human health and environmental effects,* assess violations of water quality standards, and sample fish and wildlife for atmospherically deposited pollutants. Section 112(m) specifically requires that EPA establish atmospheric deposition monitoring networks in the Great Waters. In addition, EPA must determine whether the other regulatory programs under section 112 are "adequate to prevent serious adverse effects to public health and serious or widespread environmental effects" associated with atmospheric deposition to the Great Waters. Based on this determination, EPA is directed to take additional measures that are necessary and appropriate to prevent serious effects to human health and the environment.

In addition to the above requirements, section 112(m)(5) directs EPA, in cooperation with NOAA, to submit, by November 1993 and

*Environmental effects include both ecological effects and other welfare effects such as the commercial impacts of depleted fish populations or lost recreational opportunities.

Monitoring Progress, 1990-1993

Through the cooperative efforts of various EPA, NOAA, and State programs, much progress has been made in deposition monitoring.

Great Lakes

- Five master/regional background stations (one per lake) are collecting wet and dry toxic deposition samples (since 1992).
- Complementary stationary stations and ship-based intensive collection efforts are being undertaken for toxic loading and mass-balance work in Lake Michigan, 1993-1996.

Lake Champlain

- NOAA, the Lake Champlain Research Consortium, and the Vermont Monitoring Cooperative, in cooperation, are conducting mercury monitoring and research on wet and dry deposition of nutrients and hazardous air pollutants (HAPs) in the Lake Champlain basin.

Chesapeake Bay and Other Coastal Waters

- From 1990-1993, three stations collected wet and dry toxics deposition samples in the Chesapeake Bay.
- Intensive collection efforts for characterization of urban plume influence to the Chesapeake Bay are being undertaken.
- NOAA conducts the Atmospheric Nutrient Input to Coastal Areas (ANICA) program to determine the fraction of coastal nutrient pollution that comes by way of the atmosphere. So far, efforts have centered on the northeastern U.S. coast, with a focus on Chesapeake Bay, and have been accomplished through enhancement of existing monitoring networks.
- NOAA conducts the Atmospheric Integrated Research Monitoring Network, which is intended to supply data to atmospheric modelers for evaluating the changes in nitrogen and other atmospheric contaminants due to legislated emission reductions. Progress to date includes the establishment of four daily monitoring stations in or near northeastern U.S. Great Waters drainage basins.
- A screening-level toxics monitoring effort in Galveston Bay is collecting various toxics, comparable in method to work in the Chesapeake Bay.

every 2 years thereafter, a Report to Congress on atmospheric deposition to the Great Waters. The report is to describe "results of any monitoring, studies, and investigations conducted pursuant to" section 112(m) and, at a minimum, is to include an assessment of:

- the contribution of atmospheric deposition to pollution of the Great Waters,
- environmental and human health effects of air pollutants that are deposited to the Great Waters,
- sources of air pollutants that are deposited to the Great Waters,
- whether atmospheric deposition contributes to violations of drinking water standards or water quality standards or exceedances of Great Lakes Water Quality Agreement objectives, and
- regulatory changes needed to ensure protection of human health and the environment.

In the 1990 Amendments to the Clean Air Act, Congress placed special emphasis on mercury as a toxic air pollutant. Several subsections in section 112 contain special requirements for the study of mercury and the regulation of mercury emissions. Section 112(c)(6) lists mercury, along with six other pollutants (all of which are listed in this report as Great Waters pollutants of concern) and requires that EPA identify and regulate the sources responsible for at least 90 percent of total air emissions of each pollutant. Section 112(n)(1)(A) requires EPA to perform a study of the hazards to public health that are anticipated to occur as a result of emissions from electric steam-generating units. A report to Congress on the results of this study is scheduled to be completed in November 1995. Section 112(n)(1)(B) requires EPA to conduct a study of the air emissions of mercury from electric utilities, municipal waste combustors, and other sources, including area sources. A separate report on the results of this study is due to Congress by November 1994. Section 112(n)(1)(C) directs the National Institute of Environmental Health Sciences to conduct a study to determine a "threshold" level for human health effects from mercury exposure, including a threshold for mercury concentrations in fish that may be eaten by humans. Because of the strong evidence indicating the importance of atmospheric deposition of mercury to waterbodies, as well as the attention given mercury in section 112, this

International Joint Commission

report includes mercury as a "case study" pollutant in each of the three main scientific sections.

Report Objectives

The main objectives of this Report to Congress are to describe what is known about atmospheric deposition of toxic chemicals to the Great Waters and to present any appropriate regulatory recommendations based on the *currently* available scientific information. The report, along with its supporting documentation, is intended to assemble information that will allow EPA to (1) determine the extent to which air pollution is a significant contributor to water quality problems in the Great Waters, (2) evaluate the effectiveness of current regulatory programs in addressing known or potential problems, and (3) decide whether additional regulatory actions are needed. The report also summarizes progress to date on research initiatives under section 112(m), identifies critical research needs, and describes the program strategy that is being developed to address research and regulatory needs. It also serves as a starting point for EPA's future assessments of the scientific data available for subsequent biennial reports. This Report to Congress on the deposition of air pollutants to the Great Waters addresses only atmospheric deposition of toxic chemicals and nitrogen compounds, and *not* acid rain. Atmospheric deposition of nitrogen compounds is addressed because of nitrogen's role in eutrophication of many coastal estuarine and marine waters.

Report Preparation

This Report to Congress summarizes the current understanding of atmospheric deposition of toxic chemicals to the Great Waters and identifies key regulatory and research needs, based in part on inputs from leading independent scientists.

EPA's and NOAA's approach to preparing this report relied heavily on participation by independent scientists who have conducted research on atmospheric deposition of toxic pollutants. As a first step, EPA sponsored a literature search on the topic of atmospheric deposition of chemicals to surface waters, identifying more than 1,100 scientific publications.[5] EPA then convened three committees of leading independent scientists and charged them with evaluating and summarizing the scientific literature in the three areas identified in section 112(m): exposure and effects of atmospheric deposition to the Great Waters, relative atmospheric loadings to the Great Waters, and sources contributing to atmospheric deposition to the Great Waters. Each committee prepared a draft paper, and, in November 1992, EPA sponsored a 2-day workshop to discuss and comment on the draft papers. Attendees of the workshop included the committee members, other independent scientists, EPA scientists, EPA program representatives, and representatives of groups that include NOAA, State agencies, and industry and environmental groups. Following the workshop, the committees prepared final

documents that represent syntheses of much of the scientific knowledge in the three areas identified.[6,7,8] This Report to Congress condenses the information in these three scientific background papers into a relatively concise and readable report. All scientific data and conclusions presented in this Report to Congress, except those specifically referenced to other sources, are drawn from the three background papers, which are fully referenced.

This document fulfills the section 112(m)(5) requirements for the first Report to Congress on atmospheric deposition to the Great Waters. Chapter 1 introduces the section 112(m) requirements and the report objectives, as well as provides examples of studies that identify atmospheric deposition as a contributor to chemical pollution of the Great Waters. Chapter 2 summarizes the activities of EPA's Great Waters Program and highlights the questions it is addressing. Chapter 3 addresses the scientific questions of section 112(m) by summarizing information on exposure and effects, relative loadings, and sources. Chapter 4 presents conclusions from an evaluation of the science and provides regulatory recommendations. All references cited (i.e., the references for information taken from sources other than the three scientific background papers) are listed in Chapter 5. The Appendices include, among other items, a description of past EPA regulations, a summary of current section 112 activities, a summary of progress to date on research initiated in response to section 112(m), and a description of research needs. Future reports to Congress, required every 2 years, will provide more complete information as it becomes available.

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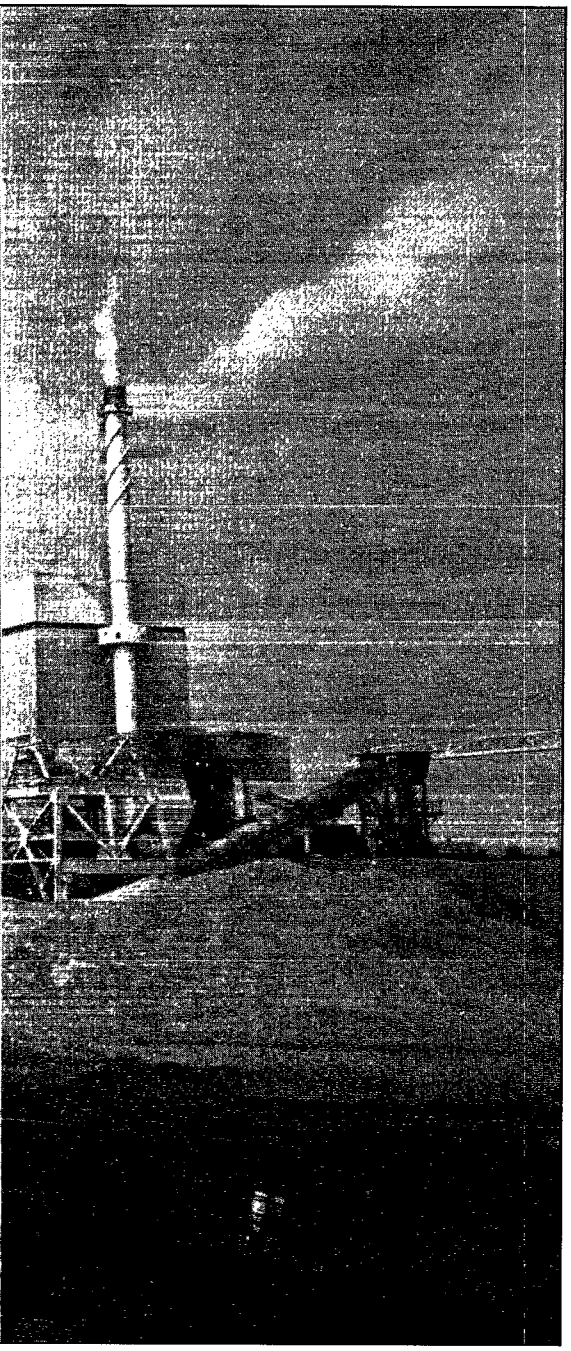
Chapter Two
Overview of the Great Waters Program



Section 112(m) of the Clean Air Act, as amended in 1990 (1990 Amendments), raises numerous questions regarding the extent and significance of atmospheric deposition of toxic chemicals to the Great Waters. EPA's Great Waters Program is attempting to answer these questions. Answers to these scientific questions will provide the information necessary to determine the need for additional regulatory actions to reduce atmospheric deposition to the Great Waters.

Scientists have long recognized the basic process by which air pollutants can enter rivers, lakes, and other waterbodies. The steps in this process are shown in Figure 2.

- First, pollutants are *released* to the air from a source, which may be natural or anthropogenic (i.e., created by humans). Anthropogenic sources include point sources, such as industrial smokestacks or any other fixed location that releases pollutants, and area sources, such as pesticide applications on agricultural fields and vehicle exhaust. Natural sources also can be classified as either point or area sources and include, for example, forest fires, volcanic eruptions, windblown dust and soil, and sea spray. Pollutants can be released either as gases or as particles.

- Second, pollutants released to the air are *transported* away from their source to other locations. Depending on weather conditions and the chemical and physical properties of the pollutant, air pollutants may be transported either short or long distances from their sources and may undergo physical and chemical changes while in transit.

- Third, air pollutants are *deposited* to the earth, in most cases directly to a waterbody or to a land area that drains into a waterbody. Pollutants are deposited by "wet deposition" or "dry deposition." In wet deposition, pollutants are removed from the air by a precipitation event such as rain or snow. Dry deposition occurs when particles settle out of the air and into water. Air pollutants can also enter a waterbody indirectly, by first depositing onto surrounding land or tributaries and then moving into the waterbody by other routes, such as stormwater runoff or inflow from tributary streams.

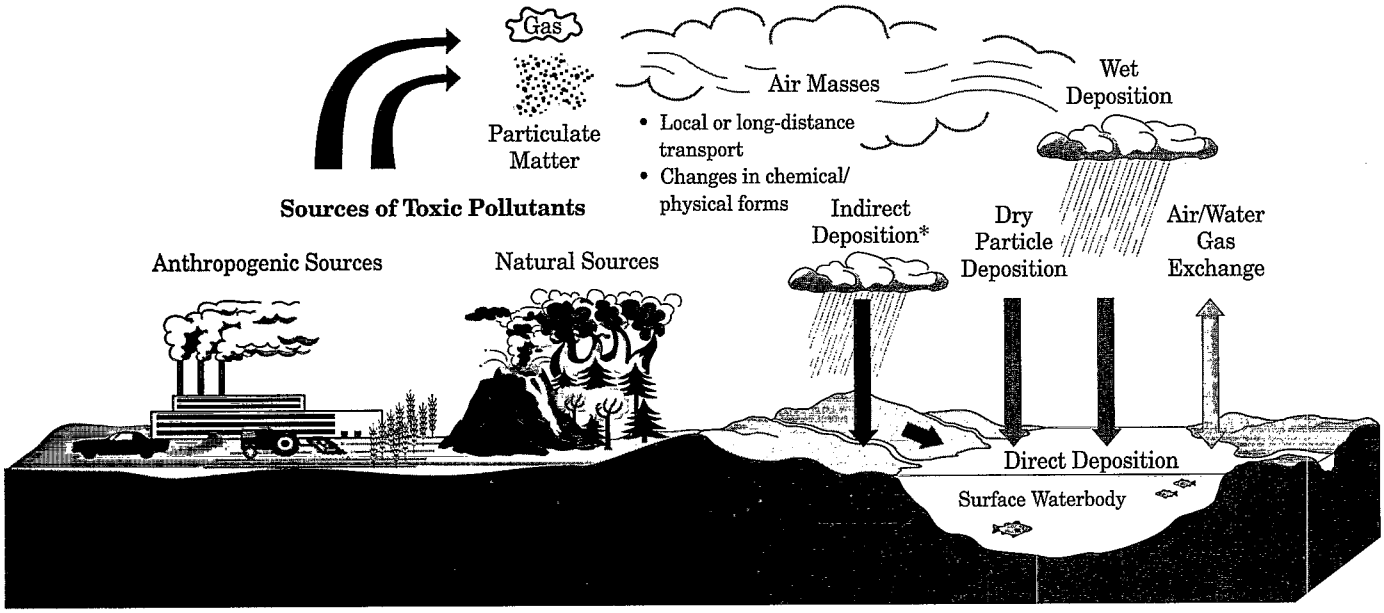


Figure 2. How does atmospheric deposition occur?
* Indirect deposition is direct deposition to land followed by runoff or seepage through groundwater to a surface waterbody.

Current understanding of the details of each of these steps is limited, although it is growing as a result of recent scientific research (see Figure 3). As early as 1907, localized atmospheric deposition of metals around smelters was reported.[10,11] In the late 1960s, the discovery of anthropogenic chemicals (i.e., chemicals created by humans) in Antarctic snow provided strong evidence that air pollutants can travel long distances and be deposited in remote areas. Additional confirmation of toxic chemical contamination caused by atmospheric deposition was provided in the 1970s by, among other studies, the reporting of anthropogenic chemical contamination in Arctic mammals and the discovery of PCBs and toxaphene in fish in the isolated waters of Siskiwit Lake on an island in Lake Superior. Studies of atmospheric deposition continued throughout the 1980s and included mass balance studies of PCBs and other toxic chemicals in the Great Lakes, implicating air pollution as a major contributor to contamination of waterbodies. These studies have yielded considerable information about how atmospheric deposition occurs and the role and significance of air pollution in influencing water quality.

As part of the Great Waters Program, four major activities have been identified that will increase the current understanding of atmospheric deposition. Each of these activities addresses different scientific and regulatory questions, as shown in Table 1. These activities provide a logical framework for deciding what actions are needed to reduce atmospheric deposition to the Great Waters, thereby minimizing the effects caused by the deposited pollutants.

The United States and Canada, through joint efforts of EPA, Environment Canada, and Ontario's Ministry of Environment and Energy, have been implementing a bilateral program on airborne toxic substances in the Great Lakes basin since 1990. This program includes bilateral cooperation in monitoring of toxic air deposition as part of the Great Lakes Water Quality Agreement (GLWQA) Integrated Atmospheric Deposition Network and in managing and assessing loadings of toxic air pollutants to the Great Lakes Basin.

Many Federal, State, and local government agencies, government agencies of other countries, and other organizations and independent researchers are involved in efforts to address the scientific questions related to atmospheric deposition to the Great Waters. Some of the groups involved are:

- U.S. EPA Offices (including the Office of Research and Development, EPA Program Offices, and EPA Regional Offices)
- U.S. National Oceanic and Atmospheric Administration
- Agency for Toxic Substances and Disease Registry
- Environment Canada
- International Joint Commission
- Great Lakes Commission
- Chesapeake Bay Research Consortium
- Vermont Monitoring Cooperative
- State agencies
- Local government agencies.

The four major activities listed in Table 1 are discussed in sequence below.

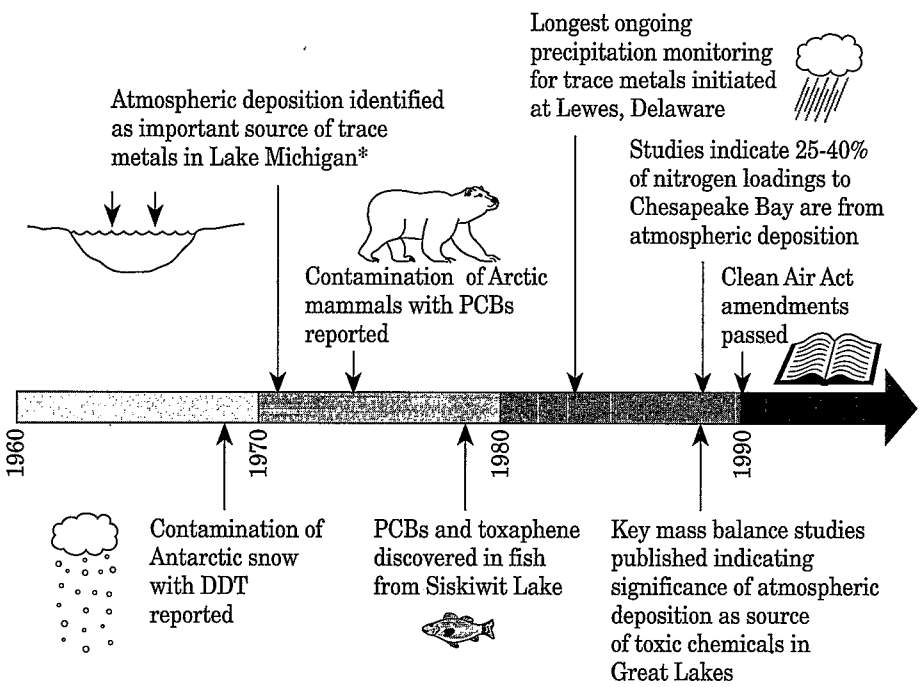
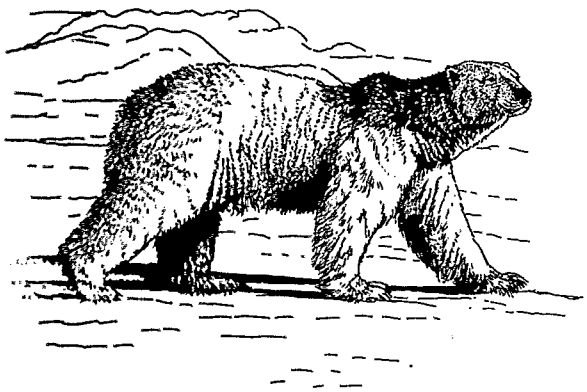


Figure 3. Significant milestones in understanding atmospheric deposition of toxic air pollutants to aquatic ecosystems.
* Reference 9, all other information from References 6-8.

■ **Analysis of pollutant exposure and effects in the Great Waters.** EPA, in cooperation with other Federal, State, and local agencies, has identified, and will continue to identify, air pollutants that are of possible concern for atmospheric deposition based on how long they persist in the environment, their ability to travel long distances when released from sources, their tendency to accumulate in animals and plants, and other factors. At the same time, EPA is evaluating whether these pollutants are associated with exposures to humans, animals, and plants and, subsequently, with human health and environmental effects. This first activity focuses on whether the pollutants of concern for atmospheric deposition can be linked to any environmental or public health impacts that appear to be significant enough to warrant action.



- **Analysis of how pollutants of concern actually get into the Great Waters.** EPA and others are evaluating how much of the toxic chemical pollution in the Great Waters comes from direct atmospheric deposition and how much comes from other routes, such as direct discharge, groundwater seepage, stormwater runoff, and inflow from connecting streams and rivers (these routes may also carry pollutants that are from indirect atmospheric deposition). If it is established that atmospheric deposition adds significantly to pollutant loadings of specific chemicals to the Great Waters (and therefore to the associated impacts identified in the first activity), actions to reduce atmospheric deposition of these chemicals may be warranted.

- **Identification and evaluation of air pollution sources that are contributing to pollutant loadings to the Great Waters and that could be targeted if reductions are needed.** EPA and NOAA are working to identify sources that emit pollutants of possible concern into the air and are determining which sources appear to add significantly to the deposition of air pollutants to the Great Waters.

Table 1. Major Activities and Questions Addressed by the Great Waters Program

Scientific Questions	Regulatory Questions	Major Activities
What do we know about atmospheric deposition to the Great Waters?	What action is needed to reduce atmospheric deposition to the Great Waters?	
What human health and environmental effects are associated with pollutants of concern in the Great Waters?	Are impacts or risks significant enough to be of concern?	Analyze pollutant exposure and effects in waterbodies
What is the relative importance of atmospheric deposition in causing contamination in the Great Waters?	Are loadings from the air significant enough to need reduction?	Evaluate pollutant loadings to waterbodies
Where and what are sources of air emissions of pollutants of concern? What sources are significant contributors to the Great Waters?	If reductions are needed, what emissions sources should be targeted?	Identify and evaluate air emission sources
Would emission reductions be effective in reducing effects of atmospheric deposition to the Great Waters?	What are the options for implementing reductions? What are the costs and benefits of the various options?	Identify and evaluate emission reduction options

- **Determination, based on the information developed from the three preceding activities, of whether further emission reductions, beyond those expected as a result of implementing section 112 of the 1990 Amendments, may be needed.** EPA will identify and evaluate any needed emission reduction options and, as appropriate, will make recommendations for regulatory action under the Clean Air Act or other Federal laws.

Chapter 3 of this report presents the information obtained, to date, for each of the first three activities, including information on pollutant exposures and effects, relative loadings, and sources. Much is known as a result of research conducted to date, as evidenced by this report. EPA will continue to analyze data as they become available. Future reports to Congress will update the information presented in this report, with greater detail and more certainty as research provides additional data on the subject of atmospheric deposition of pollutants to the Great Waters.

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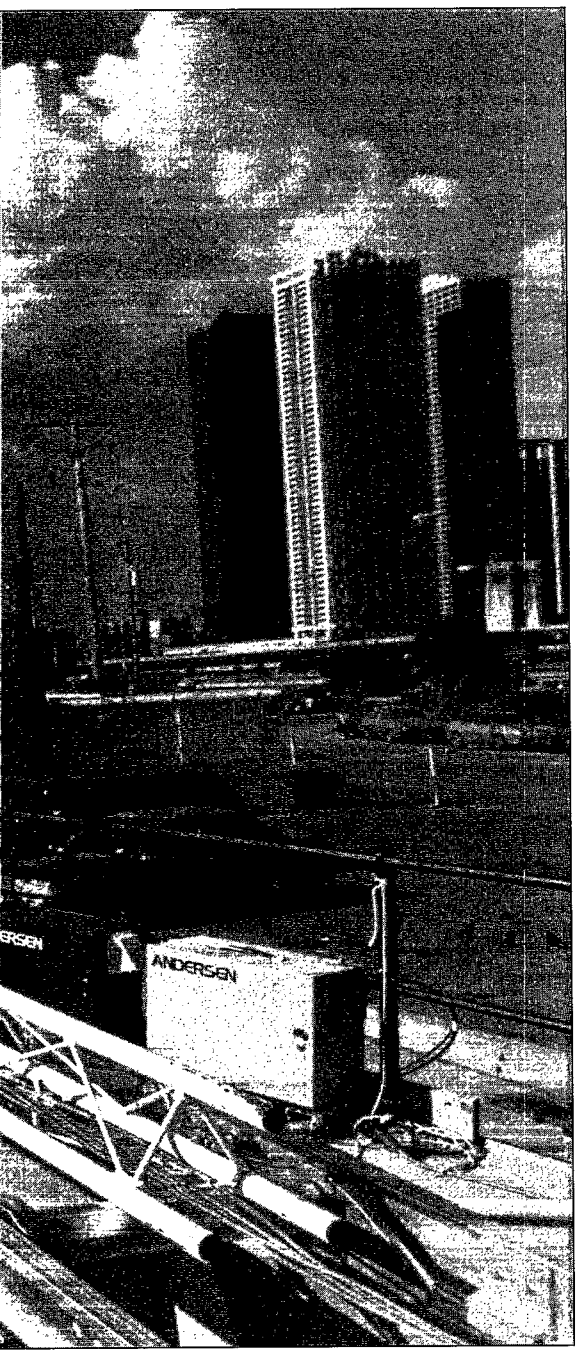
Chapter Three
Answering the Scientific Questions of Section 112(m)



What is known about the extent and significance of atmospheric deposition to the Great Waters? To answer this central question and to address the issues raised in section 112(m) of the Clean Air Act, as amended in 1990 (1990 Amendments), this chapter considers three scientific questions (see sidebar next page). The first section addresses the question of exposure and effects to evaluate the human health and environmental effects associated with exposure to pollutants in the Great Waters. This section identifies pollutants of concern, describes pathways by which humans, animals, and plants may be exposed to these Great Waters pollutants, summarizes information on pollutant levels in the Great Waters and related adverse effects, and presents several case studies. The second section addresses the question of relative loading to evaluate how much of the pollution in the Great Waters comes from atmospheric deposition. This section summarizes the current understanding of atmospheric deposition processes and presents the results of mass balance case studies for selected chemicals and waterbodies. The third section addresses the question of sources to evaluate the origins of the toxic air pollutants being deposited to the Great Waters. This section summarizes what is known about various sources that emit pollutants of concern and provides several case studies as examples.

In each of these three areas, it is difficult to establish, in a rigorous scientific manner, definitive cause and effect relationships. In other words, it is very unlikely that one will find evidence proving that atmospheric deposition of a specific pollutant from a specific source caused a specific effect in a specific waterbody. In fact, a major challenge is proving cause and effect for any one of these links, much less for the entire chain of events. Reasons for this difficulty include: (1) the very large number of factors that could contribute to causing an observed effect and the challenges of determining the contribution of each; (2) the inability to conduct fully controlled experiments in real-world field situations; (3) the inability to apply the findings of controlled laboratory experiments, with full confidence, to real-world conditions; and (4) the logistical difficulty, time, and expense required to collect the kinds of data needed to identify cause and effect relationships. Scientists do, however, employ a number of approaches to investigate cause and effect relationships. These approaches include: (1) establishing correlations and associations among factors, (2) evaluating trends in field data over long time periods, (3) analyzing for consistency between field observations and controlled laboratory data, and (4) confirming

preliminary findings in different settings by different investigators. Using these and other tools, scientists frequently can accumulate enough information to determine the likely causes of a given effect.

Effects: What Human Health and Environmental Effects Are Associated with Exposure to Great Waters Pollutants of Concern?

In assessing exposure and effects, consideration must be given to both human health and environmental effects (and the exposures that cause both types of effects). Both types of effects are important in their own right, and, in many cases, ecological effects are early indicators of human health effects. For example, pollutants in water that accumulate in the tissues of fish may result in direct effects in fish-eating birds, such as decreased populations. These ecological effects, in turn, may be indicators of potential human health effects related to the consumption of contaminated fish. In a widely circulated 1990 report, *Reducing Risk: Setting Priorities and Strategies for Environmental Protection*, EPA's Science Advisory Board strongly emphasized the very close link between human health and ecological health and pointed out that "most human activities that pose significant ecological risks . . . pose direct or indirect human health risks as well."[12]

The mandates of section 112(m) of the 1990 Amendments require EPA to assess the environmental and public health effects caused by water pollution attributable to atmospheric deposition to the Great Waters and to determine whether pollutant loadings to the Great Waters cause or contribute to exceedances of drinking water or water quality standards (including, for the Great Lakes, violations of the specific objectives of the Great Lakes Water Quality Agreement). Although a large number of pollutants are potentially of concern for atmospheric deposition, this report focuses on only 15 pollutants. Table 2 lists the 15 pollutants addressed in this report, along with examples of their uses (and use restrictions) in the United States. Thirteen of these pollutants are on the 1990 Amendments list of air pollutants; dieldrin and nitrogen are not on the list. All 15 are known air pollutants in the vicinity of at least some of the Great Waters, and all are known to be present in atmospheric deposition (e.g., rainfall). Data indicate that they are present in the Great Waters and that atmospheric deposition is a pathway by which they reach the waterbodies. All of the pollutants, with the exception of nitrogen, are of concern because of their persistence in the environment (length of time a pollutant remains in the environment), potential to bioaccumulate (potential to accumulate in living organisms), and toxicity to humans and the environment. The range of potential effects associated with exposure to these pollutants (except for nitrogen) includes cancer, effects to the reproductive system, developmental effects (i.e., effects on the developing human, including effects on embryos, fetuses,

Scientific Questions

- **Effects**
  What human health and environmental effects are associated with pollutants of concern in the Great Waters?
- **Relative Loading**
  What is the relative importance of atmospheric deposition in causing contamination in the Great Waters?
- **Sources**
  What sources are significant contributors to atmospheric loadings to the Great Waters?

Regulatory Question

- **Regulatory**
  Is action warranted to reduce atmospheric deposition?

Table 2. Selected Pollutants of Concern in the Great Waters[a]

Pollutant	Examples of Uses[b]
Cadmium and compounds	Naturally occurring element used in metals production processes, batteries, and solder. Often released during combustion of fossil fuels and waste oil and during mining and smelting operations.
Chlordane	Insecticide used widely in the 1970s and 1980s. All U.S. uses except termite control canceled in 1978; use for termite control voluntarily suspended in 1988. Use of existing stocks permitted.
DDT/DDE	Insecticide used widely from introduction in 1946 until significantly restricted in U.S. in 1972. Still used in other countries. Used in U.S. for agriculture and public health purposes only with special permits.
Dieldrin	Insecticide used widely after introduction in late 1940s. Used in U.S. for termite control from 1972 until registration voluntarily suspended in 1987.
Hexachlorobenzene	Fungicide used as seed protectant until 1985. Byproduct of chlorinated compound and pesticide manufacturing. Also a byproduct of combustion of chlorine-containing materials. Present as a contaminant in some pesticides.
α-Hexachlorocyclohexane (α-HCH)	Component of technical-HCH, an insecticide for which use is restricted in U.S., but used widely in other countries.
Lindane (γ-Hexachlorocyclohexane) (γ-HCH)	Main component of lindane, an insecticide used on food crops and forests, and to control lice and scabies in livestock and humans. Currently used primarily in China, India, and Mexico. U.S. production stopped in 1977. Use was restricted in 1983; however, many uses are still registered, but are expected to be voluntarily canceled in the future.
Lead and compounds	Naturally occurring element commonly used in gasoline and paint additives, storage batteries, solder, and ammunition. Released from many combustion and manufacturing processes and from motor vehicles. Use in paint additives restricted in U.S. in 1971. U.S. restrictions on use in gasoline additives began in 1973 and have continued through the present, with a major use reduction in the mid-1980s.
Mercury and compounds	Naturally occurring element often used in thermometers, electrical equipment (such as batteries and switching equipment), and industrial control instruments. Released from many combustion, manufacturing, and natural processes. Banned as paint additive in U.S., for interior paint (1990) and for exterior paint (1991).
Polychlorinated biphenyls (PCBs)	Industrial chemicals used widely in the U.S. from 1929 until 1978 for many purposes, such as coolants and lubricants and in electrical equipment (e.g., transformers and capacitors). In the U.S., manufacture stopped in 1977 and uses were significantly restricted in 1979. Still used for some purposes because of stability and heat resistance, and still present in certain electrical equipment used throughout U.S.
Polycyclic organic matter (POM)[c]	Naturally occurring substances that are byproducts of the incomplete combustion of fossil fuels and plant and animal biomass (e.g., forest fires). Also, byproducts from steel and coke production and waste incineration.
2,3,7,8-Tetrachlorodibenzofuran (2,3,7,8-TCDF)	Byproduct of combustion of organic material containing chlorine and of chlorine bleaching in pulp and paper manufacturing. Also a contaminant in some pesticides.
2,3,7,8-Tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD)	Byproduct of combustion of organic material containing chlorine and of chlorine bleaching in pulp and paper manufacturing. Also a contaminant in some pesticides.
Toxaphene	Insecticide used widely on cotton in the southern U.S. until the late 1970s. Most U.S. uses banned in 1982; remaining uses canceled in 1987.
Nitrogen compounds	Byproducts of combustion processes and motor vehicles. Also, compounds used in fertilizers.

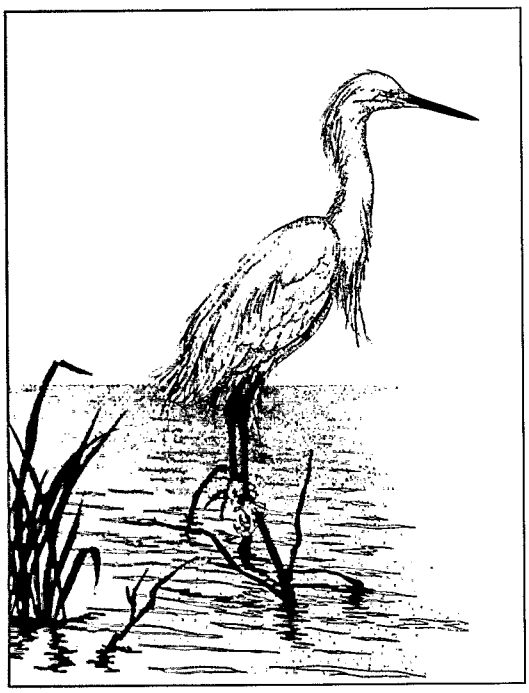
[a]Data for this table are taken from References 13 through 27.
[b]Applicable restrictions (including bans) on use or manufacture in the United States also are described.
[c]POM is a large class of chemicals consisting of organic compounds having multiple benzene rings and a boiling point greater than 100 °C. Polycyclic aromatic hydrocarbons (PAHs) are a chemical class that is a subset of POM.

and children), neurological effects (i.e., effects on the brain and nervous system), effects on the endocrine system (i.e., effects on hormone production and function), and other noncancer effects (e.g., liver or kidney damage). The potential for effects will depend on the level and duration of exposure and the sensitivity of the exposed organism.

Furthermore, although some differences exist, the pollutants in Table 2 overlap substantially with several sets of Great Lakes chemicals of concern selected by other scientific and regulatory groups, and they also are generally consistent with the toxic air pollutants that ranked the highest in a 1991 EPA study to identify priority chemicals for the Great Waters Program.[28] In addition, all of the pollutants in Table 2, except 2,3,7,8-TCDF and nitrogen compounds, are included on the list of pollutants that are the initial focus of the EPA/State Great Lakes Water Quality Initiative, and 10 of the 15 Great Waters pollutants of concern are designated as chemicals of concern that have the potential to bioaccumulate (the highest priority group).[29]

These pollutants, excluding nitrogen, are also of concern based on the priorities set by the Great Lakes Water Quality Board (GLWQB) of the International Joint Commission, which is an advisory committee comprised of representatives from the United States and Canada. In addition, 5 of the 15 pollutants (cadmium, benzo[*a*]pyrene [indicator for polycyclic organic matter (POM)], lead, mercury, and PCBs) are on the Chesapeake Bay Toxics of Concern List, and two (dieldrin and toxaphene) are on the list of potential substances to be added to the Chesapeake Bay list.

Nitrogen compounds were added to the list of pollutants considered in this report because of nitrogen's role in nutrient enrichment in coastal waters and because data indicate that atmospheric loadings of nitrogen to Chesapeake Bay are significant. Accelerated eutrophication, which results from excessive loadings of nitrogen, can cause ecological effects such as reduced fish and shellfish populations.



The first 14 pollutants in Table 2 represent air pollutants of priority concern for the Great Lakes. Because of the potential for these 14 pollutants to cause harm in the Great Lakes, it is likely that they have the potential to cause harm in other fresh water systems as a result of their tendency to bioaccumulate in living organisms, to persist in the environment, and to be toxic to humans and ecosystems. However, the pollutants listed in Table 2 are not inclusive of all chemicals that may, now or in the future, be an important component of atmospheric deposition to the Great Lakes or other Great Waters.

Other pollutants are of potential concern for the effects that they may cause after being deposited to the Great Waters. In the proposed Water Quality Guidance for the Great Lakes Systems, 28 "bioaccumulative chemicals of concern" (BCCs), many of which are air pollutants, are identified. A BCC is defined as "any chemical which, upon entering the surface waters, by itself or as its toxic transformation product, bioaccumulates in aquatic organisms by a human health

bioaccumulation factor greater than 1000, after considering metabolism and other physicochemical properties that might enhance or inhibit bioaccumulation, . . . "[29] The guidance proposes that additional controls be established for these BCCs to obtain reductions in loadings and to ensure that new problems do not develop in the future with pollutants in the Great Lakes ecosystem that show a propensity to bioaccumulate and to persist in the environment.[29] Ten of the fifteen Great Waters pollutants of concern appear on this list, and one other appears on the list of potential bioaccumulative chemicals. These two lists are provided in Appendix A. Future reports to Congress on atmospheric deposition to the Great Waters may include many of these additional chemicals.

The remainder of this section describes exposure and associated effects for the selected pollutants of concern. Exposure can be thought of as the contact between a chemical and a living organism. Because atmospheric deposition is a significant source of pollutants for some Great Waters, it is reasonable to hypothesize that atmospheric deposition is contributing, to some extent, to exposure and effects occurring in the Great Waters. Moreover, some studies have found correlations between atmospheric deposition of pollutants and subsequent exposure and effects in the Great Waters. Few studies, however, have directly and strongly linked atmospheric deposition of pollutants to exposure and, subsequently, to effects observed in the Great Waters. Because of the data limitations and the overall difficulty in documenting cause and effect relationships, this section generally does not attempt to attribute specific effects to atmospheric deposition. Instead, it describes effects of selected pollutants of concern known to be present in atmospheric deposition. This section is further limited in its scope because much of the available information is from the Great Lakes and Chesapeake Bay, since relatively little research has been devoted to other Great Waters.

Current Understanding of Exposure

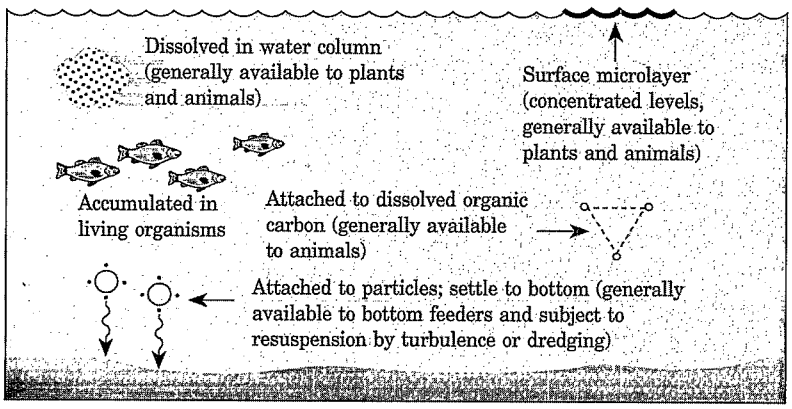


Figure 4. Distribution of pollutants within a waterbody.

Figure 4 illustrates, in a simplified form, the distribution of pollutants in a waterbody. Once in a waterbody, pollutants will bind to the surface of particles or dissolved organic material, concentrate at the surface of the water, or dissolve and remain in solution. Most of the selected pollutants of concern tend to bind to small particles suspended in water. Over time, pollutants associated with particles tend to deposit to, and accumulate in, sediments. In some cases, pollutants in the sediments may not be available for chemical degradation in the water. However, contaminated sediments can serve as a major reservoir of pollutants that continually recycles

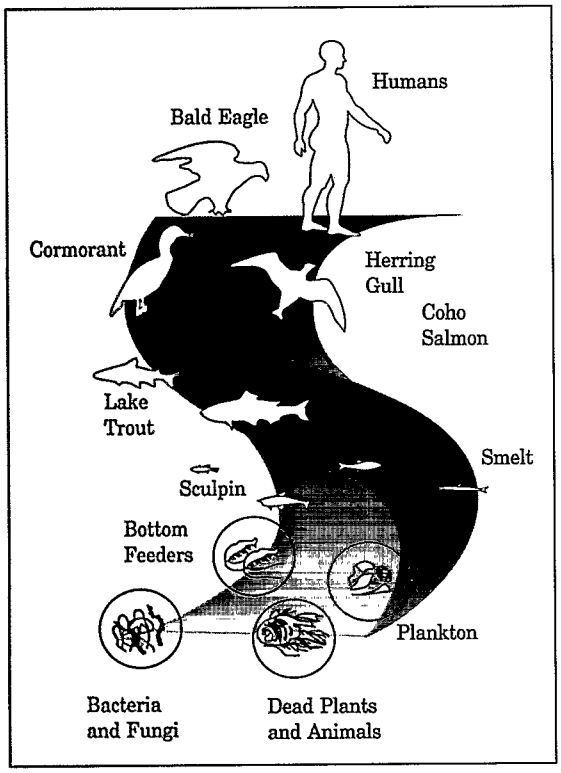


Figure 5. Simplified overview of a food web in the Great Lakes.

the pollutants back into the ecosystem. Pollutants thus are available for uptake by bottom-dwelling organisms and bottom-feeding fish. Binding to dissolved organic material may also affect the pollutants' availability for uptake. Many pollutants tend to concentrate in the surface microlayer, which is the uppermost layer of water surface (approximately 50 μm in depth). The surface microlayer is enriched with nutrients and is an important feeding area for many microscopic plants and animals. It also is the site for the transfer of chemicals between air and water. Pollutants dissolved in the water tend to be readily available for uptake by plants and animals. The uptake of dissolved pollutants may be the most important means by which many plants and animals are exposed.

For animals and plants, the possible exposure routes for toxic pollutants present in waterbodies are intake of food, intake of drinking water, and direct contact with the water. For fish-eating birds and mammals, intake of food is the main exposure route of concern for pollutants that are persistent in the environment and that have the tendency to bioaccumulate.

Bioaccumulation is the uptake and retention of a chemical by a living organism as a result of either intake of food, intake of drinking water, direct contact, or intake of air. Biomagnification refers to the phenomenon in which chemicals become more concentrated in animals at higher levels in the food web. The selected pollutants for this report tend to accumulate in fatty tissue, and, as a result of food web interactions, the highest pollutant concentrations are found in animals at the top of the food web. Figure 5 provides a simplified overview of a food web in the Great Lakes. In this example, organisms near the top of the food web, such as humans and bald eagles, would tend to have higher body concentrations of chemicals that biomagnify than organisms lower in the food web.

The exposure route of most concern for human health is intake of food. Intake of drinking water is another exposure route; yet, for pollutants with the capacity to bioaccumulate, this is typically not a significant exposure route of concern.

Estimates of exposure levels can be made using data on pollutant concentrations in various parts of the ecosystem. Researchers have investigated surface water, sediment, and fish tissue concentrations of toxic chemicals in the Great Lakes and Chesapeake Bay, to determine the pattern and extent of contamination. In the Great Lakes, they found that the influence of anthropogenic sources since the 1940s has resulted in significant increases in the levels of many persistent toxic chemicals. However, there were few reliable data on toxic chemical concentrations in water in the Great Lakes until 1980 or later.[30] As a result, it is very difficult to draw an accurate picture of time trends in water concentration data. Recent research, however, has shown that most pollutants of concern usually are found in water samples at very low levels, although

even at low levels some pollutants may cause significant effects in plants and animals in the Great Waters.

In general, pollutant levels in sediments have decreased, relative to the 1970s, and the bulk of toxic pollutant influx into the Great Lakes ecosystem occurred in the 1960s and 1970s.[30] However, sediment concentrations are still particularly elevated in sediment basins, harbors, and delta regions (near the mouth of the river), indicating that runoff and industrial discharge may be the source of some pollutants.[31] Compared with water concentrations, sediment concentrations of pollutants of concern are considerably higher, reflecting the tendency of these pollutants to attach to particles and settle to the sediments.[30] Sediment processes such as resuspension and resolubilization can reintroduce significant pollutant loads.

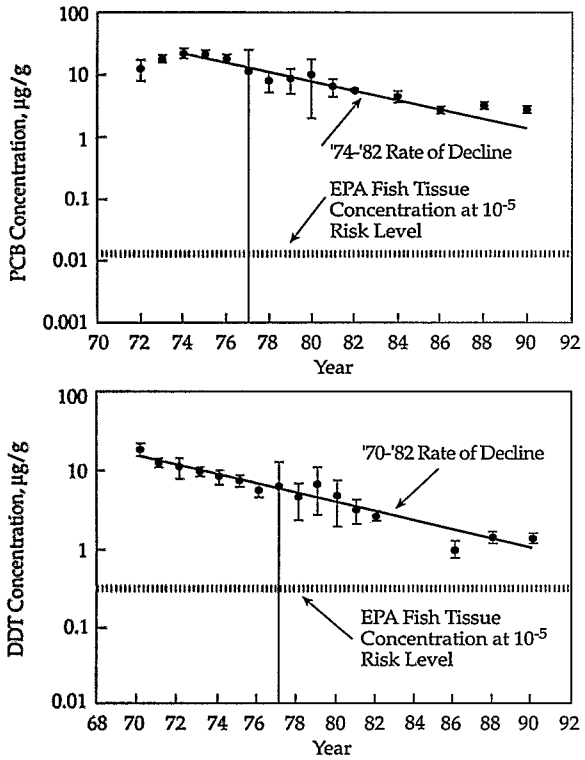
As described earlier, pollutants in sediments and water can be transferred to fish and other aquatic animals by direct contact and by intake of food and water and can be concentrated in the animal by the process of bioaccumulation. Numerous studies have documented cases of elevated levels of persistent toxic pollutants in various fish species compared to levels in water and, in many cases, compared also to levels in sediment, reflecting the tendency of these pollutants to bioaccumulate.

Because many Great Waters bird and mammal species rely on fish and shellfish as a primary food source, bioaccumulation and biomagnification of these pollutants is a problem for wildlife. In fact, predators such as the herring gull, bald eagle, and turtle in the Great Lakes region have some of the highest reported tissue concentrations of persistent toxic chemicals. The measurement of persistent toxic chemicals in herring gull eggs has been used as an indicator of pollutant levels in the Great Lakes ecosystem.[30] One study suggested that levels of toxic pollutants in herring gull eggs have decreased from the 1970s to the early 1980s.[30] Another study shows a general decline in levels of DDE and PCBs during the early and mid-1980s, with an increase for PCBs in 1989. DDE levels have gradually ebbed, without recent rises.[32] Data on concentration levels in Great Lakes mammals are scarce because most research has centered on birds.

The presence of the same pollutants in the tissue of humans and other fish-eating animals as those identified in water, sediment, and fish indicates that biomagnification through the food web has the potential to be a significant exposure concern for humans. Studies indicate that people who regularly consumed fish from Lake Michigan in the 1970s had significantly higher concentrations of PCBs and pesticides, such as DDT, in their bodies compared with those who did not consume fish.[8]

The limited human tissue residue data available indicate that the general population residing in the Great Lakes basin is probably not exposed to higher levels of the most persistent pollutants than people residing elsewhere in North America. However, individuals (e.g., native peoples, sports anglers) who consume large amounts of contaminated fish and wildlife have greater exposure to persistent pollutants than the

Total PCB and DDT Concentrations in Lake Michigan Lake Trout



Source: Reference 29.

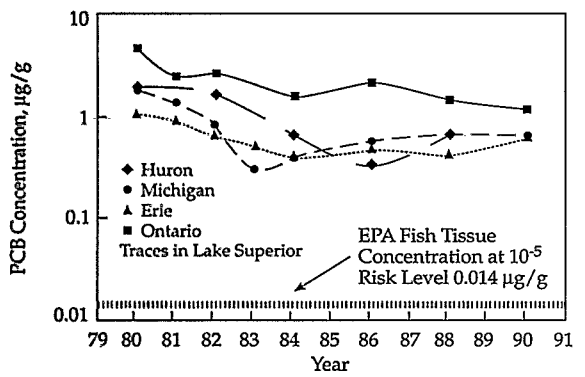
Trends in Total PCB and DDT Concentrations in Great Lakes Fish

Concentrations of PCBs and DDT in Lake Michigan lake trout have declined markedly since the latter half of the 1970s, reflecting the relatively rapid response of the water column to decreases in pollutant loadings. Beyond 1982, however, concentrations in lake trout have been higher than predicted levels. Although concentrations are still declining, the rate of decline is slowing and may be leveling off, although at concentrations well above water quality criteria.

These substances appear to be approaching equilibrium in the Great Lakes system at unacceptably high levels due to continuing loadings from a variety of sources, such as: (1) historically contaminated sediments; (2) tributary inputs resulting from point sources, spills and runoff from both urban and rural areas, and resuspension from contaminated sediments; and (3) atmospheric deposition of pollutants. In 1990, concentrations of PCBs and chlorinated pesticides measured in fish tissue exceeded the fish tissue concentrations that correspond to current EPA Clean Water Act 304(a) water quality criteria by several orders of magnitude.* If a new equilibrium is being reached given current mass loadings, then substantial reductions to the Great Lakes system will be necessary to eliminate fish advisories.[29]

The slowing in the rate of decline of PCBs in fish tissue is also supported by coho salmon data. Because coho are stocked and are in the lake for only 18 months, they respond much more quickly to changes in water column concentrations than lake trout, which have an average life span of 6 years and can, therefore, accumulate greater PCB concentrations. After significant declines between 1980 and 1984, PCB concentrations in coho salmon have been relatively constant in all the Great Lakes since the mid-1980s, with the exception of a general decline in Lake Ontario.

PCB Concentrations in Coho Salmon

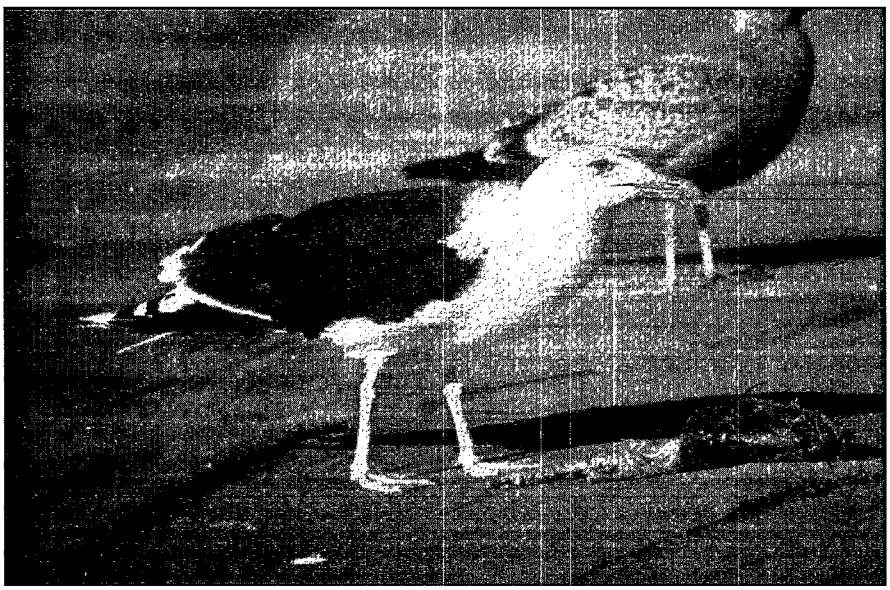


Source: Reference 29.

*Section 304(a) of the Clean Water Act establishes numeric water quality criteria for the protection of the health and welfare of (including, but not limited to) plankton, fish, shellfish, wildlife, plant life, shorelines, beaches, aesthetics, and recreation, which may be adversely impacted due to the presence of pollutants.

general population.[32] Studies of these sensitive subpopulations are currently being performed by the Agency for Toxic Substances and Disease Registry of the Department of Health and Human Services.

Given the elevated levels of the selected pollutants of concern in water, sediments, fish and wildlife, and humans, concern for human exposure is warranted. One means of limiting human exposure is the establishment of fishing or fish consumption advisories or restrictions.



James F. Parnell

A fishing advisory or fish consumption advisory is issued when fish taken from a particular body of water are found to contain levels of contaminants that exceed recommended intake, or threshold, levels. The majority of the advisories are recommendations to the general public about the dangers of fish consumption. Advisories to regulate commercial fishing are enforceable by health departments and are often referred to as "fishing restrictions." Tables 3 and 4 summarize current fish consumption advisories and fishing advisories for some of the Great Waters. As shown in Table 3, portions of all of the Great Lakes and many associated waterbodies have had or do have some type of advisory on fish consumption. As shown in Table 4, fish consumption and fishing advisories have also been issued in Chesapeake Bay and Lake Champlain. The elevated tissue levels in fish relative to water concentrations, which have resulted in restrictions in fish consumption, emphasize the importance of bioaccumulation, and subsequent biomagnification in the food web, when considering potential for human exposure.

Summary of Current Understanding of Exposure

1. **What Are the Major Routes of Exposure to Pollutants Derived from Atmospheric Deposition?**

   For water pollutants that are derived from atmospheric deposition, the major routes of exposure are fairly well understood. For animals, routes of exposure include intake of food (especially significant because of biomagnification in the food web), intake of drinking water, and direct contact. Routes of exposure for plants include water uptake and direct contact. For humans and for fish-eating mammals and birds, the primary route of exposure is intake of food.

   Although routes of exposure have been identified, the amounts of toxic pollutants to which humans, animals, and plants are exposed are not easily determined given the currently available data.

Table 3. Current Great Lakes Fish Consumption Advisories[a]

Waterbody (including tributaries)	Pollutant	Restrictions[b]	Do Not Eat
Lake Superior	PCBs, chlordane, and mercury	Lake trout, chinook salmon, and walleye	Lake trout over 30", walleye over 26", catfish, northern pike, and white sucker
Lake Michigan	PCBs, chlordane, DDT, dieldrin, and mercury	Lake trout, coho salmon, chinook salmon, brown trout, and walleye	Lake trout over 23", chinook salmon over 32", brown trout over 22", carp, and catfish
Green Bay	PCBs and pesticides	Splake	Lake trout, brook trout, rainbow trout, chinook salmon, brown trout, splake over 16", northern pike, walleye, white bass, and carp
Lake Huron	PCBs	Brown trout, lake trout, and rainbow trout	Brown trout over 21" and rainbow trout over 21"
Saginaw Bay	PCBs and dioxins	Rainbow trout and brown trout	Carp and catfish
Lake Erie	PCBs and chlordane	——	Carp and catfish
Lake Ontario	PCBs, dioxins, and chlordane	——	American eel, catfish, lake trout, chinook salmon, coho salmon, rainbow trout, brown trout
St. Mary's River	Mercury	Walleye	——
St. Clair River	PCBs and mercury	Freshwater drum and gizzard shad	Carp
Lake St. Clair	PCBs and mercury	Walleye, white bass, smallmouth bass, white perch, carp, rock bass, largemouth bass, bluegill, freshwater drum, carpsucker, catfish, and northern pike	Muskie, sturgeon, and catfish over 22"
Detroit River	PCBs and mercury	Freshwater drum	Carp
Niagara River	PCBs and dioxins	Carp and smallmouth bass	Channel catfish, American eel, lake trout, chinook salmon, rainbow trout, coho salmon, and brown trout
St. Lawrence River	PCBs	All fish	Channel catfish, American eel, chinook salmon, brown trout, lake trout, coho salmon over 21", and rainbow trout over 25"

[a]Data for this table are taken from Reference 13.
[b]Restrictions: Nursing mothers, pregnant women, women who anticipate bearing children, female children of any age, and male children age 15 or under should not eat fish taken in these locations. Other persons should limit their consumption to one meal per week and follow preparation and cooking recommendations.

Table 4. Current Fishing Advisories in Selected Great Waters[a]

Waterbody	Advisories
Chesapeake Bay (Maryland)	**Chlordane:** Black crappie and carp from Lake Roland; channel catfish and American eel from Baltimore Harbor and Back River
Chesapeake Bay (District of Columbia)	**Chlordane:** Potomac River **PCBs:** Potomac River
Lake Champlain	**Vermont and New York** **PCBs:** Lake trout over 25 in. **Mercury:** Walleye over 19 in. **Cumberland Bay only** **PCBs:** American eel, Brown bullhead

[a]Data for this table are taken from References 23 through 39. Includes information for the selected pollutants of concern only.

2. To What Extent Does Atmospheric Deposition Contribute to Overall Exposure to Toxic Chemicals in the Great Waters?

Current understanding of the extent to which atmospheric deposition contributes to overall exposure is limited because overall exposure to toxic water pollutants has not been fully quantified and because complete and accurate information on all pollutant inputs and outputs is not available. For example, the presence of pesticides, such as toxaphene, that have never been used extensively in the Great Lakes region can be attributed primarily to atmospheric deposition. Similarly, reductions in lead concentrations in fresh water fish have been attributed to decreases in lead emissions from motor vehicles, suggesting the importance of atmospheric deposition for exposure.[40] On the other hand, many pollutants such as 2,3,7,8-TCDD are derived from several sources, and determining how much comes from atmospheric deposition alone is difficult.

Pat Cunningham, RTI

Current Understanding of Effects

Much of the ecological effects information presented in this section is based on observed effects from field studies in which ecological effects data were correlated with pollutants present in the environment. This information generally is supported by laboratory study data, as detailed in the technical contractor report.[8] Because of the limited observed effects data for humans and the difficulties in obtaining such data, the human health effects information presented also refers to data on effects in animals that are suggestive of potential human health effects.

Environmental Justice: Great Lakes Fish Contamination

Numerous early studies have examined the relationship between the consumption of fish from the Great Lakes and observable health effects in certain subpopulations. Subpopulations identified as especially vulnerable to exposure to persistent toxic substances in the Great Lakes include: pregnant and nursing females (who are more vulnerable to effects), sport anglers, Native Americans, and the urban poor (who have high fish consumption for reasons of economic need or cultural tradition).[41] This makes environmental justice an important issue.

Examples of study results:

- In 1990, a study entitled "Fish Consumption Patterns and Blood Mercury Levels in Wisconsin Chippewa Indians" was conducted by the Center for Disease Control to investigate blood mercury levels among Wisconsin's Ojibwa population. Of the 357 adults (from five Ojibwa tribes) tested, 64 were found to have blood mercury levels in excess of 5 µg/L. Since a report from the Institute of Medicine suggests that delayed development of infants may occur following in utero exposure to maternal blood mercury levels of 5 to 10 µg/L, the results of the Wisconsin Chippewa study warrant concerns for human health.[41]

Jimmy W. Crawford, KY

- A 1989 survey of Michigan sport anglers showed a tendency for fish consumption to increase with age, for minority ethnic groups to consume more fish than whites, and for fish consumption to diminish as the educational level of a household increases.[42]

- A 1992 survey of 300 Detroit riverbank anglers showed that 32% of those who ate the fish were children between the ages of 5 and 18 years, and 26% were women of childbearing age. Minorities made up 94% of those fishing the Detroit River within the city limits, with 90% being African American. Almost 35% of the anglers indicated that they did not feel adequately informed about the risk associated with eating fish from the Detroit River.[43]

Cuyahoga Bay, OEPA

It should be noted that there are concerns related to methodology that can be raised with respect to many, if not all, of these studies. These concerns led the EPA to create a Fish Contamination work group, which is writing technical guidance designed to improve the quality of such surveys.[42] In addition, the Department of Health and Human Services, Agency for Toxic Substances and Disease Registry, is in the process of completing a report summarizing the "Impact on Public Health of Persistent Toxic Substances in the Great Lakes Region," with a review of available studies on consumption of contaminated fish.[41]

Although concerns over methodology exist, results of studies suggest that certain subpopulations will be more likely to consume greater amounts of Great Lakes fish and, therefore, be more exposed to toxic chemicals and their effects. These issues need to be considered in decisionmaking on toxic substance control.

Ecological Effects

Ecological effects associated with pollutants known to be present in atmospheric deposition are evident in numerous studies describing birth defects, reproductive failure, disease, and premature death in fish and wildlife species native to the Great Lakes.[8]

In general, ecological effects of exposure to toxic pollutants can occur at both the individual level and the ecosystem level. Effects at the individual level include both cancer and noncancer effects. There is a broad spectrum of noncancer effects, including changes in enzyme functioning and effects on the endocrine, immune, nervous, and reproductive systems. Effects at the ecosystem level may include changes in populations (e.g., reproduction rates) and communities (e.g., species diversity). Another effect, eutrophication, to which atmospheric deposition can contribute, can produce both individual- and ecosystem-level effects.

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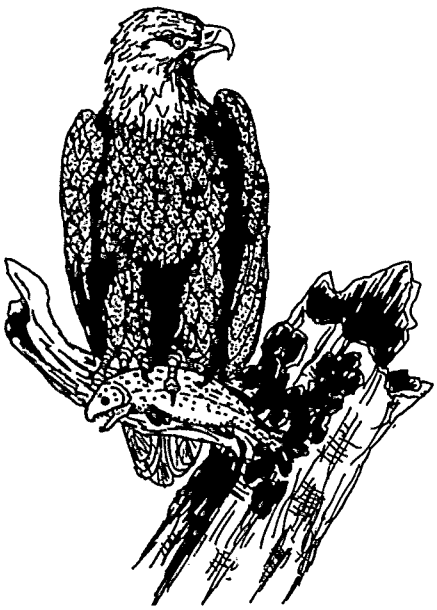
Ecological effects associated with pollutants of concern range from short-term, chemical-specific effects (e.g., fish disease, wildlife disease, effects on reproduction) to gradual, cumulative effects (e.g., population declines, community changes). Effects on the reproductive system can have negative impacts both on an individual's reproductive success and on the ecosystem by reducing a population's rate of reproduction. In addition, most pollutants of concern bioaccumulate to high levels in fish and fish-eating wildlife. At these higher exposure levels, fish and wildlife are more likely to suffer various cancer and noncancer effects. The remainder of this section briefly discusses some of the important effects of the pollutants of concern on aquatic organisms and other wildlife.

Effects on Aquatic Organisms and Other Wildlife. Several of the selected pollutants of concern cause changes in enzyme functioning. Studies have reported that the activity of enzymes responsible for the breakdown of foreign compounds is greatly increased by most of the chemicals of concern. In fish, the increased activity of these enzymes has been shown to result from exposure to PCBs and PAHs (PAHs are a subset of POM). In birds, "wasting" syndrome (i.e., the condition in which an animal slowly loses body weight until it can no longer sustain itself) has been related to altered enzyme activity resulting from exposure to environmental pollutants.

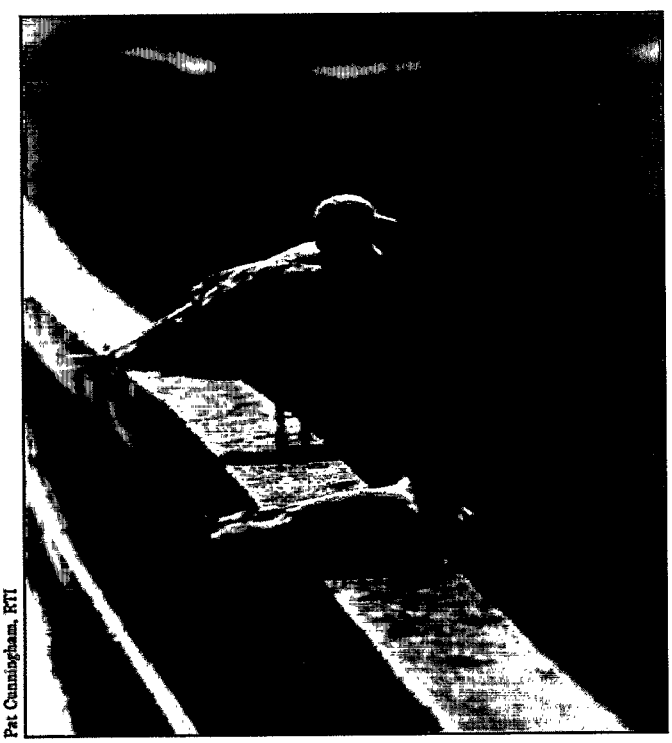
Effects on system functioning are reflected in findings of deficiencies in the immune system of beluga whales during a long-term study in the St. Lawrence River (located in the Great Lakes basin). This study indicated that these populations of beluga whales have significantly higher tissue concentrations of PCBs, DDT, and other toxic chemicals



than other marine mammal populations. Researchers attributed the generally poor health of the St. Lawrence beluga whales to suppressed immune system activity resulting from exposure to environmental chemicals. Other studies in the Great Lakes region also have found associations between PCBs and DDT and decreased immune system function. In the Chesapeake Bay region, diminished immune response was demonstrated in bottom-dwelling fish of the Elizabeth River exposed to sediment contaminated with PAHs.

Particular concern is warranted for humans and other animals because of the effects these pollutants have on other body systems such as the nervous system (including behavioral effects) and endocrine system. Recent data indicate that effects to these systems may occur at very low exposure levels. For example, populations of Great Lakes herring gulls, Forster's terns, and ring-billed gulls have exhibited behavioral changes such as female-female pairings, which result in abnormal incubation activities and nesting behavior, including nest abandonment. Exposures to pollutants of concern have resulted in effects on the endocrine system such as thyroid disorders, loss of reproductive functions in certain species, deficiencies in hormones such as insulin, and changes in reproductive success related to hormone function.

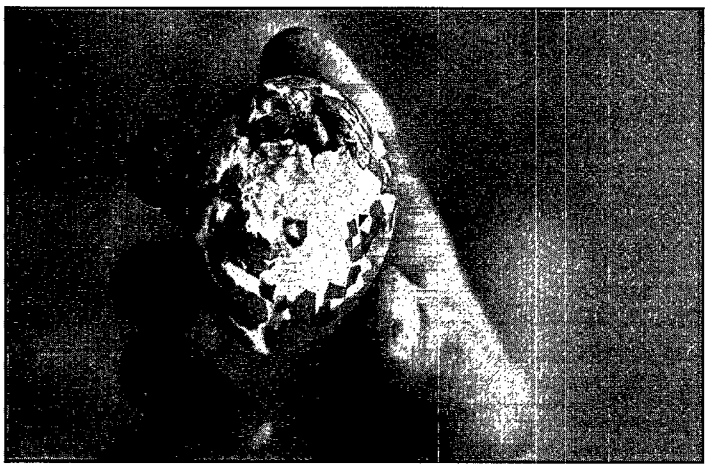
Effects on the overall health of individual aquatic organisms are reflected in reports of skin and liver cancers in fish and beluga whales. In some cases, these cancers have been attributed to concentrations of PAHs. In one study of stranded beluga whales in the St. Lawrence River, tumors were discovered in 40 percent of the whales examined. In another study in the Great Lakes, bottom-feeding fish such as bullhead were found to have increased tumor occurrence and a broad variety of tumors. These tumors were linked to exposure to PAHs.



Pat Cunningham, RTI

Effects on Great Waters ecosystems are evident in changes in fish communities present in the Great Lakes and Chesapeake Bay and population declines in many fish species. Another indicator of ecosystem effects is the drastic change in bottom-dwelling communities in the Great Lakes.[38] Exposure of these communities to toxic chemicals has resulted in significant changes in species diversity and populations.[38] In addition, populations of bottom-dwelling invertebrates have shown higher frequencies of deformed mouth parts and head capsules.[38] Changes in the ecosystem are reflected in other wildlife also.[38] Bald eagles, herring gulls, and Forster's terns in the Great Lakes region have undergone significant population declines since the 1960s.[38] Only in recent years, as concentrations of water pollutants in the Great Lakes have declined, have some species (e.g., bald eagles) begun to recover.[4]

Reproductive Effects. Effects on reproduction include embryo toxicity, hatching success, abnormalities in offspring, parental behavior change, and changes in mating. These effects are often accompanied by higher concentrations of PCBs, DDT, dieldrin, and other chlorinated compounds in animals. Specific effects noted in various species include reduced fertility, reduced hatchability, reduced survival of offspring, impaired hormone activity, changed adult sexual behavior, and sparser shoreline populations relative to inland populations. Pollutants of concern that have been linked with reproductive impairment include toxic metals (e.g., cadmium, mercury, and lead), lindane, PCBs, DDT/DDE, dieldrin, and 2,3,7,8-TCDD.



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Usually, observed reproductive effects cannot be linked conclusively to specific pollutants; however, linkages often are made through similarities of effects across species and geographic locations. For example, eggshell thinning in a number of bird species and associated reproductive loss are linked to DDT in the 1960s and 1970s, and decreases in environmental concentrations of DDT have resulted in population recoveries. However, populations in certain regions of the Great Lakes still exhibit reproductive failure. For example, bald eagle populations near the Great Lakes show much lower reproductive success than populations inland. Many eggs in shoreline nests contain lethal concentrations of PCBs, DDE, and dieldrin, resulting in bald eagle reproduction rates too low to maintain a population. In laboratory studies, mink that were fed PCB-contaminated fish responded with decreased reproduction and lower offspring survival. PCB levels in the fish used in that study were similar to those found in some regions of the Great Lakes.

Eutrophication. Eutrophication, which refers to the ability of a waterbody to produce organic material, is a natural process that takes place over geologic periods of time, but which can be accelerated by anthropogenic additions of nutrients (see Figure 6). Eutrophic lakes, which occur when nutrients such as nitrogen and phosphorus are present in excess amounts, are characterized by very high productivity and by high organic content from the decay of plants and recycling of carbon. In freshwater lakes, concentrations of phosphorus, which has only minor atmospheric inputs, generally are limited and therefore control productivity. Atmospheric deposition is not thought to be a major factor in eutrophication of freshwater lakes.

In coastal waters, nitrogen, which can have significant atmospheric inputs in the form of various nitrogen compounds, generally is the nutrient that controls eutrophication.

Eutrophication of a lake or estuary may result in undesirable effects such as oxygen depletion in the water or reduced oxygen levels, nuisance algal blooms, dieback of underwater plants (due to reduced light penetration), and reduced populations of fish and shellfish. The reduction in oxygen levels may reduce bottom-feeder populations, create conditions that favor different species, or cause dramatic fish kills, resulting in an altered food web. In fact, eutrophication recently has been identified by EPA as one of the most serious pollution problems facing estuarine waters of the United States.[45] The atmospheric deposition of nitrogen compounds can contribute significantly to eutrophication in coastal waters.

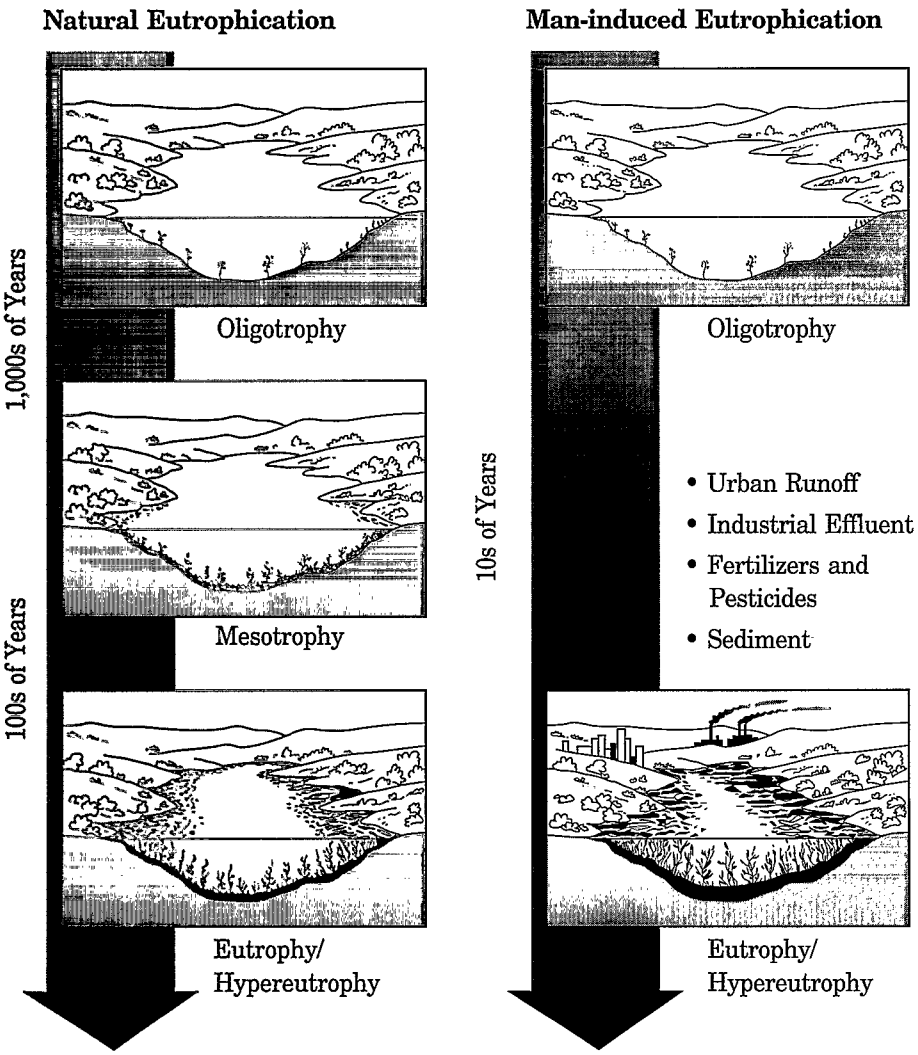


Figure 6. The eutrophication process.[44]

Human Health Effects

Unlike the documented effects of certain pollutants of concern on ecological health in the Great Waters, the effects on human health are not as easy to detect. The potential effects of the selected pollutants of concern, based largely on laboratory studies, range from a broad spectrum of noncancer effects on specific organs to probable human carcinogenicity. However, in thorough human studies, few of these effects have been proven to result from exposures to Great Waters pollutants of concern. Table 5 summarizes the potential human health effects for each of the selected pollutants of concern. Except for nitrogen compounds, all of these pollutants or common compounds of the pollutants bioaccumulate to some degree in humans. The range of effects includes cancer, reproductive effects, developmental effects (i.e., effects on the developing human, including effects on embryos, fetuses, and children), neurological effects (i.e., effects on the brain and nervous system), effects on the endocrine system (e.g., effects on hormone synthesis and function), and other noncancer effects (e.g., liver or kidney damage).

Table 5. Potential Human Health Effects[a] Associated with Pollutants of Concern[b]

Pollutant[d]	Potential Effects on Human Health[c]					
	Cancer[e]	Reproductive/ Restrictions[f]	Neurological/ Behavioral	Immuno-logical	Endocrine	Other Noncancer[g]
Cadmium and compounds	Probable[h]	●	●	●		Respiratory and kidney toxicity
Chlordane	Probable[h]	●[f]	●	●	●	Liver toxicity[h]
DDT/DDE	Probable[h]	●[h]	●[f]	●		Liver toxicity[h]
Dieldrin	Probable[h]	●[h]	●[f]	●	●	Liver toxicity[h]
Hexachloro-benzene	Probable[h]	●	●[h]	●	●	Liver toxicity[h]
α-HCH[i]	Probable[h]					Kidney and liver toxicity
Lindane	Probable[j]	● (γ-HCH)	●	●		Kidney and liver toxicity[h]
Lead and compounds	Probable[h]	●[k]	●[k]	●	●	Kidney toxicity[k]
Mercury and compounds		●	●	●	●	Kidney toxicity
PCBs	Probable[h]	●	●	●	●	Liver toxicity
Polycyclic organic matter	Probable[h]	●		●		Blood cell toxicity
2,3,7,8-TCDF	Not classifiable[h]	●		●	●	Liver toxicity
2,3,7,8-TCDD	Probable[j]	●[l]	●[l]	●[l]	●[l]	Integument toxicity[l]
Toxaphene	Probable[h]	●[f]	●[f]	●	●	Cardiovascular effects; liver toxicity[f]

[a]These data are based on a compilation of results from both human and animal studies. Potential for effects will depend on the level and duration of exposure and the sensitivity of the exposed organism.

[b]Where footnoted, data for this table are taken both from EPA sources [48-54] and the applicable Agency for Toxic Substances Disease Registry (ATSDR) Toxicological Profile [14-22, 24-26, 55]; otherwise, all data are taken from the applicable ATSDR Toxicological Profile alone.

[c]For this table, a chemical was considered to induce an effect if human or laboratory mammal data indicating a positive result were available. Blanks mean that no data indicating a positive result were found in the references cited (not necessarily that the chemical does not cause the effect).

[d]Nitrogen compounds are not included in this table because they are considered a pollutant of concern only for eutrophication.

[e]A chemical is classified as a "probable human carcinogen" when there is limited or no evidence of human carcinogenicity from epidemiologic studies but sufficient evidence of carcinogenicity in animals (corresponds to EPA weight-of-evidence category B). A chemical is classified as "not classifiable as to human carcinogenicity" when there is inadequate human and animal evidence of carcinogenicity or when no data are available (corresponds to EPA weight-of-evidence category D).

[f]Data from the applicable EPA Health Effects Assessment (HEA) document.[50-53]

[g]This is only a sample of other noncancer effects that may occur as a result of chronic exposure to the pollutant. Additional adverse human health effects may be associated with each chemical.

[h]Data from EPA's Integrated Risk Information System.[49]

[i]Toxicity data are available primarily for γ-HCH and technical-HCH (a mixture of several HCH isomers), with limited data available for α-HCH.

[j]Data from EPA's Health Effects Assessment Summary Tables (HEAST).[48] HEAST classifies these chemicals as probable human carcinogens; however, these carcinogenic evaluations are currently under review by EPA.

[k]Data from EPA's Reportable Quantity (RQ) Document for lead.[54]

[l]Data from *Biological Basis for Risk Assessment of Dioxins and Related Compounds*.[56]

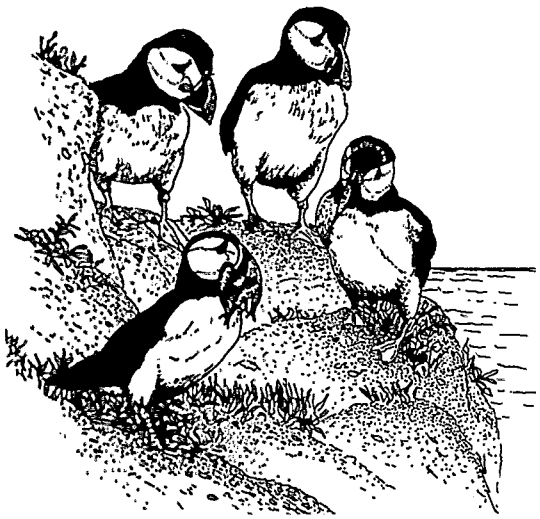
In general, few of these chemicals are acute toxicants or genetic toxicants at concentrations found in the Great Waters; however, several are developmental toxicants that, through low-level exposures to parents, are capable of altering the formation and function of critical physiological systems and organs in children.

Two studies in the United States looked at infants and children who were nonoccupationally exposed to PCBs during prenatal development. Both studies found nervous system deficits. One study showed that children of mothers who ate PCB-contaminated fish (on average 2 to 3 meals per month of lake trout or salmon) from Lake Michigan before 1980 (when PCB concentrations in fish were higher than at present) exhibited deficits in cognitive function.[8] In another study, children in North Carolina showed motor abnormalities at birth and psychomotor delay at up to 2 years of age.[8] Both studies have generated controversy, mainly over study design, data analysis, selection of appropriate statistical tests, and even whether psychological tests are appropriate instruments in population studies.[46]

In a followup to the Lake Michigan study, the same children were evaluated at 4 years of age. These children were found to have subtle deficits in short-term memory and speed of information processing, which could impact the child's ability to master basic reading and arithmetic skills in school. An 11-year followup study on these children has begun.[47]

Summary of Current Understanding of Effects

1. What Are the Major Effects Associated with Pollutants of Concern for Atmospheric Deposition?



The potential human health and environmental effects associated with the selected pollutants of concern are generally well documented. In humans, the potential effects include cancer, reproductive and developmental effects, neurological effects, endocrine and immune system effects, and organ system toxicity. All of the pollutants of concern (except nitrogen compounds) are known to bioaccumulate in animals, including humans. In animals and plants, the potential effects of individual pollutants are not always well defined; however, linkages have been made between exposure to pollutants of concern and observed fish and bird deaths, reproductive effects, deformities in wildlife, and population declines. In the environment, it is difficult to relate a specific effect of concern (e.g., reproductive effects) to a single pollutant, because most affected animals have elevated body concentrations of many pollutants. It is known, however, that exposure to pollutants of concern can result in serious ecological and human health effects, particularly when animals are exposed to the pollutant through intake of food.

In addition, it is well established that nitrogen is usually the limiting nutrient controlling eutrophication in coastal waterbodies and that eutrophication in these systems can cause severe system-wide ecological effects.

2. **What Is the Contribution of Atmospheric Deposition to Adverse Human Health and Environmental Effects?**

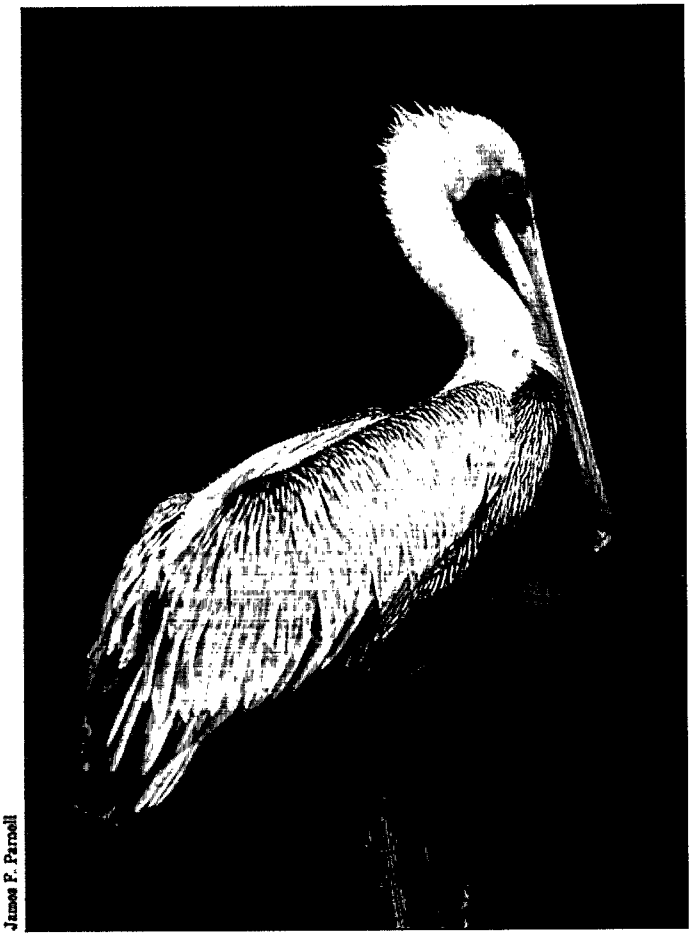
   The relationship between adverse effects of toxic pollutants and atmospheric deposition is not well understood. Some correlations and linkages between specific pollutants of concern and effects in the Great Waters can be established. Yet, at this time, quantifying the contribution of atmospheric deposition of each pollutant of concern to ecological and human health effects is not possible. For example, a pollutant may produce reproductive effects at a given concentration under certain exposure conditions, but the pollutant present in a waterbody generally is derived from many sources, and the link between an observed reproductive effect and atmospheric deposition is very difficult to determine.

Comparisons with Water Quality Benchmarks

As one means of assessing the significance of contamination of the Great Waters caused by the selected pollutants of concern, available water sampling data can be compared with various water quality criteria. Such comparisons are consistent with requirements in section 112(m) of the 1990 Amendments for EPA to assess the contribution of atmospheric deposition to exceedances of certain water quality standards and criteria. This section first describes several sets of relevant water quality benchmarks—EPA's national ambient water quality criteria (AWQC), EPA's recently proposed Great Lakes water quality criteria (pGLWQC), and the U.S.-Canadian Great Lakes water quality objectives (GLWQOs)—and then summarizes how the available Great Waters sampling data compare with the criteria. Because of limited sampling information for many of the selected pollutants of concern in Great Waters other than the Great Lakes, this summary focuses primarily on the Great Lakes.

This section compares water sampling data with water quality benchmarks, rather than comparing sediment contamination data or biological contamination data to appropriate benchmarks, for two main reasons: (1) the specific emphasis of section 112(m) requirements on water quality standards and benchmarks, and (2) the limited availability of Federal or other widely accepted numerical benchmarks for sediments or living organisms for the selected pollutants of concern. However, because of the strong tendency of most of the selected pollutants of concern to bind to sediments and to bioaccumulate, comparisons of sediment and biological contamination levels to appropriate benchmarks, where such benchmarks are available, has advantages over comparisons

based on water contamination levels. First, for most of the pollutants, sediment and biological contamination levels generally are much higher, and therefore easier to measure, than water contamination levels. Similarly, the levels of concern (i.e., the benchmarks) for these pollutants are generally much lower for water than for sediments or for living organisms, resulting in the need to be able to measure very low water concentrations to ensure that criteria or standards are not being exceeded. For example, the newly proposed Great Lakes water quality criterion for 2,3,7,8-TCDD is 9.6×10^{-6} parts per trillion (9.6×10^{-12} milligrams per liter), an extremely low level. Second, sediment and biological contamination levels better reflect the overall pollutant loading over time in a waterbody because of the tendency of the selected pollutants of concern to accumulate in sediments and in living organisms. Therefore, the absence of water quality benchmark exceedances for pollutants that have a strong tendency to bind to sediments and to bioaccumulate does not necessarily indicate the absence of contamination at levels of potential human health or ecological concern.


James F. Parnell

Maximum contaminant levels (MCLs) are "maximum permissible level[s] of a contaminant in water which is [are] delivered to any user of a public water system."[57] These levels are developed under the Safe Drinking Water Act, and the goal in developing MCLs is to approach as closely as possible ideal human health-based levels while still taking into account the cost of achieving the levels as well as the availability of technology to achieve them. At this time, some chemicals do not have MCLs. Few violations of existing levels have been found in Great Lakes drinking water systems, and, for the pollutants that do exceed their MCL, the distribution system rather than the water source may be the principal cause.[31]

AWQC are designed to protect humans and freshwater and saltwater animals and plants from harmful effects resulting from both chronic and acute exposures.[58] The development of AWQC is an ongoing process that is meant to reflect current knowledge on health and welfare effects, dispersal of pollutants across media, and effects on animal and plant communities and on reproduction. Several different AWQC values may be published for an individual chemical, including values designed to protect freshwater aquatic organisms (for both acute and chronic exposure), marine aquatic organisms (for both acute and chronic exposure), and humans (for chronic exposure through consumption of both fish and drinking water and for chronic exposure through fish consumption only). EPA's national AWQC, which are based entirely on scientific data, are provided as guidelines and are not directly applicable as enforceable water quality standards. Rather, AWQC are intended to be used by States as a basis for developing regulations.

Galveston Bay NEP

Applicable national AWQC are available for all of the selected pollutants of concern except 2,3,7,8-TCDF and nitrogen compounds.

In April 1993, EPA proposed and requested public comment on new water quality criteria specifically for the waters in the Great Lakes system.[29] When the criteria are made final, they will form the basis for new water quality standards to be issued by States in the Great Lakes basin. Criteria are proposed to protect aquatic life (for both acute and chronic exposure), wildlife (for exposure through food webs), and humans (for chronic exposure through consumption of both fish and drinking water and through water-related recreation). The proposed methods for deriving these criteria differ in some respects from the methods used for deriving national AWQC, and, in general, the proposed Great Lakes criteria are lower to account for bioaccumulation.

The Great Lakes Water Quality Agreement of 1978 is an agreement between the United States and Canada that adopted the principle of "virtual elimination" of persistent toxic substances to the Great Lakes (i.e., a goal of zero discharge).[59] For a number of chemicals, the Agreement includes specific GLWQOs that are set to protect the most sensitive user of the water among humans, aquatic life, and wildlife. For other persistent toxic chemicals, such as hexachlorobenzene, no specific GLWQO has been established; for such chemicals, concentrations in water and in aquatic organisms should be lower than detection levels. Specific GLWQOs are available for 8 of the 15 selected pollutants of concern, and recommended values are available for four additional ones.

The table in Appendix B shows which of the selected pollutants of concern have potentially exceeded national AWQC, pGLWQC, and GLWQOs, based on Great Lakes sampling data since 1980. The table provides comparisons of maximum open water (i.e., away from shore and not as strongly influenced by direct discharge) concentrations of the pollutants with their respective water quality benchmarks. In interpreting these results, it is important to remember that the sampling data used in the comparisons are maximum values (generally based on a total of 10 to 30 samples) and are not necessarily representative of ambient water concentrations throughout the lake. Dieldrin, mercury, and PCBs exceeded criteria in all five Great Lakes, while DDT/DDE exceeded criteria in all but Lake Superior (mercury and PCBs almost always exceeded more than one of the applicable criteria). In addition, hexachlorobenzene exceeded criteria in Lakes Erie and Ontario, and cadmium exceeded criteria in Lake Erie. Thus, open water concentrations of several pollutants of concern in the Great Lakes—which are expected to be substantially lower than maximum near-shore concentrations—have potentially exceeded applicable water quality criteria at some locations in the recent past. Moreover, maximum concentrations of most of the other selected pollutants (i.e., those that did not exceed criteria) in most of the lakes were within a factor of 10 of the lowest applicable criterion, indicating contamination that approaches levels of concern.

Sampling data for the selected pollutants of concern in Lake Champlain are considerably more limited than for the Great Lakes. Except for lead, the sparse sampling data available generally indicate that these pollutants, if present at all, are below detection limits (i.e., below the levels that can be detected by the sampling and analysis methods used).[60] For lead, concentrations have exceeded applicable water quality criteria at some locations in the recent past.[60]

No widespread concentrations of metals exceed EPA water quality criteria in the mainstem Chesapeake Bay. Yet, a limited number of measured concentrations in the tidal tributaries to the Chesapeake Bay exceeded EPA water quality criteria and State water quality standards for cadmium and lead (as well as for copper and zinc). No exceedances of water quality criteria or standards were reported for the remaining air pollutants of concern within Chesapeake Bay.[39]

Case Studies of Exposure and Effects

As illustrations of exposure and effects resulting from pollutants in the Great Waters, the following case studies are presented. These case studies look at PCB concentrations in the food web in Lake Ontario, the effects to humans from exposure to mercury through intake of contaminated fish, the effects associated with low-level exposures to pollutants of concern in Forster's terns, and the effects of nitrogen loadings on Chesapeake Bay. In reviewing these case studies, remember that the overall contribution of atmospheric deposition to exposure levels and effects is generally not well understood.

Subtle Effects Are Associated with Exposure to Low Levels of Pollutants of Concern


James F. Parnell

Exposure to low levels of many pollutants of concern may result in ecological or human health effects that are not easy to recognize. The effects from high-dose exposures, such as acute toxicity and death, are far easier to observe than those from low-dose exposures, which often are delayed, long-term effects. In offspring, these effects may be the result of low-level exposures to parents. As a result, subtle health effects in wildlife and human populations resulting from low-level exposures could be overlooked as conditions in the environment improve and exposure levels decrease. This point is illustrated in the following study of a Forster's tern colony (fish-eating birds) in Green Bay, Wisconsin.

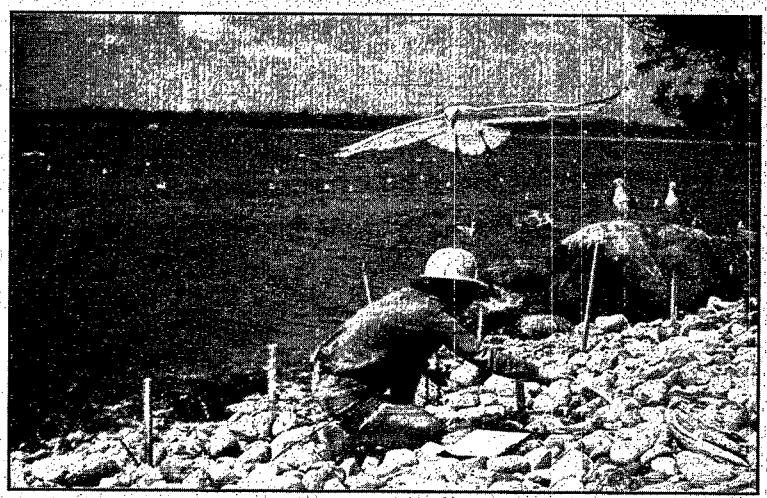
In 1983 and 1988, research teams studied the reproductive success of a colony of Forster's terns nesting on a waste disposal facility in Green Bay as compared with a control population that was nesting on an inland lake and that was not dependent on food sources in the Great Lakes. In 1983, tern offspring from the Green Bay colony experienced lower hatch rates of eggs, lower chick body weight, lower rates of chicks learning to fly, and decreased parental care as compared with the control colony. Within 17 days after hatching, 35 percent of the chicks had died, and the birds had abandoned the area. Researchers linked

New Reports on the Effects of "Environmental Hormones"

Mounting evidence indicates that many chemicals, including DDT, lindane, atrazine, PCBs, dioxin, and mercury, that have been released to the environment can disturb the hormonal (or endocrine) systems of humans and wildlife. Two recent scientific articles describe the major concerns with the release of these chemicals, often called "environmental hormones"—the variety of noncancer effects, such as developmental and male and female reproductive impacts, linked to chemicals that mimic estrogens (female hormones) and other hormones, and a proposed link between exposure to environmental estrogens and breast cancer.

Disruptions to the endocrine system, especially during development in utero or in very early life, and of the organs that respond to endocrine signals are of concern because the effects caused by exposure during an organism's development are permanent and irreversible, and they may go undetected until an organism reaches adulthood and, for example, tries to reproduce.[61]

Environmental hormones work by being accepted by organs as a hormone, but without producing appropriate effects. Exposure of fetuses to these chemicals can profoundly disturb organ development. Organs that appear to be at particular risk for developmental abnormalities are those affected by female/male hormones, including both female and male reproductive organs. In both sexes, the external genitals, brain, skeleton, thyroid, liver, kidney, and immune system are also potential targets for endocrine-disrupting chemicals.[61]



International Joint Commission

In wildlife, exposure to endocrine-disrupting chemicals has been associated with decreased fertility in birds, fish, shellfish, and mammals; decreased hatching success in fish, birds, and turtles; demasculinization and feminization of male fish, birds, and mammals; defeminization and masculinization of female fish and birds; and alteration of immune function in birds and mammals.[61]

In laboratory studies, mammary (breast) cancer has been linked with environmental estrogens, such as organic compounds (e.g., DDT or dioxins) and polycyclic aromatic hydrocarbons (PAHs). In addition, recent epidemiologic studies have found that breast fat and blood fats of women with breast cancer contain significantly elevated levels of some chlorinated organic compounds compared with noncancer controls. Breast cancer, in the majority of cases, is thought to arise from interactions between genetic and environmental factors. It is hypothesized that environmental estrogens increase the risk of breast cancer by mechanisms that include interaction with breast-cancer susceptibility genes.[62]

these effects to elevated concentrations of PCBs in the chicks and eggs. In 1988, the median concentrations of PCBs in the Green Bay colony had decreased significantly from those measured in the 1983 study. During the first 17 days of the 1988 study, certain types of effects for the Green Bay colony—including hatching success, weight gain, and the rate of chicks learning to fly—were comparable to those for the control group in 1983. However, on day 18, chicks began to show signs of "wasting" and by day 31, 35 percent of the young had died—the same percentage that died within 17 days in 1983. To date, "wasting" appears to be the most sensitive effect in Forster's terns resulting from low-level exposure to PCBs, and it appears to be delayed as exposure is reduced. Also, because of the delayed onset of the effects, if the 1988 study had been conducted for only the same period of time as the 1983 study, incorrect conclusions about environmental recovery would likely have been drawn.

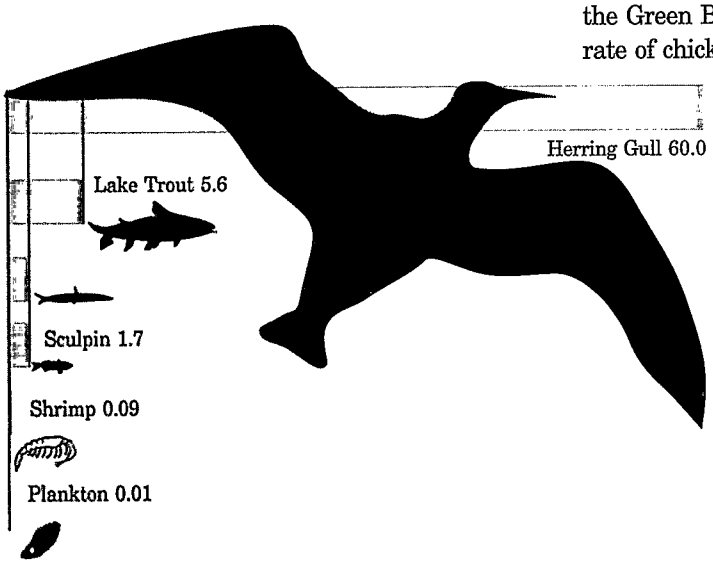


Figure 7. Biomagnification of PCBs in the Lake Ontario food web, 1982. [32]
PCBs shown in parts per million.

PCBs Concentrate in the Food Web

PCBs are a class of compounds that are persistent in the environment and are known to be toxic to humans and ecosystems. PCBs are associated with cancer, neurological effects, and effects on reproduction and development in humans. In wildlife, they have been associated with premature deaths, effects on reproduction, and immune system effects. Because of low ambient concentrations in water, the importance of exposure routes other than drinking water should be considered when assessing the exposure and effects of PCBs.

The most important route of exposure to PCBs is through intake of food following biomagnification through the food web. This is illustrated by the concentrations of PCBs in plants and animals in the food web in Lake Ontario (see Figure 7). Floating microscopic plants (phytoplankton) and animals (zooplankton) take in and retain PCBs. Fish then eat these plankton, taking in the chemicals present in the plankton. When an animal cannot break down or eliminate the PCBs it takes in, the PCBs accumulate in the animal's fatty tissue. Soon, the animal has higher concentrations than the surrounding environment. The process leading to increasing concentrations of chemicals at higher levels of the food web is known as biomagnification. Biomagnification continues up the food web as predator fish feed on smaller fish, and as birds and mammals (including humans) then feed on predator fish. Biomagnification results in far greater concentrations of PCBs in animals than would be expected based strictly on concentrations in the water.

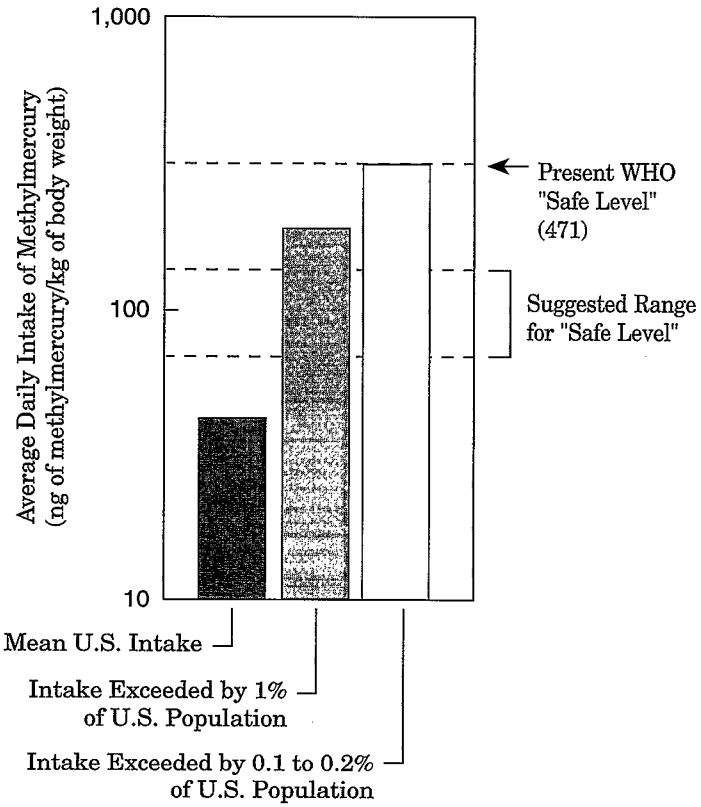


Figure 8. U.S. daily intakes of methylmercury versus World Health Organization "Safe Levels."[63]

Current "Safe Levels" for Mercury May Not Be Protective of Human Health

Mercury exists in the environment in three forms: metallic mercury, methylmercury (and other organic forms), and mercury salts. Metallic mercury is poorly absorbed by the body, and high doses in humans may result in no effects.[38] Methylmercury and mercury salts, on the other hand, have been demonstrated to cause serious human health effects.[38] Methylmercury is of greatest concern because it is readily absorbed into body organs and tends to bioaccumulate to high levels in animals, including fish and humans.

Exposure of humans to mercury may result in kidney damage, damage to the brain and nervous system, and developmental effects. Prenatal exposure to methylmercury is of special concern because recent research indicates that prenatal exposure to methylmercury concentrations fivefold to tenfold lower than current World Health Organization (WHO) "safe levels" may result in subtle neurological effects in children, such as abnormal reflexes and delayed motor skills development.[63] As shown in Figure 8, 0.1 to 0.2 percent of the U.S. population (about 250,000 to 500,000 people) currently exceed the WHO "safe level" for daily methylmercury intake. Based on the recent data assessing the effects associated with prenatal exposure, however, a lower limit to protect from developmental effects may be warranted. As shown in Figure 8, a new "safe level" of about one-tenth the current level has been suggested. It is evident that a substantial fraction of the U.S. population exceeds this suggested new level. The National Institute of Environmental Health Sciences is performing studies to determine whether a lower limit should be established.

Mercury is found naturally in the environment, and it is also released from human activities such as fossil fuel combustion, waste incineration, and other industrial processes. Subsequent atmospheric deposition may lead to increased concentrations in aquatic ecosystems. Human exposure to mercury generally is through the ingestion of fish, which bioaccumulate mercury in muscle tissue.[63] Anthropogenic releases of mercury, combined with naturally occurring mercury and the bioaccumulation potential of mercury, may result in exposure of human residents in the Great Lakes basin to levels of mercury that exceed the current WHO and EPA guidelines for fish consumption. This risk is greatest for population groups that consume affected fish from the Great Lakes, such as some American Indian tribes.[64]

Nitrogen is the Nutrient Controlling Eutrophication in Chesapeake Bay


Steve Minnich

Eutrophication has been recognized as a problem in the Great Lakes, Chesapeake Bay, and coastal waters for over 30 years. In the Great Lakes and other fresh waterbodies, phosphorus was recognized as the nutrient of primary concern, and efforts to reduce phosphorus loadings, such as removal of phosphate from detergents and improved sewage treatment techniques, resulted in substantial phosphorus reduction in waterbodies and improved water quality.[40]

However, in coastal waters such as the Chesapeake Bay, decreases in phosphorus loadings have not resulted in significant decreases in eutrophication. Researchers now identify nitrogen, in forms such as ammonium and nitrates, as the nutrient of primary concern in the Chesapeake Bay and many other coastal waters. Nitrogen loadings cause increased eutrophication in waterbodies, and this can result in oxygen depletion in the water or reduced oxygen levels, nuisance algal blooms, dieback of underwater aquatic plants, and reduced populations of fish and shellfish. This is significant in the context of atmospheric deposition because up to 40 percent of nitrogen in Chesapeake Bay is estimated to result from atmospheric deposition.[40] Thus, atmospheric deposition appears to be contributing significantly to eutrophication problems in Chesapeake Bay and other coastal waters.

Chesapeake Bay has experienced a 20 percent decrease in water column phosphorus concentrations since 1984, but water concentrations of nitrogen have remained relatively constant.[40] During this same time, dissolved oxygen levels, an indicator of recovery from eutrophication, have not increased. Eutrophication in Chesapeake Bay has contributed to depleted fish and shellfish stocks, loss of fish and plant habitats, and losses of underwater aquatic plants related to increased algal growth and decreased light penetration. Studies to improve the health of Chesapeake Bay focus on understanding how nitrogen cycles through the bay and on techniques for decreasing inputs of nitrogen compounds into this waterbody. Recently, tributary basin nutrient reduction goals were established, which has focused attention on atmospheric deposition of nitrogen as a potential source of controllable loadings.[65]

Conclusions Related to Effects

Section 112(m) of the 1990 Amendments directs EPA to assess the environmental and human health effects of pollutants that are attributable to atmospheric deposition to the Great Waters and to determine whether pollutant loadings to the Great Waters cause or contribute to

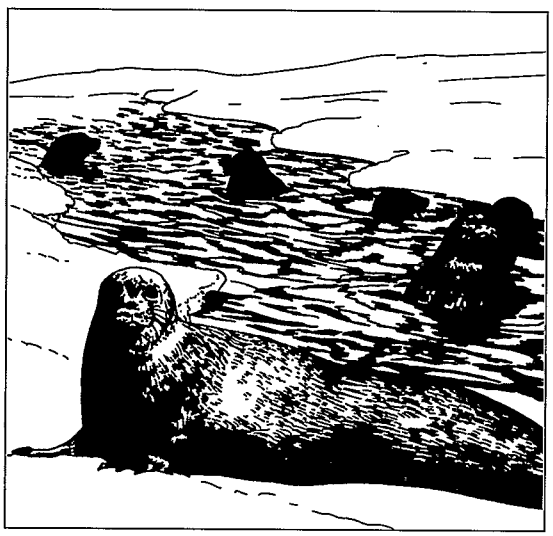
violations of drinking water standards or water quality standards. This section reviewed some of the principal ecological and human health effects associated with selected pollutants of concern in the Great Waters. The following conclusions are drawn from this review of currently available scientific information.

1. **Some ecological effects and human health effects caused by pollutants that are present in atmospheric deposition to waterbodies are subtle, result from long-term exposures to low levels of pollutants, and may be delayed in onset and even occur over multiple generations.**

   The effects associated with the selected pollutants of concern often result from long-term exposures to one or more pollutants present at low concentrations. (For example, long-term exposure to low levels of mercury may result in kidney or nervous system damage. Because of gradual exposure and bioaccumulation in the body, the onset of these effects may go undetected.) Similarly, exposure to many pollutants of concern may have little or no measurable effect on the adult but may result in developmental effects in the fetus or developing newborn. For example, maternal exposures to certain pollutants may be passed to a newborn through feeding or passed to a fetus across the placenta, potentially resulting in adverse effects in the infant or in the fetus, both of which are more sensitive than the adult to environmental pollutants. In addition, effects that have a delayed onset may occur, and these delayed effects are often extremely difficult to detect.

2. **Noncancer effects of the pollutants of concern are of great concern, particularly for animals higher up in the food web.**

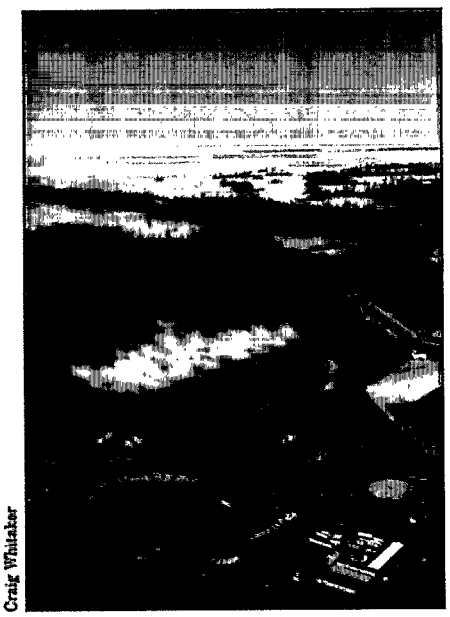
   Laboratory studies indicate that the selected pollutants of concern have a wide range of effects on both wildlife and humans. Field studies indicate that certain Great Waters pollutants can cause immune system effects, effects on reproduction, and neurological effects in wildlife, thereby signalling concern that humans may also be exposed to levels that cause these and other effects. Though many of the pollutants are probable carcinogens, most cause noncancer effects that are also of significant concern. For example, a majority of the 15 selected pollutants of concern may cause harder-to-detect developmental effects, such as delayed development of motor skills. Additionally, many of the pollutants of concern may cause brain or nervous system damage. All of the pollutants of concern also can affect the endocrine system, which may in turn affect reproduction, nervous system function, and development of the immune system. These effects may be as devastating to an individual person or animal as cancer and may also have population impacts. Effects may also occur after little or no latency period and may occur at very low exposures.



Further, because exposure is cumulative, the potential for impacts on the population is extremely high.

3. **Though atmospheric deposition is a significant pathway into the Great Waters for some toxic pollutants, the relationship between atmospheric deposition and the effects on humans and ecosystems is not clearly understood.**

   Recent research has indicated that atmospheric deposition is a significant contributor of some toxic pollutants to the Great Waters; however, the relationship—especially the quantitative relationship—between atmospheric deposition and effects in (or risks to) humans, animals, and plants, for most pollutants, is not clear. Many pollutants of concern for atmospheric deposition also have a long history of discharges to the Great Waters, and current levels of many of these may be the result of recycling from the sediments. Thus, the specific contribution of atmospheric deposition to exposure and effects cannot be quantified at this time.


Craig Whitaker

4. **In many temperate estuarine systems, such as Chesapeake Bay, atmospheric deposition of nitrogen compounds is a major contributor to accelerated eutrophication.**

   Regulations to reduce phosphorus discharges have had limited success in controlling eutrophication in coastal systems. Recent evidence indicates that nitrogen is usually the limiting nutrient in temperate estuarine systems. Moreover, for some waterbodies, such as Chesapeake Bay and Delaware Bay and potentially for many others, a significant percentage of the nitrogen load is from atmospheric deposition (see Table 8, page 55).

5. **Persistence in the environment, tendency to accumulate in animal tissue, and toxicity to humans and other organisms are important indicators of the hazard potential of air pollutants that are deposited to waterbodies.**

   For identifying air pollutants of concern, these three characteristics are most important. Pollutants with these characteristics have the ability to cause health effects in humans and the environment; to biomagnify in the food web, allowing for higher exposure levels to animals at the top of the food web; and to remain in the environment for long periods of time, increasing the opportunity for exposure.

Relative Loading: What Is the Relative Importance of Atmospheric Deposition in Pollutant Loadings to the Great Waters?

A critical step in evaluating atmospheric deposition to the Great Waters is to assess the extent to which hazardous air pollutants actually enter waterbodies from the air. This type of analysis enhances our ability to attribute adverse human health and environmental effects to atmospheric deposition and helps make it possible to trace the air pollutants of concern back to the sources that release them.

Understanding the extent of atmospheric deposition requires an analysis of the amount of a given pollutant that enters a waterbody over some period of time, commonly referred to as the pollutant "loading." It is necessary to analyze not only the total loading of a pollutant, but also the relative loading (i.e., how the loading from atmospheric deposition compares to that from other pathways, such as groundwater seepage and inflow from connecting surface waterbodies, where portions of the pollutant load may not be of atmospheric origin). Once a clear picture of relative loading exists, the importance of atmospheric deposition to the Great Waters can be understood and a determination can be made of the possible measures for controlling air and water quality in an overall context.

The essential framework for evaluating the relative inputs of chemicals to a body of water is an input-output budget, or "mass balance" model. In such a model, the total amounts of a given chemical that enter and exit the waterbody by various pathways are estimated. Figure 9 shows the basic features of a mass balance model for lakes and estuaries. Pollutants can enter lakes, such as one of the Great Lakes or Lake Champlain, from connecting streams and rivers, groundwater inflow, and atmospheric deposition. Pollutants can also reenter lakes through release from the lake bottom. Atmospheric deposition occurs as pollutants are carried down from the air along with falling rain or snow, settle onto water in the form of dry particles, or transfer into the water in the form of a gas. Atmospheric deposition may occur either as direct deposition (i.e., pollutants are deposited directly from air to a waterbody) or as indirect deposition (i.e., pollutants are deposited from

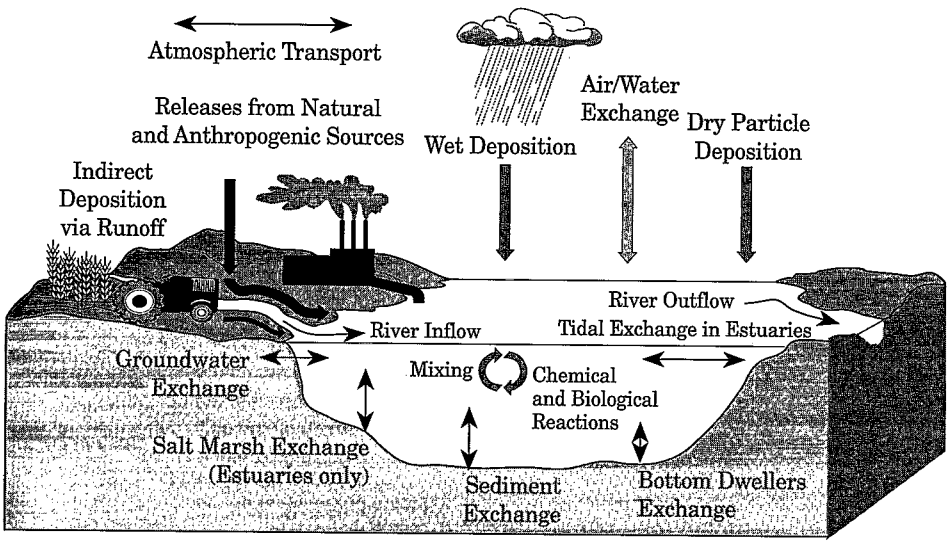


Figure 9. Mass balance model for lakes and estuaries.

The Mass Balance Model

- The mass balance model is the essential framework for estimating relative loadings of pollutants to a waterbody.
- The model establishes a process for identifying and consistently evaluating all ways that pollutants can enter and exit a waterbody.
- Only with a clear understanding of all inputs and outputs of a given pollutant can we begin to understand the relative importance of atmospheric deposition.

air to land and enter a waterbody via runoff or seepage through groundwater). Pollutant outputs from the lake include evaporation, stream and river outflow, breakdown by chemical and biological processes, settling and burial at the bottom, and seepage into groundwater. Pollutants entering surface waterbodies may be diluted by mixing and may undergo reactions that change their physical and chemical forms. Estuaries such as Chesapeake Bay differ from lakes in that they are semi-enclosed bodies of water where fresh water from the land mixes with salt water from the ocean. Estuarine waters have physical and chemical characteristics that make them different from fresh and salt waters. The features of a mass balance model for estuaries differ significantly from those of lakes because of the importance of tidal exchanges and the influence of coastal marshes and waters.

The remainder of this section describes chemical mass balances for the Great Waters to evaluate the relative loading of pollutants by atmospheric deposition. The section begins by summarizing the current understanding of atmospheric deposition processes and mass balances in surface waters. The section then presents mass balance case studies for selected pollutants and waterbodies and uses these case studies to develop generalizations and conclusions that apply to the Great Waters as a whole.

Current Understanding of Relative Loadings

A substantial body of knowledge exists concerning atmospheric deposition processes and loadings to surface waters. Significant additional research is required, though, to better determine, with certainty, the atmospheric loadings of pollutants of concern to the Great Waters and their relative importance in causing human health and environmental effects. Table 6 provides an explanation of several scientific terms that are relevant to relative loadings from the atmosphere, including how airborne pollutants are brought to the earth by "wet" and "dry" deposition processes, the degree to which airborne pollutants are transferred "directly" versus "indirectly" to surface waterbodies, and the influence of a pollutant's chemical and physical form on deposition and cycling between environmental media. This table is followed by answers to four basic questions that address the current scientific capabilities for evaluating atmospheric deposition to the Great Waters.

1. Do We Have the Conceptual Understanding Required to Estimate the Relative Atmospheric Loadings of Pollutants to the Great Waters?

The mass balance approach, in which all pollutant loadings to, and releases from, a waterbody are identified and estimated, provides an appropriate conceptual framework in which to determine the relative importance of atmospheric deposition in causing contamination in the Great Waters. While the amounts of pollutants that enter and exit

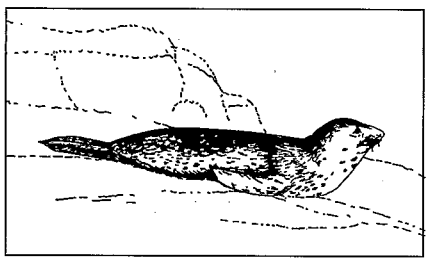


Table 6. Explanation of Atmospheric Deposition Terms

Term	Explanation
Wet deposition	Pollutants in the atmosphere in a gaseous phase can enter water droplets in the air by a variety of processes and be deposited to the earth along with rain, snow, and other forms of precipitation. Metals and organic chemicals that are bound to airborne particulates also are incorporated into precipitation. Wet deposition of gases and particles has been evaluated extensively by several investigators at several locations.
Dry deposition	Dry particles in the air can settle onto water and land surfaces at a rate that depends on the particle size, wind speed, and other factors. Gaseous pollutants also can transfer from the air to the water and land. Currently, methods for measuring dry deposition have large uncertainties compared to methods for measuring wet deposition, and there are no widely accepted methods for estimating how much dry deposition occurs. Recent studies, however, suggest that chemical transfers between air and water play an important role in the mass balance of systems like the Great Waters.
Indirect versus direct deposition	Air pollutants are not only deposited directly to the surface of waterbodies, but are also deposited to watersheds and then discharged into the waters indirectly, through stormwater runoff, tributaries, and groundwater seepage. Where the watershed is large relative to the open water, indirect loading can exceed direct loading. Although indirect loadings are included as a component of a mass balance, procedures are not available for determining these loadings with much certainty.
Reentrainment, Resuspension	Reentrainment is the removal of deposited particles from a water or land surface by air flow above the surface. Whether a particle will be resuspended depends on the adhesion between the particle and the surface, balanced against the lifting force created by wind turbulence.
Volatilization	Previously deposited gaseous chemicals can be reemitted to the atmosphere as the result of many factors, including chemical reactions and changes in temperature or windspeed.
Chemical and physical forms of pollutants	The chemical and physical form of a pollutant affects its mobility in environmental media, its tendency to transfer between media, and its toxicity. Significant aspects of pollutant forms and how they influence pollutant behavior in the environment have been identified. However, numerous uncertainties still exist, and additional study of specific chemicals is needed.

surface waters by some pathways are difficult to quantify and require new research initiatives, development of the mass balance framework is straightforward.

2. **Do We Currently Have Data of Sufficient Accuracy and Precision to Estimate Relative Atmospheric Loadings of Pollutants to the Great Waters?**

   With very few exceptions, the construction of a mass balance for each pollutant has not been possible due to a lack of consistent, coherent, and simultaneous measurements, on a pollutant by pollutant basis, of all loadings to a waterbody. While technically possible, such integrated measurements require a significant commitment in order to generate adequate information to develop scientifically credible mass balances.

3. **Do We Have the Tools to Determine Atmospheric Deposition Rates with Accuracy and Precision?**

   Methodologies of suitable accuracy and precision currently exist to determine the rate of wet deposition of many chemicals to specific locations in the Great Waters. For example, deposition of trace elements and some organic pollutants during rainfall can be adequately measured. Conversely, dry deposition of pollutants attached to particles and the transfer of gaseous pollutants between air and water can, at present, only be estimated by indirect methods. Tools for accurately and precisely measuring dry deposition are not yet available, but some are being developed.

4. **What Is the Scientific View of Our Current Understanding of the Processes Resulting in Atmospheric Deposition?**

   During the past 30 years, the scientific community has recognized the importance of atmospheric deposition in causing surface water contamination and has developed and refined models describing the physical and chemical processes responsible for atmospheric deposition. At present, estimates have been made of atmospheric loadings of a few specific pollutants to a few specific waterbodies, yet uncertainties are associated with these estimates. Knowledge of processes in the atmosphere and in surface waters is not sufficient to determine, with confidence, the magnitude and impact of atmospheric deposition of all of the pollutants of concern to all of the Great Waters. However, specific studies do indicate that the relative contribution of pollutant loading from atmospheric deposition can be significant.

Relative Loading Case Studies

As illustrations of relative contaminant loadings to the Great Waters and other similar waterbodies, mass balances are presented below for PCBs in Lake Superior, mercury in lakes in Wisconsin and Sweden, nitrogen in several Atlantic estuaries, and cadmium in Delaware Bay. To construct these mass balances, researchers used data on the concentration, amount, and movement of pollutants in different environmental media, as well as mathematical models to estimate pollutant transfers into and out of waterbodies over time. This process requires numerous assumptions and is filled with uncertainties. Nevertheless, as summarized below, **these mass balance case studies provide a strong indication that the relative contribution of pollutant loading from atmospheric deposition can be substantial, depending on the particular pollutant and waterbody.**

The Atmosphere is Both a Major Contributor to, and Recipient of, PCBs in Lake Superior

Of the organic chemicals for which a mass balance can be developed, PCBs have been studied the most because of their tendency to bioaccumulate and because of their persistence, widespread distribution in the environment, and toxicity. PCBs provide an interesting case study because they have been banned from further production, although they are still in use for limited purposes, and because they are still apparently being released to the air. Several researchers have accumulated sufficient information on the amount and cycling of PCBs in Lake Superior to develop a PCB budget (see Figure 10). Lake Superior is the largest of the Great Lakes, accounting for more than 50 percent of the Great Lakes water volume. In addition to having a large surface area compared to its drainage area, Lake Superior is strongly influenced by atmospheric interactions (for PCBs). Major inputs to the lake include river flows (which contain municipal and industrial wastes) and atmospheric deposition. PCBs may be lost from the lake by flow through the St. Mary's River, by settling into the bottom sediments, by chemical or biological degradation, and by release to the air.

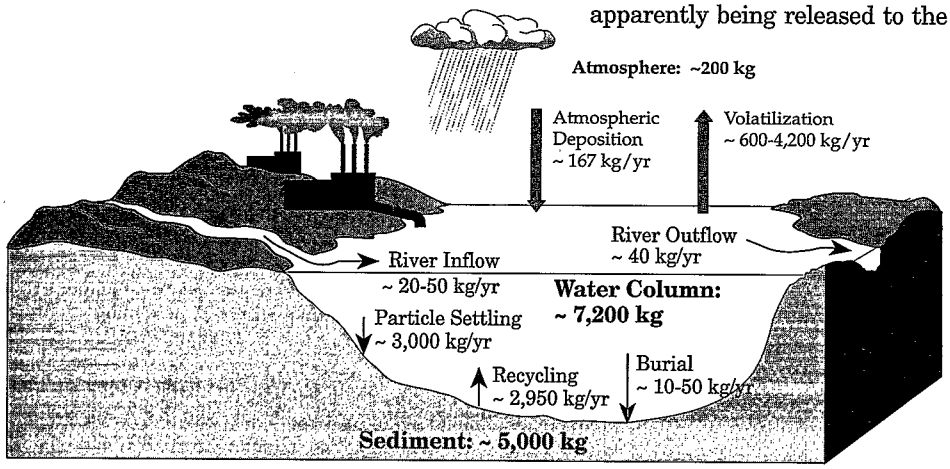


Figure 10. Mass balance of PCBs in Lake Superior. Numbers presented are approximations. Data taken from reference 6.

Mass balance calculations indicate that atmospheric deposition currently contributes approximately 77 to 89 percent of the total yearly input of PCBs to Lake Superior. Annual atmospheric loadings (per unit

Mercury Deposition in the Lake Champlain Basin

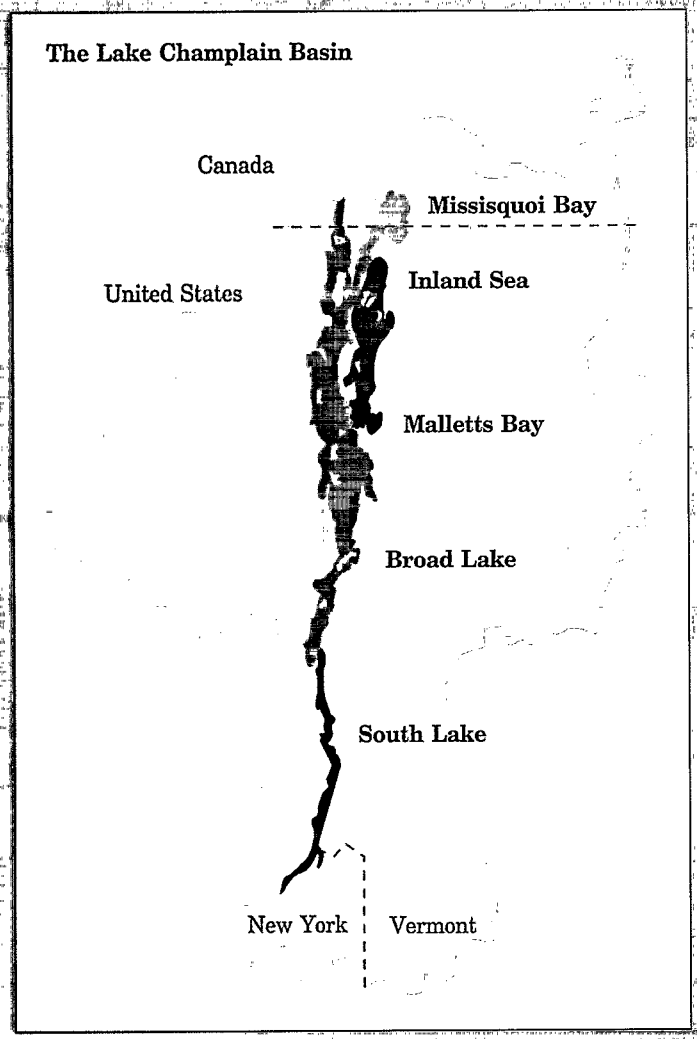
The importance of the atmosphere in the cycling of mercury in the Lake Champlain Basin is being investigated for the first time by a team of researchers from the academic community (the University of Vermont and the University of Michigan), State agencies (State of Vermont, Vermont Monitoring Cooperative), and the National Oceanic and Atmospheric Administration. The project objective is to assess the magnitude and seasonal variations in the levels of atmospheric mercury and mercury deposition in the Lake Champlain Basin (LCB). Emphasis is also being placed on the processes that lead to indirect deposition of mercury to Lake Champlain since the lake surface represents only 5 percent of the drainage basin.

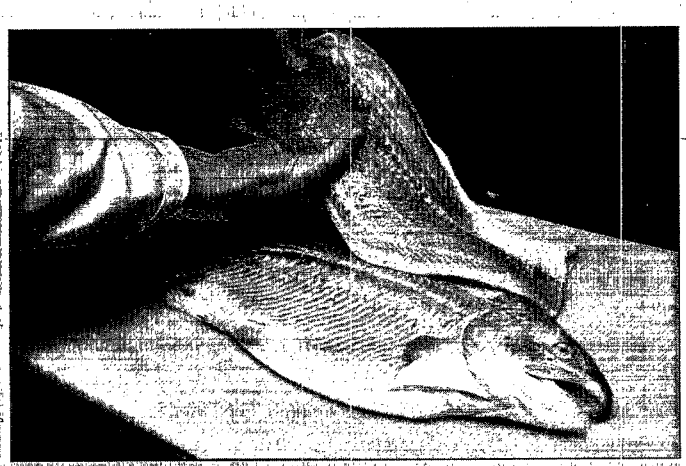
Results from the first year of monitoring, which commenced in December 1992, reveal that mercury in precipitation varies seasonally, with the highest concentrations being observed during the warm months from May to August. The total mercury wet deposition was similar to the observed deposition in northern Michigan but less than the amounts measured at southern Michigan sites.

Preliminary meteorological analysis of the atmospheric mercury measurements revealed that the highest concentrations of mercury in precipitation were associated with air masses reaching the Vermont site from the northwest, out of Canada. However, elevated mercury levels in the LCB are also associated with air masses arriving from the southwest, from the Midwest Region. In comparison, the highest concentrations of mercury in precipitation recorded in Michigan are often associated with air masses reaching the Michigan sites from the south-southwest.[66,67]

The ambient mercury levels observed in the LCB were also similar to those measured in the State of Michigan.[68]

While fish consumption advisories are in effect for Lake Champlain due to mercury and PCBs, it is unclear at this time what fraction of the mercury in the fish is accounted for by atmospheric deposition.



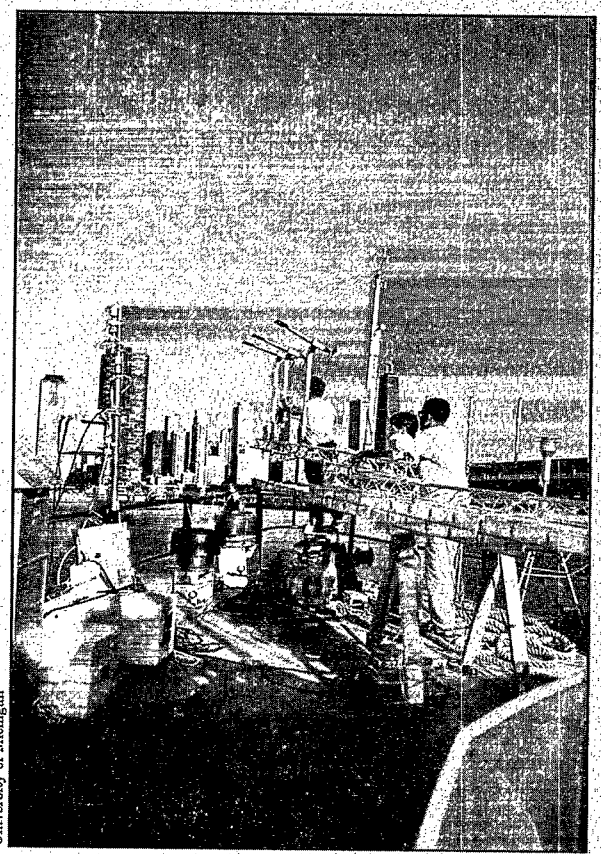


Jimmy W. Crawford, PTI

New Reports on Mercury Measurements

A recently completed report on the Lake Michigan Urban Air Toxics Study (LMUATS) provides new insight on the levels and behavior of atmospheric mercury in the southern Lake Michigan Basin.[69] The report documents the measurements of total mercury performed simultaneously at three locations during the summer of 1991 as part of the month-long intensive study. This project was one of the first designed to observe the behavior of the many different classes of compounds* as they moved from the urban/industrial source regions across Lake Michigan.

The LMUATS revealed that ambient mercury concentrations, both gaseous and particulate (i.e., attached to particles), are significantly higher (approximately 5 times higher) in the Chicago urban/industrial area compared to the levels measured at the same time in surrounding areas. At the urban Chicago location, the levels of atmospheric mercury varied greatly from day to day as well as within days for mercury gas with the highest concentrations observed during the daytime hours.



University of Michigan

Measurements of particulate mercury provided new data on the levels, particle size, and form of this critical pollutant. The concentrations of this kind of mercury are significantly greater than those observed previously in the Great Lakes Region (10 to 50 times greater and also attached to larger particles than expected). Since the 1991 study on Lake Michigan, over-water measurements of mercury have been performed in the southern Lake Michigan Basin, with levels exceeding those measured during LMUATS.[69] These findings indicate that most dry deposition estimates for mercury have probably underestimated the mass loading of this toxic compound to both terrestrial and aquatic systems. Additional studies are needed to allow us to understand what factors are controlling the transport and deposition of mercury from the atmosphere.

*A comprehensive suite of hazardous air pollutants including other heavy metals, PCBs, PAHs, and selected pesticides were also investigated during the study.

surface area) to the Great Lakes are similar to those to Chesapeake Bay, suggesting rapid atmospheric mixing and transport of PCBs over North America. PCB losses from Lake Superior occur primarily by volatilization (evaporation), which represents nearly 90 percent of total losses, while river outflow and burial in bottom sediments each represents only about 5 percent of total PCB losses. Based on these mass balance calculations, PCBs appear to be gradually leaving Lake Superior and transferring back into the atmosphere. It appears that the majority of the decrease in PCB concentrations is due to volatilization.

Atmospheric Deposition Dominates Mercury Loadings to Lakes Studied in Wisconsin and Sweden

The importance of atmospheric deposition in the cycling of mercury in large lakes has been demonstrated in two major mercury investigations: (1) the "Mercury in Temperate Lakes Program" in Wisconsin, and (2) an investigation of mercury in Sweden. Both studies indicate that most of the mercury that enters these lakes comes from atmospheric deposition and that slight increases in atmospheric mercury loading could result in higher levels of mercury in fish. The Wisconsin and Sweden studies show broad agreement.

The Wisconsin study indicates that atmospheric deposition accounts for the majority of the mercury in water, fish, and sediments of Little Rock Lake and other similar lakes (see Figure 11). Measurements of airborne particulate matter and precipitation indicate that particulate mercury in the air falls to the earth with rain. The lake ecosystem is very sensitive to atmospheric inputs of mercury, to the extent that even small increases in mercury loadings from the air could lead to elevated levels in fish.

Similarly, analyses of the mercury flows into and out of typical lakes in the southern half of Sweden show that mercury enters lakes mainly through atmospheric deposition. The Swedish study also suggests that a large portion of the mercury that eventually enters a lake is first stored in the soils of forests located within the lake's drainage area. Current atmospheric deposition into drainage areas was found to be, on average, 10 times greater than the amount of mercury leaving the areas through stormwater runoff. As a result, even if anthropogenic emissions of mercury were to suddenly cease, mercury that has accumulated in the soil would continue to be released to lakes from the forest soils for a long time, perhaps several centuries.

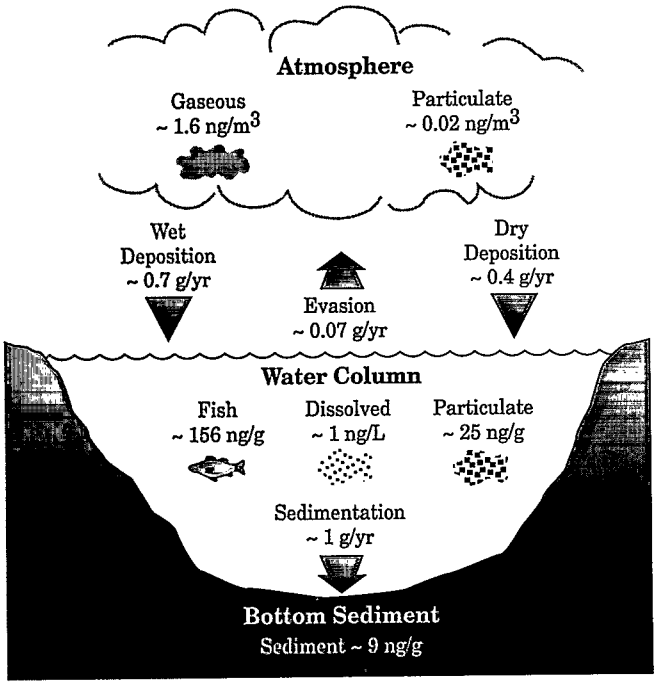


Figure 11. Mercury in Little Rock Lake, WI. Values shown are approximations.[6]

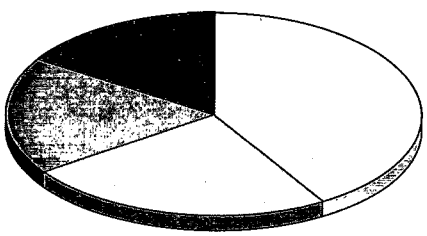


Figure 12. Annual nitrogen loadings to Delaware Bay. Values shown are approximations.[6]

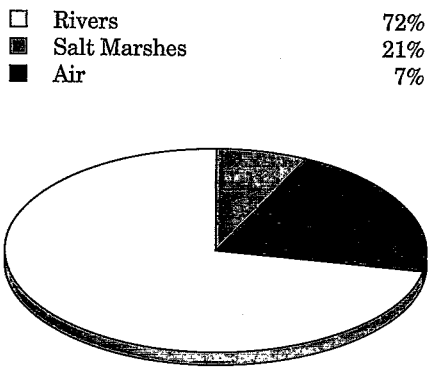


Figure 13. Cadmium loadings to Delaware Bay. Some unknown quantity of the river input includes atmospheric fallout into the watershed.[6]

Atmospheric Inputs of Nitrogen to Atlantic Estuaries Appear Significant

Nitrogen, in forms available to living organisms, can enter estuaries in a variety of ways. Of these, atmospheric deposition of nitrogen can be important. For example, 10 percent of the total nitrogen inputs to Long Island Sound is estimated to come from the direct deposition of nitrogen compounds (nitrate and ammonium) from the air.[70] Indirectly, the inputs of nitrogen from the air to Long Island Sound are probably much greater, since over half of the nitrogen entering the Sound comes from upstream sources and urban runoff,[70] and much of this nitrogen is probably derived originally from the atmosphere. Studies of the watersheds of the entire Chesapeake Bay and of the upper Potomac River have estimated that 25 to 40 percent and 28 percent, respectively, of the nitrogen inputs to these systems come from atmospheric deposition.

In Delaware Bay, a more heavily urbanized estuary adjacent to Chesapeake Bay, direct and indirect atmospheric deposition provide about 14 percent of the annual nitrogen input (see Figure 12). During late spring and early summer, however, atmospheric deposition to Delaware Bay is estimated to provide 25 percent of the total nitrogen loading, due to greater atmospheric loading coupled with lower river flows. Comparison of these results for Chesapeake Bay and Delaware Bay shows that these two nearby waterbodies receive similar inputs of nitrogen from the atmosphere. However, because of the more urbanized nature of the area surrounding Delaware Bay (and the corresponding higher nitrogen loadings from municipal and industrial discharges), the relative atmospheric loading to the Delaware is slightly less, though still significant.

The Atmosphere Contributes Important Loadings of Cadmium to Delaware Bay

The mass balance for cadmium in Delaware Bay is one of the few that is rather complete and realistic, as it considers both river inflows and atmospheric deposition, the effects of tides, chemical transfers between the Bay's water and bottom sediments, and outputs as well as inputs to the estuary. More research is needed, however, to determine the amount of cadmium that enters Delaware Bay from groundwater and indirect atmospheric deposition.

Cadmium in Delaware Bay comes from three main inputs: rivers, salt marshes, and direct atmospheric deposition. As presented in Figure 13, direct atmospheric deposition accounts for 7 percent of the cadmium loading to the Bay. Rivers are the dominant cadmium input (72 percent), but some fraction of the cadmium entering from rivers originally comes from the air. Most of the cadmium loading is exported from the Bay to coastal waters and, consequently, does not settle into bottom sediments. The release of cadmium from the bottom sediments of the bay into the water is seven times faster than the rate at which cadmium gets buried in the bottom.

Conclusions Related to Relative Loadings

The research findings and case studies lead to a number of conclusions concerning the importance of atmospheric deposition to surface water contamination. The conclusions presented below apply both to surface waters in general and to the Great Waters in particular.

1. **Although uncertainties still exist, case studies demonstrate that atmospheric deposition may be an important, and in some cases primary, contributor of toxic chemical contamination and nitrogen enrichment to the Great Waters.**

Current understanding of relative loadings from atmospheric deposition is limited by a lack of data for many chemicals, undetermined flows into and out of waterbodies for many pathways, and insufficient monitoring data. Nevertheless, relatively complete information for a few cases clearly shows that atmospheric deposition may be a significant contributor to contamination in surface waterbodies, including the Great Waters. For example, atmospheric deposition is the primary way mercury gets into some waterbodies (see Tables 7 and 8 and Figure 11). For PCBs, direct atmospheric deposition supplies approximately 77 to 89, 63, and 58 percent of the annual PCB loadings to Lakes Superior, Huron, and Michigan, respectively, as shown in Figure 14. Moreover, atmospheric deposition is the primary way lead has been entering the Great Lakes (Table 7). Atmospheric deposition contributes an estimated 25 to 40 percent of the total nitrogen loading to Chesapeake Bay and a significant fraction of nitrogen loadings to other U.S. coastal waterbodies (Table 8).

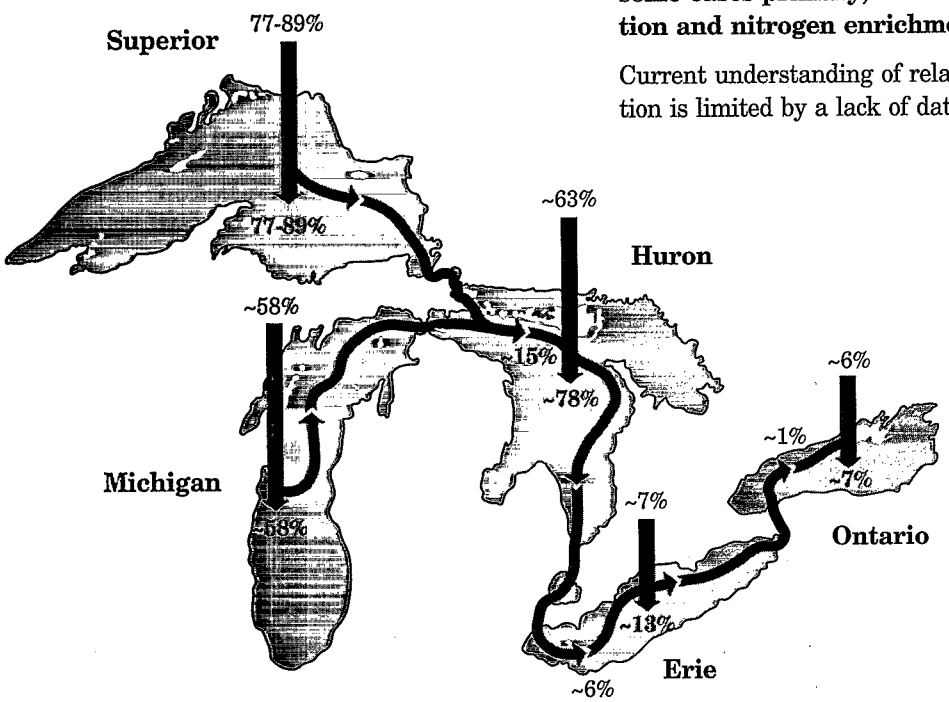


Figure 14. Atmospheric loading of PCBs to the Great Lakes. Arrows and flow depict pollution that deposits from the atmosphere directly to water surfaces and travels through the Great Lakes system. The percentages reflect the amount of such pollution compared to that from all other routes. For example, approximately 63% of Lake Huron's PCB loading is from atmospheric deposition to the lake itself and approximately 15% is from atmospheric loading to the upstream lakes. The remainder of Huron's PCB loading is from nonatmospheric sources (approximately 22%). Data taken from reference 6.

2. **The relative importance of atmospheric loading for a specific chemical in a given waterbody depends on characteristics of the waterbody, properties of the chemical, and the location of sources.**

Broad generalizations concerning the relative importance of atmospheric deposition are not possible because loadings depend on numerous chemical-specific and site-specific factors. Chemical-specific factors include pollutant form, persistence, and bioaccumulation

Table 7. Contribution of Atmospheric Deposition to Total Loadings of Pollutants of Concern for Selected Waterbodies

Waterbody	Lead (%)	PCBs (%)	POM[a] (%)
Lake Superior	95	76-89	96
Lake Michigan	95	58	96
Lake Huron	95	63	80
Lake Erie		20	79
Lake Ontario		13	72
Mid Atlantic Bight	96		

[a]Measured as benzo(*a*)pyrene.

NOTE: The data in Tables 7 and 8 represent several studies with varying margins of error. The data are limited to pollutants for which relative loading estimates exist. Also, it is important to consider the magnitude of inputs from sources other than the atmosphere when comparing relative loadings. For example, if a lake is polluted heavily by riverine inputs, its atmospheric contribution will be a smaller percentage of the total. Likewise, a remote lake with no other inputs will have a high atmospheric component, even if the overall sum of pollutant inputs is small.

Table 8. Contribution of Atmospheric Deposition to Total Loadings of Nitrogen for Selected Waterbodies

Waterbody	Nitrogen (%)
Baltic Sea	25
Chesapeake Bay	25-40
Delaware Bay	14-25
Laholm Bay, Sweden	7
Narragansett Bay	12
New York Bay	10
Ocholockonee Bay, FL	100
Potomac River	28
Rehoboth/Indian River Inland Bays, DE	8
Rhode River, MD	40

potential, all of which affect transport processes, mobility, residence times, and toxicity. Site-specific factors include the hydrology of a given waterbody (e.g., river inflows and outflows, size of drainage areas) and the location relative to natural and anthropogenic sources of air pollution. All of these factors must be considered together to develop specific conclusions for particular chemicals and waterbodies.

3. **Chemicals in the environment may cycle between soil, air, water, and biota for many years.**

The amount of PCBs and other persistent semivolatile organic compounds in air over the upper Great Lakes has not changed much since the late 1970s, suggesting a continuing transfer between the atmosphere and the terrestrial reservoir of these contaminants. Because of the reservoirs of persistent chemicals, cycling between environmental media occurs and results in continued atmospheric deposition of certain organic chemicals without additional inputs. Cycling of pollutants will continue for some time into the future. Similarly, mass balance studies show that significant reservoirs of mercury reside in fish, soils, and bottom sediments of waterbodies, and these studies indicate that mercury may continue to cycle among environmental media for years to come.

4. **When possible, relative loadings to the Great Waters should be evaluated using a mass balance approach.**

   Mass balance analyses provide an essential framework for determining the relative importance of various input sources and output mechanisms in a waterbody, as well as a process for understanding the transport and distribution of pollutants in the water and estimating the length of time that a pollutant resides in any part of an ecosystem.

   Atmospheric loading information should not be overlooked as a source of information when there are not enough data for a complete mass balance, particularly when atmospheric deposition is suspected of being a significant contributor to total loadings.

Sources: What Sources Are Significant Contributors to Atmospheric Loadings to the Great Waters?

As summarized in Section 3.1, there is clear evidence of exposures and adverse effects associated with toxic pollutants in some of the Great Waters and, as discussed in Section 3.2, it appears that a large amount of certain pollutants enter the water from the atmosphere. The next step is to determine the source of the pollutants that are released to the air, to help identify what, if any, emission reductions may be needed to protect the Great Waters.

A source is any activity at any location that may release pollutants to the air. As illustrated in Figure 15, pollutants may be released to the air by natural sources and by anthropogenic, or manmade, sources. Examples of natural sources include forest fires, volcanoes, windblown dust and soil, and sea spray. Examples of anthropogenic sources include industrial activities (such as waste incinerators, power plants, and chemical manufacturers), pesticide applications at farms, and motor vehicles. Some sources release pollutants to the air from a single point at a fixed location, such as a smokestack at a factory; these sources are commonly referred to as "point sources." Pollutants also may be released over broad areas, called "area sources,"* such as when pesticides volatilize after application to a farmer's field or when smoke rises from a widespread forest fire. Area sources also may include small sources in a given area that are too numerous to be counted individually as point sources. Examples of these are dry cleaning facilities and households, both of which use various chemicals. Sources that are not stationary, such as cars, planes, and other vehicles, are considered "mobile" area sources since they release pollutants to the air while moving.

In general, both local and distant air emission sources contribute to a pollutant load at a given location. For the purposes of this report,

Natural Sources

Anthropogenic Sources

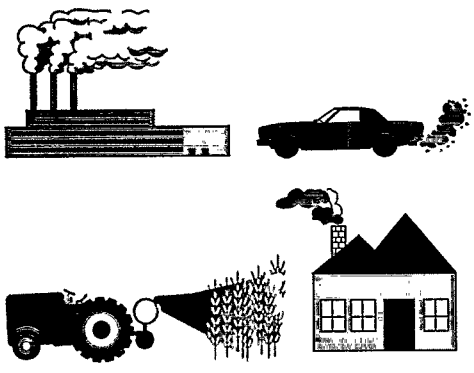


Figure 15. Examples of sources.

*Note that this definition of area sources is different from the definition in the Clean Air Act.

Evaluation of Sources

- **Source characterization:** Identifying and evaluating the sources that emit pollutants of concern to the air.
- **Source apportionment:** Determining the relative contribution of different sources to the air pollution levels at a given location, such as over a waterbody.

local sources are defined generally as those sources located in States and Provinces adjacent to the Great Waters. For example, local sources for the Great Lakes are located in Illinois, Indiana, Michigan, Minnesota, New York, Ohio, Pennsylvania, Wisconsin, and Ontario, Canada. Distant sources are sources located outside the States and Provinces adjacent to the Great Waters. Both local and distant sources include natural and anthropogenic sources and area and point sources.

Determining what sources and source categories (i.e., groups of individual sources having similar activities and air emissions) are significant contributors to atmospheric deposition to the Great Waters requires two basic tasks. First, sources must be characterized to identify what the sources are, where they are located, and what pollutants they emit. Source characterization includes identifying all sources that release a given chemical, grouping sources into categories, measuring or estimating the amounts of chemicals released to the air from individual sources, and examining the relative importance of each source category in the total atmospheric loading of a given chemical. Source characterization also includes analyses of the identity, form, and relative concentrations of chemicals released from a source, information that can be used to link pollutants found in the air with a particular source or source category.

Second, air pollution levels at a given location, such as over a waterbody, must be apportioned to various sources that may have emitted the pollutants. Linking air pollution with sources is a complicated task, as air pollution at any one location usually consists of a mixture of chemicals released from many sources, including some that are nearby and others that are far away. Source apportionment is further complicated by complex and ever-changing weather conditions, variations in emissions from a given source, and changes in the chemical and physical forms of pollutants as they move through the atmosphere. Scientists have developed and are using a variety of mathematical models as tools for determining what sources are contributing most to air pollution levels at a given location, such as over the Great Waters.

In the remainder of this section, important sources of air pollutant emissions are identified and an evaluation is made of which sources contribute significantly to atmospheric deposition to the Great Waters.

The section begins with a review of the current understanding of source characterization and source apportionment. Source case studies are then presented, followed by conclusions for the Great Waters.

Current Understanding of Sources of Air Pollutants

Over the last several years, a large amount of information has been collected to identify the major anthropogenic sources of toxic air pollutants. Substantial new information is currently being collected by EPA to identify and inventory sources in the United States that emit mercury and other pollutants to the air. Nevertheless, at present, a

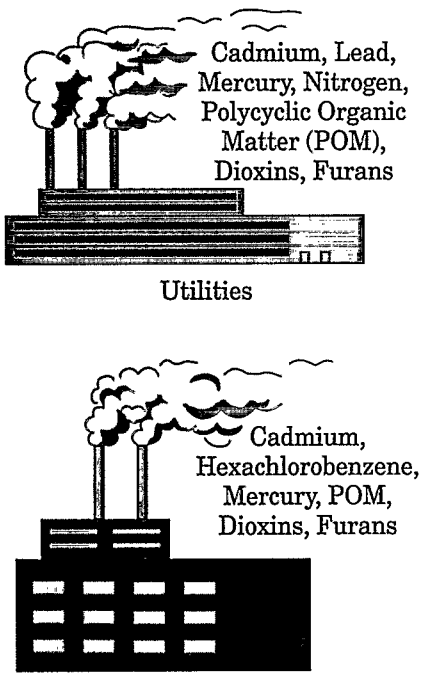


Figure 16. Pollutants of concern emitted from selected sources.

complete and comprehensive inventory of the locations of particular sources and the amounts of individual toxic pollutants that each source emits to the air is lacking. This basic source characterization information is needed to predict the transport of toxic air pollutants from sources to the Great Waters and also to apportion existing air pollution levels in the vicinity of the Great Waters to sources. In addition, there is considerable uncertainty regarding the relative importance of local and distant sources to air pollution levels over the Great Waters. Answers to five basic questions that summarize current scientific knowledge about these sources are provided below.

1. **What Sources Emit Great Waters Pollutants of Concern to the Air?**

   Many source categories of air pollutants of concern for the Great Waters have been defined, and chemicals associated with each source category have been identified. These categories include primarily industrial activities and processes involving combustion. Less information is available on natural sources and some types of area sources, although several of these sources are known to emit certain pollutants of concern. In some cases, there also is limited understanding about the original source of certain pollutants that appear to be originating from natural sources. For example, emissions of mercury from soils have historically been classified as natural sources when, in fact, a substantial portion of the mercury may have been deposited to soils originally from human activities and then resuspended in the air. In cases such as this, the current source (i.e., the soil) may be distant from, and have quite different release characteristics from, the original anthropogenic or natural source. Table 9 identifies key sources known to emit pollutants of concern, and Figure 16 identifies several pollutants of concern emitted from three important source categories: utilities, municipal waste combustors, and motor vehicles.

2. **How Good Are Available Emissions Data for the Various Sources?**

   The quality of available data on the emission of toxic air pollutants from individual sources varies widely. For some source categories, such as chemical manufacturing, data on the composition and rate of emissions are of good to excellent quality. For other sources, data on the types and rates of pollutants emitted are incomplete, and emission estimates are crude. For example, the rates of emissions of metals and persistent organic compounds in the States around the Great Lakes, Lake Champlain, and Chesapeake Bay are difficult to assess because of the diversity of emissions data reported by several research groups. For most of the metals, emission estimates differ by a factor of 10 or more and are continually being revised.

Table 9. U.S. Sources of Air Pollutants of Concern[a]

Pollutant	Sources of Air Emissions
Cadmium and compounds	Fossil fuel combustion; aluminum production; cadmium, copper, lead, and zinc smelting; iron and steel production; battery manufacturing; hazardous waste and sewage sludge incineration; municipal waste combustion; petroleum refining; lime manufacturing; cement manufacturing; pulp and paper production; combustion of waste oil; pigment manufacturing; soil-derived dust; volcanoes.
Chlordane	Insecticide application[b]; volatilization from soils, water, and treated building foundations due to past insecticide application; suspension of eroded soil particles.
DDT/DDE	Insecticide application[b]; volatilization from soils and water due to past insecticide application.
Dieldrin	Insecticide application[b]; volatilization from soils and water due to past insecticide application.
Hexachlorobenzene	Manufacture of chlorine and related compounds; combustion of materials containing chlorine; pesticide manufacturing; municipal waste combustion; fungicide application[b]; volatilization from soils and water due to past fungicide application.
α-HCH	Insecticide application[b]; volatilization from soils and water due to past insecticide application.
Lindane	Insecticide application[b]; volatilization from soils and water due to past insecticide application.
Lead and compounds	Fossil fuel combustion; aluminum production; lead smelting; ferroalloys production; iron and steel production; battery manufacturing; hazardous waste and sewage sludge incineration; municipal waste combustion; petroleum refining; lime manufacturing; cement manufacturing; asphalt and concrete manufacturing; pulp and paper production; combustion of waste oil; paint application[b]; motor vehicles[b]; forest fires; suspension of eroded soil particles; volcanoes.
Mercury and compounds	Fossil fuel combustion; copper and lead smelting; hazardous waste, municipal waste, medical waste, and sewage sludge incineration; lime manufacturing; cement manufacturing; chlorine and caustic soda manufacturing; paint application[b]; suspension of eroded soil particles; evasion from soils and water; volcanoes.
PCBs	Incineration and improper disposal of PCB-contaminated waste; disposal of waste oil; malfunction of PCB-containing transformers and capacitors; electrical equipment manufacturing; pulp and paper production; volatilization from soils and water; municipal solid waste incineration and unregulated combustion.
Polycyclic organic matter	Combustion of plant and animal biomass and fossil fuels; municipal waste combustion; petroleum refining; steel production; coke byproduct recovery; aluminum production; plywood and particle board manufacturing; surface coating of auto and light duty trucks; asphalt processing; dry cleaning (petroleum solvent); fabric printing, coating, and dyeing; forest fires.
2,3,7,8-TCDF	Hazardous, industrial, and medical waste and sewage sludge incineration; municipal waste combustion; combustion of fossil fuels and organic materials containing chlorine; byproduct of various metals recovery processes, such as copper smelting; accidental fires of treated wood products and PCB-containing transformers and capacitors; improper disposal of certain chlorinated wastes; pesticide production, application, and spills; pulp and paper production; volatilization from, and erosion of, dust from landfill sites; forest fires.
2,3,7,8-TCDD	Hazardous, industrial, and medical waste and sewage sludge incineration; municipal waste combustion; combustion of fossil fuels and organic materials containing chlorine; byproduct of various metals recovery processes, such as copper smelting; accidental fires of treated wood products and PCB-containing transformers and capacitors; improper disposal of certain chlorinated wastes; pesticide production, application, and spills; pulp and paper production; volatilization from, and erosion of, dust from landfill sites; forest fires.
Toxaphene	Insecticide application[b]; volatilization from soils and water due to past insecticide application.
Nitrogen compounds	Fossil fuel combustion and other types of combustion; fertilizer application; animal waste.

[a]Data for this table are taken from References 5, 13 through 27, 71, and 72.
[b]Not currently a significant source in the United States due to manufacturing restrictions.

3. What Local Sources Are Important Contributors of Pollutants of Concern to the Great Waters?

Ongoing studies in the United States and Canada have identified and characterized local sources around the Great Waters for many pollutants of concern. Major local sources of metals include fossil fuel combustion in industrial, commercial, and residential units; municipal waste combustion and hazardous waste and sewage sludge incineration; and various manufacturing processes, such as cement production. Polycyclic organic matter originates locally from fossil fuel and biomass combustion, petroleum refineries, motor vehicles, and industrial, commercial, and residential units. Pesticide application is an important local source of pollutants to many Great Waters, such as Chesapeake Bay and other coastal waters. The impact of local sources is also influenced strongly by the location of large point sources relative to the waterbody. For example, 50 of the largest U.S. power plants (as judged by emissions of sulfur oxides) are found in a belt from Missouri to Illinois, Indiana, Michigan, Ohio, West Virginia, and Pennsylvania. Many of these States are adjacent to the Great Lakes region. Lake Champlain and Chesapeake Bay also are located near significant sources.

4. What Distant Sources Are Important Contributors of Pollutants of Concern to the Great Waters?

Although no definitive information exists that indicates in precise quantitative terms the relative contribution of local and distant sources, evidence strongly suggests that distant sources can contribute a significant amount of air pollution over the Great Waters. The extent to which pollutants reach the Great Waters from distant sources depends on many factors, including the height of emission stacks, temperature and velocity of exhaust gases, meteorological conditions, and the physical and chemical forms of the pollutants. Some distant sources believed to be responsible for pollutants deposited to the Great Waters are located in other U.S. regions and also in Canada. A portion of some pollutants over the Great Waters probably originates from sources in other countries. For example, significant amounts of air pollutants are emitted in Mexico from motor vehicles and metals production plants. Also, some pesticides that are present in the Great Lakes are still being used in the Caribbean and Mexico but are restricted from use in the United States and Canada. Pinpointing the sources of some pollutants deposited to the Great Waters is difficult because these pollutants are widely distributed throughout the United States and other countries, even showing up in such remote locations as the Arctic. Pollutants such as PCBs, which exist as particles or in the gaseous phase, may be deposited to the ground and returned to the air many times. This

phenomenon, referred to as the "grasshopper effect," can result in long-distance transport of pollutants and similar background concentrations of air pollutants worldwide.

5. Are Sufficient Data and Techniques Available to Relate Air Pollution Levels Over the Great Waters to Particular Sources or Source Categories?

There is a general understanding of the major factors that affect the transport of air pollutants between their release from the source and their downwind locations, as well as a general understanding of how these factors interrelate. Based on this knowledge, a variety of techniques have been developed that can, in limited instances, relate air pollution levels over the Great Waters to particular sources. Primary source apportionment techniques include "dispersion" models, "receptor" models, and "hybrid" models (see Table 10). In most cases, these techniques have not been fully validated, and they lack complete and reliable input data, such as sufficiently detailed data on the composition and rate of emissions from some sources. Source apportionment methods and capabilities are especially weak for air pollutants that are widespread in the environment, travel over long distances, and/or are emitted in large quantities from natural sources or broad area sources.

Table 10. Source Apportionment Techniques

Type of Model	Description
Dispersion	Traces pollutants from sources to the air at various locations, such as over waterbodies and land. Can quantify relative contributions from a particular point source. Uses data on meteorological conditions and the amount of pollutants emitted from particular sources to evaluate pollutant dispersion in the atmosphere and estimate the airborne concentrations at locations of interest. Requires detailed data on the location and emission characteristics of various sources, which are often lacking. Historically, the primary tool for linking sources to air pollution levels, although limitations of dispersion models have led to the development of receptor models.
Receptor	Traces pollutants in the air at various locations, such as over waterbodies and land, back to particular source types. Can estimate contributions from a group of sources with similar emissions rather than relative contributions from a particular source. Uses data on the air pollution characteristics at the location of interest and on the composition of source emissions (not including data on meteorological conditions) to determine the likely responsible sources based on the premise that sources can be identified by unique emission characteristics (such as the forms and relative amounts of individual pollutants). The effectiveness of this modeling approach is often limited by a lack of adequate "source profile" data (sometimes called "source signatures") that allow air pollution to be linked to particular source types.
Hybrid	Similar to receptor models, but also consider meteorological data. Important for assessing how much air pollution over a receptor comes from distant sources.

Source Case Studies

This section presents source case studies for four pollutants of concern in the Great Waters. These cases identify the principal local and, when possible, distant sources contributing to air pollution levels.

Fuel Combustion, Especially Residential Wood Burning, is a Primary Source of PAHs

PAHs, a subset of POM, appear to be released from a wide variety of sources. The results of a recent study that analyzed the contributions of various PAH sources in eastern North America are shown in Figure 17. The primary source of PAHs was stationary fuel combustion (nearly

50 percent), which includes commercial and residential wood, coal, oil, and other fuel combustion, as well as electric power generation. In fact, residential wood combustion alone accounted for over 30 percent of the total PAH emissions in this study.

PAH releases in the United States appear largest in a wide diagonal belt extending from southern Illinois to the mid-Atlantic States and southern New England. Many of the largest U.S power plants are found in this belt and are known to emit large amounts of PAHs that could deposit in the Great Lakes, Chesapeake Bay, and other Great Waters. Major aluminum smelters also are significant sources of PAHs; some aluminum smelters are located in States adjacent to and near the Great Lakes, and others are in States that border Chesapeake Bay.

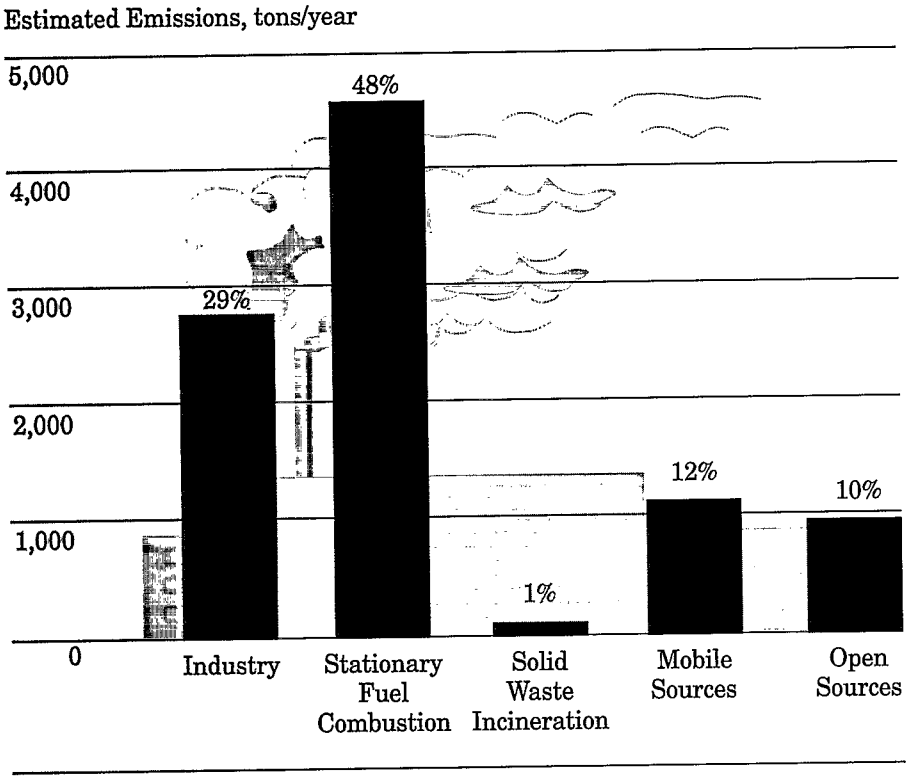


Figure 17. Sources of PAH emissions in eastern North America, 1992.

Other studies have found that the relative importance of particular sources of PAHs can be exceedingly variable. One study in New Jersey, for example, found a large seasonal variation in emissions of benzo(*a*)pyrene, a specific PAH. During the nonheating season, 98 percent of benzo(*a*)pyrene emissions (183 kg) was estimated to come from motor vehicles, while during the heating season, 98 percent of benzo(*a*)pyrene emissions (6,135 kg) was from residential wood burning. Another study found that within a particular source category (such as primary metals production), and even within a specific industry (such as aluminum reduction facilities), PAH emissions at individual facilities typically vary by an order of magnitude or more, depending on the process and raw materials involved.

Lindane Is Emitted from Sources Outside North America

The pesticide lindane, which consists mostly of γ-hexachlorocyclohexane (γ-HCH), demonstrates the importance of global sources and long-distance transport in atmospheric deposition. Though lindane is available in both the United States and Canada, its use as a pesticide is severely restricted. Yet lindane is still found in the Great Waters. Based on investigations of global wind patterns and lindane concentrations in the Arctic, it has been hypothesized that lindane deposited to the Great Waters may originate from as far away as India, China, or the former Soviet Union. Other international regions, such as Latin America, may also be significant sources.

Lindane and other pesticides are used most extensively in Asia in the spring. During spring storms in the Asian deserts, these pesticides can become airborne in the gaseous phase or as very small dust particles and become subject to long-distance transport in the atmosphere. Current understanding of meteorologic processes and wind patterns is consistent with the possibility of lindane-contaminated air from Asia blowing over the Pacific Ocean to the United States and beyond. Such long-distance transport of lindane, however, has only been hypothesized at this time and further study would be needed to obtain evidence of long-distance transport. During atmospheric transport, some of the γ-HCH in lindane naturally changes into another chemical form, α-hexachlorocyclohexane (α-HCH). Measurements of the relative amounts of γ-HCH and α-HCH can be used to evaluate how long the lindane has been in the air and how far it has travelled from possible sources.

Fossil Fuel Combustion and Motor Vehicles Are Major Sources of Nitrogen Emissions

Nationwide studies have determined that nitrogen is released to the atmosphere in various forms from a wide variety of industrial, commercial, and residential fuel combustion sources. As shown in Figure 18, the two primary U.S. sources of nitrogen oxide (NO_x) emissions in 1990 were stationary fuel combustion (such as power plants) and motor vehicles.[40] Trends from 1982 to 1991 indicate that emissions from mobile sources have decreased, while emissions from stationary fuel combustion have increased. No cap on NO_x emissions is required by the 1990 Amendments. Therefore, with continued growth, it is probable that NO_x emissions will start increasing again after the turn of the century.[73]

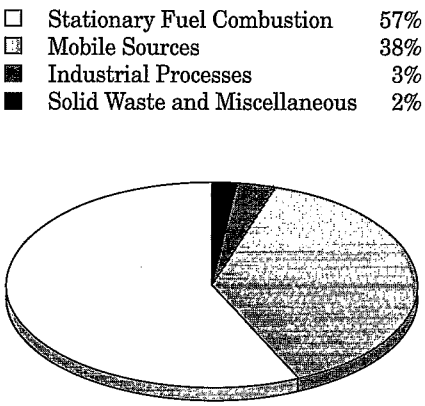


Figure 18. Anthropogenic sources of nitrogen oxide emissions in 1990.

Separate studies in the Chesapeake Bay region yield the same basic conclusion about nitrogen sources. One study determined that the majority of nitrogen compounds in air over Chesapeake Bay are emitted from power plants and motor vehicles.[40] Many large power plants are found in the mid-western, eastern, and southern parts of the country, with a higher density of plants in a few regions, including the region west and south of Chesapeake Bay.

Many Mercury Sources Are Located Outside the Great Lakes Region

A number of studies have developed interim emission estimates for mercury in the continental United States. One 1984 study (based on 1980 data) estimated that natural and anthropogenic sources combined release roughly 1,700 tons of mercury to the air each year. More recent EPA studies estimate that anthropogenic sources account for less than one-third of this total.[74] The 1984 study also found that only a small portion of the total release came from States that are adjacent to or near the Great Lakes. Mercury emissions from more distant States, such as those in the Rocky Mountain region and along the Pacific

Ocean, appear to contribute much larger fractions of the total mercury loading to the air. These conclusions are still highly uncertain, however, because dispersion modeling was not performed as part of the study. The authors did estimate that almost 25 percent of airborne mercury comes from unknown locations. As part of the EPA Mercury Study (required under section 112(n)(1)(B) of the Clean Air Act, as amended in 1990), long-range transport dispersion modeling is being performed to address this issue. The findings from this effort will be presented in 1994 in the Report to Congress on the Mercury Study.

Preliminary results also provide an indication of the types of sources that add to mercury levels in air over the Great Lakes. Waste incineration and fossil fuel combustion have been identified as major anthropogenic source categories for mercury emissions in the Great Lakes region. Natural sources also are an important source. One study suggests that a significant portion of the mercury released to air comes from natural sources such as the release of mercury from surface waters and the resuspension of soil particles. It is unclear, however, how much of these "natural" emissions are actually the result of mercury buildup in the environment caused by past anthropogenic releases.

Conclusions Related to Sources

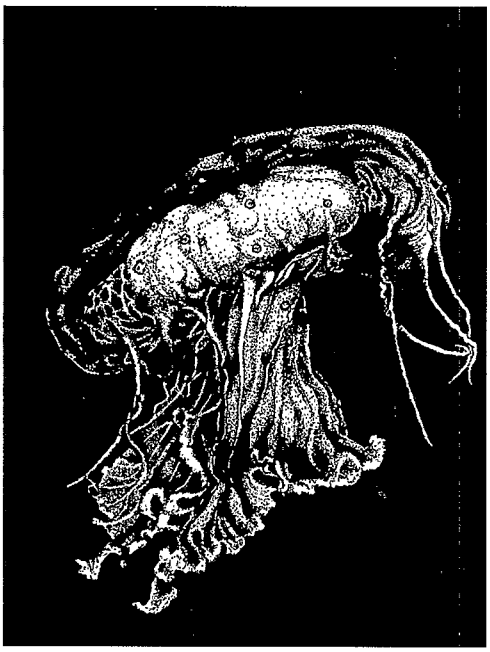
Section 112(m) requires an assessment of air pollution sources that are responsible for the atmospheric deposition of toxic chemicals to the Great Waters. Based on available research findings concerning air emission sources, the following overall conclusions can be drawn.

1. **Although atmospheric deposition appears to be an important way for some pollutants to enter the Great Waters, identifying and characterizing the specific sources that emit the pollutants is difficult.**

   In general, major sources that emit pollutants of concern to air are reasonably well known, although reliable emissions data are frequently lacking. Characterizing the emissions from a given source through measurements is often very expensive and, in some cases, extremely difficult to perform. As a result, emissions are frequently determined by estimation techniques, which are less accurate and frequently yield widely varying results across different research groups. The current lack of reliable emission data and lack of detailed emission inventories severely limits the ability to link toxic air pollution over the Great Waters to particular sources.

2. **The specific sources and source categories contributing to atmospheric deposition to the Great Waters are not well known.**

   Many of the sources of many of the pollutants of concern have been identified. Yet, to adequately identify the specific sources or source categories responsible for particular air pollutants deposited to the



Great Waters, more complete and accurate data are needed to evaluate both the emissions from individual sources and the concentrations and deposition rates of airborne pollutants over a waterbody. Source apportionment techniques also need continued modification, improvement, and verification.

3. **Atmospheric loadings to the Great Waters may be derived from local, regional, and global sources.**

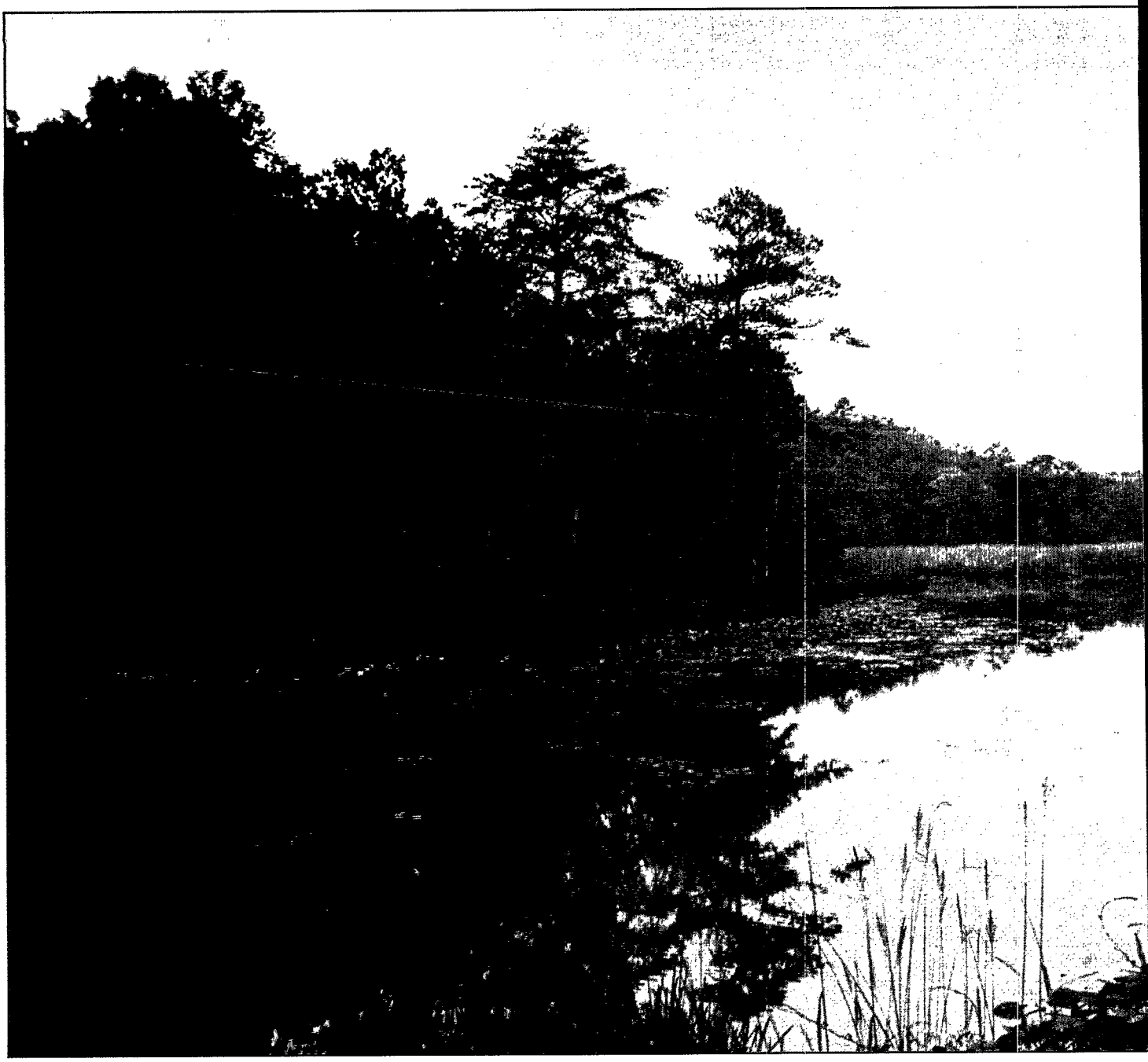
   Among other evidence, the observed presence of current-use pesticides on untreated crops adjacent to treated fields demonstrates the importance of local release and transport over short distances. The importance of regional sources and cycling to mercury deposition has been demonstrated in both North America and Europe. The presence of persistent organic compounds in the Arctic and Antarctic is evidence that long-distance transport in air and subsequent deposition is an important global pathway for these chemicals. Similarly, atmospheric mixing allows northern hemispheric emissions of elemental mercury to be transported to the southern hemisphere, leading to elevated mercury concentrations in fresh water and marine fish in areas far removed from local sources.

4. **The relative contribution of local sources and distant sources to atmospheric deposition to the Great Waters is uncertain.**

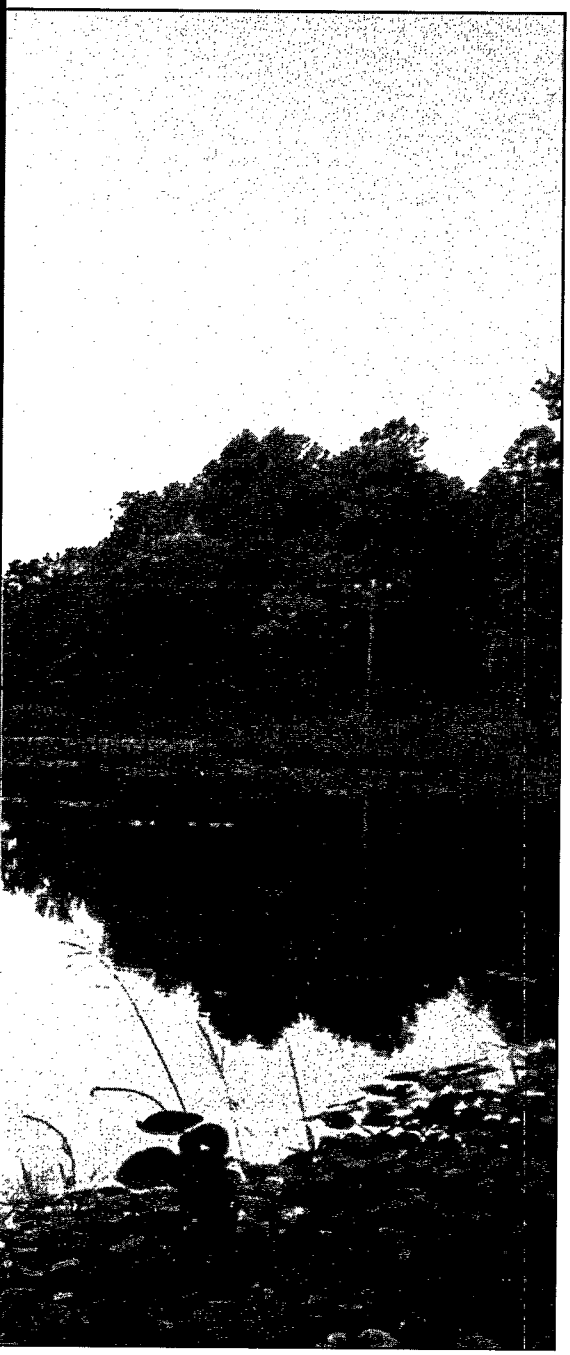
   Available emissions data indicate that many pollutants of concern originate from some sources that are near particular Great Waters, as well as some sources that are located in distant regions. The tendency for pollutants to deposit near their source or to move long distances in the atmosphere depends on a number of factors, including the height of release points, the temperature and velocity of emissions, meteorological conditions, and the physical and chemical forms of the pollutants. Results from several recent studies allow for some generalizations for certain pollutants, but more research is needed to determine how much of the pollution deposited to the Great Waters from the air comes from local versus distant sources.

5. **The environment may act as an important reservoir or source of persistent contaminants that have been released to air previously. Because of pollutant cycling in the environment, atmospheric concentrations of some pollutants may not correspond closely to current source emissions.**

   Understanding the relative contribution of various sources is further complicated by the fact that certain persistent pollutants cycle among air, soils, surface water, sediments, and other environmental media for extended periods. In many cases, it is possible that sizable fractions of some pollutants entering the Great Waters from the air today are not coming from current emissions, but rather are the result of continued cycling in the environment.



Chapter Four
Conclusions and Recommendations



Pollutants emitted to the atmosphere are transported various distances and can be deposited to aquatic ecosystems far removed from their original sources. Scientific studies show that this atmospheric deposition is often an important factor in the degradation of water quality and associated adverse health and ecological effects.

The potential for air pollutants to affect water quality has become increasingly apparent over the last two decades. Stringent controls placed on direct discharges to surface waters have had significant but limited results, making diffuse, or nonpoint, sources of water contamination more important by comparison. Water quality programs have begun to address such nonpoint sources as agricultural and urban runoff. Studies evaluating the loading of pollutants through atmospheric deposition indicate that the atmosphere must also be considered a nonpoint source of pollution that must be controlled in order to meet water quality goals.

Concern about the impact of atmospheric deposition on water quality was the basis for the inclusion of section 112(m) (i.e., the "Great Waters" provision) in the Clean Air Act, as amended in 1990 (1990 Amendments). The purpose of this section of the 1990 Amendments is to evaluate the impact of hazardous air pollutants on the Great Lakes, Chesapeake Bay, Lake Champlain, and coastal waters and to determine whether further reduction of HAPs is needed to prevent serious adverse effects on human health and the environment. To determine if further action is needed, several steps in the decision process are necessary and are inherent in the report requirements posed in section 112(m). Essentially, the questions that must be addressed are:

1. Do HAPs deposited to aquatic systems cause or contribute to serious adverse effects to human health or serious or widespread adverse effects to the environment or does atmospheric deposition of hazardous air pollutants result in exceedances of water quality standards or criteria?

2. If adverse impacts are occurring, what proportion of the exposure is due to airborne pollutants as opposed to waterborne pollutants?

3. If deposition from the air is significant, what sources are emitting these pollutants?

This report evaluated 15 chemicals that are significant water pollutants on the list of Clean Air Act HAPs (except for nitrogen compounds and dieldrin) and are known to be deposited from the atmosphere. They are:

Cadmium and compounds
Chlordane
DDT/DDE
Dieldrin
Hexachlorobenzene
α-HCH
Lindane
Lead and compounds
Mercury and compounds
PCBs
Polycyclic organic matter
2,3,7,8-TCDF
2,3,7,8-TCDD
Toxaphene
Nitrogen compounds.

Other pollutants will be evaluated in the future and will be included, as appropriate, for recommended action. Likely additions to the list are bioaccumulative chemicals of concern (BCCs) targeted for action by the proposed Water Quality Guidance for the Great Lakes System (58 FR 20802) that have significant atmospheric sources.

4. Will regulations mandated by the 1990 Amendments address these sources adequately, and, if not, what regulatory action is recommended to control these air sources to prevent adverse effects? What regulatory revisions are recommended under other Federal laws?

The requirement to include non-Clean Air Act recommendations makes it clear that the intent is for section 112(m) to identify what needs to be done in a broad arena and with a multimedia approach to prevent adverse effects caused by hazardous air pollutants that have been deposited to significant U.S. waterbodies.

This chapter summarizes findings on the impact of air pollutants on water quality and aquatic resources, discusses EPA rationale for decisionmaking at this time, and presents EPA's recommendations.

Future biennial Great Waters reports to Congress will provide updated scientific information and address further regulatory needs. The Clean Air Act studies on mercury and on electric utilities, with reports to be published in 1994 and 1995, respectively, will further augment information on hazardous air pollutants that are of concern in the Great Waters. Findings from those reports will also be used in subsequent Great Waters reports to Congress.

Conclusions

Water quality conditions for most of the Great Waters have improved substantially over the past two or three decades. This improvement demonstrates the effectiveness of Federal, State, and local programs of environmental legislation and regulation, as well as public and industry cleanup efforts. Most significant are water program accomplishments under the Clean Water Act (CWA) (see sidebar on page 69). Programs under the Clean Air Act (e.g., the phaseout of leaded gasoline), the Toxic Substances Control Act (TSCA, e.g., banning the use of PCBs), the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA, e.g., the banning of DDT), and other Federal laws have also contributed to water quality improvements. A summary of relevant EPA regulatory programs and regulations is provided in Appendix C.

As major reductions in direct discharges to surface waters were achieved, the air contribution to water quality and related ecosystem problems became more apparent. Despite the water quality improvements that have been made, the Great Waters ecosystems are far from fully recovered. In order to attain water quality goals and ecosystem protection, the atmospheric component of the water quality problem must be addressed.

Section 112(m) of the Clean Air Act, as amended in 1990, is only one part of a comprehensive program to reduce emissions of hazardous

Pollutant Reductions Resulting from the Clean Water Act

The States and EPA have implemented many important features of the CWA in the past 20 years, including:

- The development of water quality standards by each State
- A massive construction program for wastewater treatment plants
- Monitoring and reporting on the status of the States' waterbodies
- The National Pollutant Discharge Elimination System (NPDES), a program that requires permits for all dischargers and sets technology-based or water-quality-based effluent limits for toxics from industrial and municipal facilities.

air pollutants, thus reducing pollutant loadings to the Great Waters. In addition to the studies required by the 1990 Amendments, there are significant regulatory requirements for hazardous air pollutants, many of which will reduce emissions of Great Waters pollutants of concern. Some of these requirements include vehicle emission standards, reformulated fuel requirements, NO_x emission control requirements under the acid rain and ozone programs, and emission standards for stationary sources. Appendix D outlines specific activities required under section 112 and lists the Great Waters pollutants that may be affected by these activities. (Appendix E outlines EPA's progress under section 112(m).)

The most important conclusions of this Great Waters report are related to scientific concepts concerning characteristics of the pollutants, their transport through air, and their impacts after being deposited to waterbodies. The atmospheric transport and deposition of pollutants and their potential to affect human health and the environment are concepts that are widely accepted by scientists in the field, yet there is still much to learn. However, some basic information is quite clear and very important in any discussion of these pollutants.

The **effects** that Great Waters pollutants of concern can cause in humans and the environment are fairly well understood. Cancer, developmental effects, neurological effects, and effects on reproduction have been associated with exposure (usually through fish consumption) of humans and other animals to Great Waters pollutants. Though studies relate these cancer and noncancer effects to specific Great Waters pollutants, there are generally insufficient data available to prove the linkage between atmospheric deposition of the pollutants and consequent effects in humans and ecosystems.

Studies of **relative loadings** to waterbodies from atmospheric deposition have demonstrated that atmospheric deposition can be a significant contributor of toxic chemicals to the Great Waters. Studies have demonstrated that, for example, atmospheric deposition is responsible for as much as 77 to 89 percent of the loadings of PCBs to Lake Superior and as much as 40 percent of the loadings of nitrogen to Chesapeake Bay. Yet, the relative importance of atmospheric loading for a specific chemical depends on the interaction of properties of the chemical, weather patterns, and the kind and amount of airborne sources and waterborne sources. Thus, even when data are available for a particular chemical in a particular waterbody, only some of the data can be generalized to other waterbodies or other chemicals.

Many **sources** and source categories of these pollutants have been identified (see chart in Appendix F). However, because atmospheric loadings to any particular waterbody are derived from local, regional, and global sources, determining the particular sources responsible for deposited pollutants is quite difficult. Further data are needed for identification and characterization of the specific sources

responsible for pollutants that are deposited to the Great Waters and, thus, for the adverse effects of these pollutants on human health and the environment. Whether or not effects from exposure to Great Waters pollutants, such as those originating from long-range transport, are sufficient to warrant regulatory action is a question for the risk manager and has not been addressed here.

Further research is needed to better determine the full impacts of these pollutants on human health and the environment and to provide risk managers with sufficient information to ensure that decisions result in pollution prevention and regulatory measures that will yield significant benefits.

The following **conclusions** are based on the currently available scientific data:

1. **Persistence is a critical characteristic of the Great Waters pollutants of concern.** This characteristic allows them to be transported long distances, to remain in the environment for a significant period of time, and to accumulate over time. Therefore, persistent chemicals can be deposited and then reemitted and redeposited many times, resulting in transport over long distances. Their accumulation in the environment over time results in significantly greater exposure potential than for chemicals that degrade in the environment.

2. **The tendency to bioaccumulate is another critical characteristic of pollutants of concern.** This results in potentially greater exposure for predators at the top of the food web, such as eagles and humans. These pollutants are stored in animal tissues and accumulate, which results in biomagnification. That is, at each level of the food web an animal accumulates the chemicals from its diet and passes them along to the animal at the next level of the food web. Top consumers in the food web may accumulate chemical concentrations millions of times greater than that in the water. For example:

   - The cancer risk from eating 1 pound of Lake Ontario trout is greater than the risk from drinking 20 lifetimes' worth of water from Lake Ontario.[75]

   - In the Great Lakes and Lake Champlain, fish consumption advisories have been established for some fish because of unsafe concentrations of chemicals, such as mercury, in the fish. People who consume contaminated fish regularly (such as Native Americans or subsistence fishermen) have greater concentrations of the contaminants in their bodies than other people. Humans generally are exposed to

Jamey Tidwell, Texas Sea Grant

The USA is a signatory of the Great Lakes Water Quality Agreement, which requires that the input of persistent toxic substances to the Great Lakes Basin Ecosystem be "virtually eliminated." In their Fifth Biennial Report in 1989, the International Joint Commission (IJC), an advisory committee comprised of representatives from the United States and Canada, recommended that the Parties (the United States and Canada) complete and implement immediately a binational toxic substances management strategy for accomplishing, as soon as possible, the Great Lakes Water Quality Agreement philosophy of zero discharge. They concluded, on the basis of mounting evidence that "cannot be denied," that "there is a threat to the health of our children emanating from our exposure to persistent toxic substances, even at very low ambient levels." In their Sixth Biennial Report in 1991, the IJC concluded that "because persistent toxic substances remain in the environment for long periods of time and become widely dispersed, and because they bioaccumulate in plants and animals—including humans—that make up the food web, the ecosystem cannot assimilate these substances." These toxic substances, the IJC concluded, "are too dangerous to the biosphere to permit their release in any quantity."

mercury through ingestion of fish and, in the United States, 1 percent of the population has an average daily intake of methylmercury (the most toxic form of mercury) higher than the World Health Organization's suggested "safe level."

- Breast-fed babies are considered to be one level in the food web higher than their mothers. After 6 to 9 months of breast-feeding, the concentration of PCBs in a baby can reach four times that in the mother.[76]

3. **Significant adverse effects on human health and wildlife have been observed** due to exposure, especially through fish consumption, to persistent pollutants that bioaccumulate. These adverse effects range from immune system disease and reproductive problems in wildlife to subtle developmental and neurological impacts on children and fetuses. Often considered to be unrelated, the human and wildlife effects of these chemicals are essentially linked. As EPA's Science Advisory Board pointed out, "most human activities that pose significant ecological risks . . . pose direct or indirect human health risks as well."[12]

   Although research is continuing, the International Joint Commission considered the known effects sufficient reason to adopt a goal of "virtual elimination" of persistent organic chemical releases to the Great Lakes (see sidebar).

4. **Noncancer effects are a significant human health concern.** Most of the chemicals of concern are probable human carcinogens, exposure to which is expected to increase the population incidence of cancer. However, noncancer effects will impact some members of a population exposed to levels that exceed a threshold level. Many of the pollutants of concern are developmental toxicants capable of altering the formation and function of critical body systems and organs. Therefore, **the developing embryo and fetus and breast-fed infants are particularly sensitive to these chemicals.**

   Subtle changes in thought processing and activity levels were observed in one study of children of women who consumed Lake Michigan fish two to three times a month. The ultimate impact of individual impairments such as these can be characterized as a "diminishment of potential" in humans. Exposure to the mother may have been a single large exposure, small cumulative exposures, or long-discontinued exposure and may have caused no visible symptoms in the mother.

5. **Ecological effects are significant and can be subtle**, such as immune function impairment, reproductive problems, and neurological changes that affect survival. Also, these impacts can affect the offspring of the exposed individual and may not be evident until later in life. For example, a study of tern eggs and chicks, contaminated through maternal exposure to PCBs and dioxins, showed a 35 percent mortality rate due to "wasting." The deaths in one study occurred after 17 days, and, in the followup study, the same percentage died after 31 days. Such delayed effects and subtle symptoms can easily be overlooked.

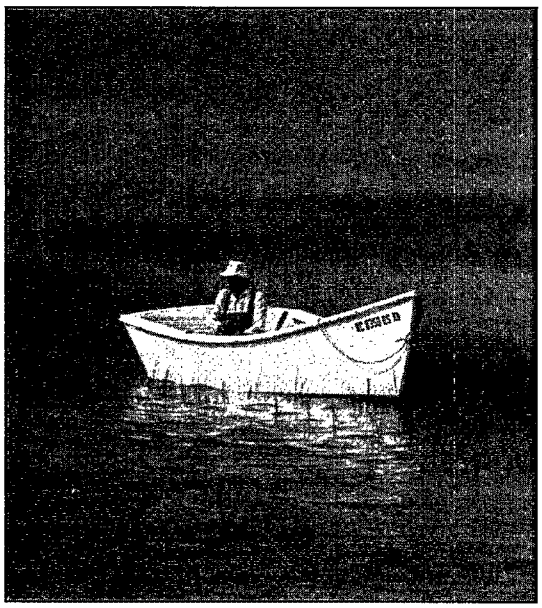
   Other ecological effects are quite obvious and can affect survival of individuals and/or populations. For example, crossed bills, associated with exposure of birds to toxic pollutants, can hinder adequate feeding.

6. **There have been many exceedances of existing water quality criteria and standards** as well as of the proposed Great Lakes Water Quality Criteria (pGLWQC) (see Appendix B). Persistence allows accumulation of a chemical to undesirable levels. The pGLWQCs are particularly important measures of the health of aquatic resources because they incorporate bioaccumulation factors.

7. **Eutrophication, caused by excess nitrogen inputs, is a major problem in U.S. coastal waters,** and, in studied estuaries, the atmospheric contribution to the total nitrogen loading is significant. Nitrogen is the limiting nutrient in most coastal estuaries. Thus, addition of nitrogen in a form usable by plants results in accelerated eutrophication of the waterbody, to the point of causing toxic effects—characterized by additional plant growth, usually of algae, which shades beneficial plants and uses up oxygen during decay. Eutrophication causes ecological effects and economic impacts that range from nuisance algal blooms to oxygen depletion and fish kills.

   In studies of the Chesapeake and Delaware Bays, the atmosphere was estimated to account for 28 to 40 percent of the total nitrogen loading, which contributes to system-wide eutrophication.

8. **Case studies have shown atmospheric deposition to be a major contributor** of mercury, POMs, PCBs, and nitrogen. Available estimates of relative loadings for studied waters and specific chemicals are listed in Tables 7 and 8, page 55.



J. Scott Taylor, Duke University Marine Lab

The relative significance of atmospheric loadings for any specific waterbody is dependent on the amount of waterborne loading to which it is being compared. **The absolute quantity of atmospheric loadings also warrants attention,** especially since loadings of even small amounts of pollutants that bioaccumulate can result in a significant pollutant burden in fish and, ultimately, in humans.

9. **Airborne emissions from local as well as distant sources contribute to pollutant loadings,** through atmospheric deposition, to waters. Transport distances depend on the characteristics of the chemicals and source emissions as well as weather patterns. While the contribution of distant air pollution sources to remote pristine regions, such as the Arctic, is well documented, more data are needed to determine sources and source locations affecting the Great Waters.

10. **Continued research is needed,** especially to help determine atmospheric contributions, to identify sources, to evaluate low-exposure effects, and to target HAPs that pose the most significant risk to human health and aquatic resources.

Conclusions could not be drawn for two areas of concern because of the lack of data. Available data are not sufficient to quantify the overall relative atmospheric loadings (for all of the Great Waters for all of the HAPs). Therefore, relative loading estimates are, and will continue to be, chemical- and waterbody-specific. Neither is it possible to identify the specific sources or source types emitting the pollutants into the atmosphere that are ultimately deposited to the Great Waters (except in localized case studies).

Recommendations and Actions

The goal of section 112(m) of the Clean Air Act, as amended in 1990, as discussed earlier, is to determine if atmospheric inputs of pollutants, and the impacts from these atmospheric inputs to the Great Waters, warrant further reductions of atmospheric releases. If reductions are warranted, a strategy to reduce atmospheric releases of the pollutants must be devised. In making the following recommendations, EPA evaluated the available scientific information and called upon the expertise of its own, as well as outside, scientists. Most important, EPA considered the implications of action and of inaction, while also recognizing that section 112(m) of the 1990 Amendments mandates a preventive approach, stating that EPA should act to "prevent" adverse effects and to "assure protection of human health and the environment."

In the 1992 amendments to the Chesapeake Bay Agreement, the Governors of Virginia, Maryland, and Pennsylvania; the Mayor of the District of Columbia; the EPA Administrator; and the Chesapeake Bay Commission chair committed "to incorporate, into the Nutrient Reduction Strategies, an air deposition component which builds upon the 1990 Amendments to the Federal Clean Air Act and explores additional implementation opportunities to further reduce airborne sources of nitrogen entering Chesapeake Bay and its tributaries."

EPA's recommendation is that reasonable actions are justified, based on evaluation of the scientific information currently available, and should now be taken and that research should continue. NOAA concurs with the principles of this policy.

Although there are significant uncertainties in the information available, there is enough convincing evidence to prompt action. Effects documented for some toxic, persistent, bioaccumulative pollutants are of concern in the Great Lakes, at least, and have prompted strong statements from the International Joint Commission. Similarly, impacts from nitrogen loading to Chesapeake Bay led the Chesapeake Executive Council to address air sources in the development of tributary basin nutrient reduction strategies (see sidebar).

"If we wait until we have all of the answers to act," many argue, it will be too late to fix the problem. In addition, some argue that further contributions of persistent bioaccumulatable pollutants add to an environmental burden that is already causing effects. EPA believes it is important to balance our present understanding of atmospheric deposition against the implications of inaction in order to define those actions that are justified at this time. EPA is committed to protecting public health and the environment and will take whatever regulatory actions are appropriate in the most cost-effective way possible. Also, EPA must target that research that is necessary to define the necessary actions. (EPA's research needs and plans are discussed in Appendix G.)

Specifically, EPA is recommending a series of actions. Because of the uncertainties, the actions EPA proposes are not specific to sources but, rather, are targeted for those chemicals about which there is evidence of potentially significant health and environmental risks. The recommended actions are focused especially on utilizing regulatory mechanisms in the Clean Air Act that address the most hazardous chemicals. EPA believes that the characteristics of toxicity, persistence, and the tendency to bioaccumulate, along with exposure and the significance of adverse effects, warrant special treatment for the Great Waters chemicals of concern, such as outlined below. EPA also believes that this treatment is consistent with the congressional intent for those regulatory mechanisms and for section 112(m).

The recommended actions also promote an integrated effort to assess the problem and reduce pollution. EPA recognizes that pollutants are transferred continuously between air, water, and land and that, to adequately address pollution problems, multimedia, multiagency approaches must be explored. The recommendations reflect an integrated effort, and plans for future Great Waters program activities reflect increased integration within and outside the Agency.

EPA continues to recommend pollution prevention as the best option for reducing or eliminating the input of these chemicals to the environment and advocates the voluntary reduction of any HAPs.

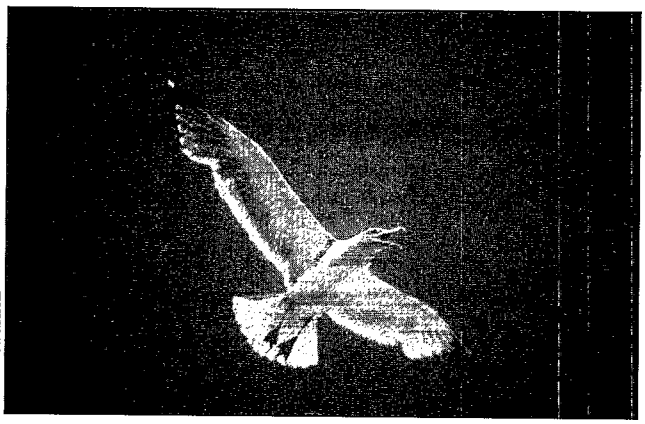
Consistent with the above, the recommendations for action, outlined below, are divided into three strategic themes.

Strategic Themes and Actions

1. **EPA will continue ongoing efforts to implement section 112 and other sections of the Clean Air Act, as amended in 1990, and will use the results of this report in taking reasonable actions to reduce emissions of Great Waters pollutants of concern.**

Action Items

a. EPA is developing standards under section 112(d) for approximately 35 source categories of Great Waters HAPs of concern, consistent with the schedule published in response to section 112(e)(3). Where possible, given other factors, EPA will publish section 112(d) standards ahead of schedule for specific source categories. Great Waters Program funds will be used to develop and publish ahead of schedule section 112(d) standards for at least one source category.



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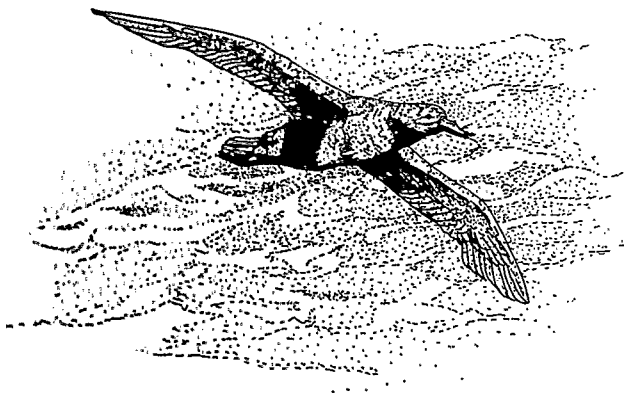
b. During the process of developing emission standards, EPA will evaluate whether the currently defined MACT floor for existing sources represents a sufficient level of control for sources that emit Great Waters pollutants of concern.

c. As soon as practicable, EPA will publish an advance notice of proposed rulemaking to notify the public of EPA's interest in establishing lesser-quantity emission rates (less than 10 tons per year) for selected Great Waters HAPs for the purpose of defining sources emitting these HAPs as "major sources" and to solicit comment. EPA will also evaluate whether any Great Waters HAPs warrant establishment of an LQER, and, if appropriate, based on that evaluation and the comments on the ANPR, EPA will develop a notice that proposes LQERs for those pollutants for which an LQER is warranted.



d. During the process of standards development for major sources, EPA will determine whether area sources of Great Waters HAPs warrant regulation under section 112(d) and, if so, which area sources. Results of the assessment will be integrated into the strategy for area sources under development in accordance with section 112(k).

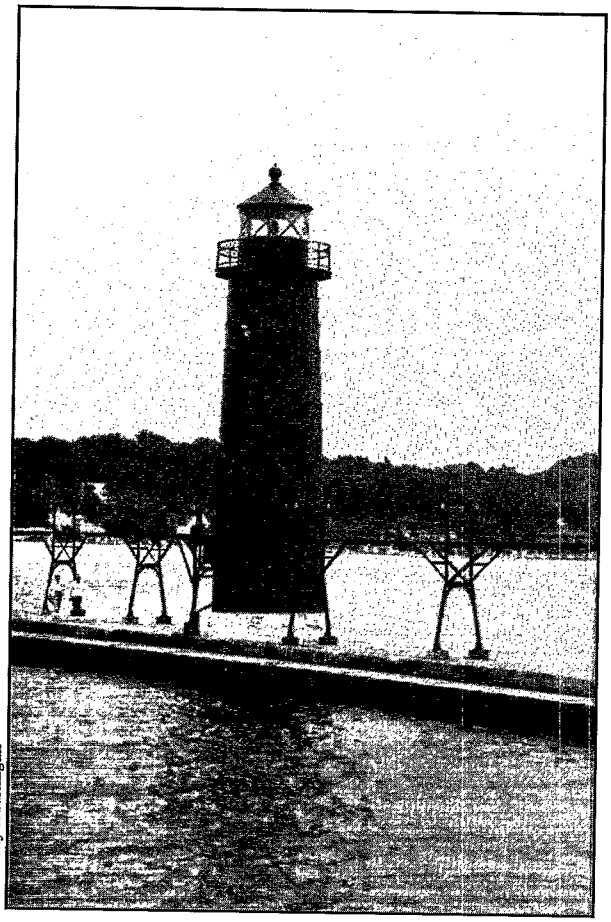
e. For the urban area source strategy (section 112(k)), EPA will evaluate public health effects on the basis of total exposure, which would include exposure by inhalation as well as exposure through ingestion of food containing bioaccumulated urban toxicants.

f. EPA will conduct a pilot project examining the use of Great Waters impacts analyses in the development of section 112(d) standards.

g. For Great Waters HAPs, EPA is proposing a cap (i.e., 0.01 ton/year) to the de minimis levels being developed under section 112(g), so that controls would be required for more sources of Great Waters HAPs as they modify their processes. EPA will determine the appropriate de minimis level on a chemical-by-chemical basis, giving consideration to the chemical's persistence, propensity to bioaccumulate, and such other factors that EPA considers relevant.

h. EPA plans to propose a revised municipal waste combustor rule, with stringent controls on mercury emissions and emissions of other Great Waters HAPs, not later than summer 1994.

i. EPA is conducting studies that will provide information for future Great Waters reports. The mercury study, under section 112(n)(1)(B), will evaluate the rate and mass of mercury emissions from all sources, the health and environmental effects of such emissions, technologies to control such emissions, and the costs of these control technologies. The utility study, under section 112(n)(1)(A), will evaluate the hazards to public health reasonably anticipated to occur as a result of emissions of all HAPs by electric utility steam-generating units. Findings of these studies will be relied upon in future Great Waters reports in the development of strategies for reducing environmental exposures to Great Waters pollutants.



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j. EPA is developing ecological effects assessment screening methods for reviewing petitions to add and delete pollutants from the HAP list and to delete source categories from the source category list. EPA will consider the Bioaccumulation Factor Methodology (58 FR 20802) in the development of these ecological effects assessment methods. The purpose is to help ensure that ecological effects, in addition to health effects, will be considered in determining whether regulation is warranted.

k. EPA will evaluate whether other pollutants, including hexachlorobutadiene and methoxychlor, which are proposed Bioaccumulative Chemicals of Concern under the proposed Water Quality Guidance for the Great Lakes System (58 FR 20802) and which have been identified as having potentially significant air sources, should be added to the list of Great Waters pollutants of concern.

l. EPA is continuing to emphasize pollution prevention as the goal in the development of control measures to reduce emissions of Great Waters pollutants of concern and is encouraging any voluntary pollution prevention and other emission reduction efforts.

m. In the development of regulations and pollution prevention or reduction strategies under the 1990 Amendments, EPA will examine the potential for reductions of oxides of nitrogen and will determine how additional NO_x reductions can be achieved for protection of coastal water quality and related resources.

n. EPA will develop Achievable Control Technology documents (ACTs) for NO_x. This is expected to result in nationwide NO_x emissions reductions, thus protecting coastal waters, as States develop regulations under the National Ambient Air Quality Standards program.

2. EPA recognizes the need for an integrated multimedia approach to the problem of atmospheric deposition of pollutants to waterbodies and, therefore, will consider authorities beyond the Clean Air Act to reduce human and environmental exposure to Great Waters pollutants of concern.

Action Items

a. EPA will establish a funding and operational mechanism for all appropriate offices to pool their resources (both dollars and personnel) to more effectively and efficiently manage this multimedia problem. The Great Waters Core Project Management Group will serve as the liaison among EPA's Assistant Administrators (AAs) and Regional Administrators (RAs). Through this group, commitments will be obtained from each of the AAs and RAs to earmark funds for implementing the recommendations of this report or to take a lead role in the implementation of specific recommendations.

b. EPA should use the discretionary authority in existing statutes to regulate or prohibit multimedia environmental releases that cause or contribute to a water quality impairment. The Administration wants to work with Congress (e.g., on Clean Water Act reauthorization) to develop approaches that would allow effective pollution control where other Federal environmental statutes are not effective and where an integrated multimedia approach is the most efficient means to reduce unacceptable risk. This would not apply to mobile sources or pesticide programs. EPA would use the most appropriate existing environmental statute (e.g., the Clean Air Act for air releases) for controlling the release and would take into account the factors of revised section 307(a)(2) of the Clean Water Act.

c. Congress, with technical support from EPA, should develop legislation to prohibit the exportation of any pesticide product which contains an active ingredient that has been banned for all or virtually all uses in the United States. The recommendation to prohibit the export of banned pesticides was presented in the *Report of the National Performance Review: Creating a Government That Works Better and Costs Less.*[77]

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d. EPA will work with other countries to explore possible alternatives to reduce or eliminate the production, export, and use of pesticides banned in the United States.

e. EPA will explore the feasibility of creating an inventory of pesticide use within the United States and of establishing a program to identify and quantify stockpiles and emissions of pesticides of known and potential concern, including banned pesticides.

f. EPA will continue to emphasize pollution prevention as a goal and to encourage voluntary pollution prevention efforts that lead to reductions in releases of Great Waters pollutants of concern. Several pollution prevention projects that address Great Waters pollutants of concern are currently under way:

- A "Virtual Elimination Pilot Project" is under way in the Great Lakes Basin, as a part of a comprehensive toxics reduction effort. The Virtual Elimination Pilot Project proposes selecting a small group of toxics as a pilot and performing an in-depth analysis of opportunities for reduction from all sources.

- EPA has initiated a project to reduce risks from PCBs by asking all utilities in the Great Lakes area to voluntarily decommission their PCB electrical equipment.

- The Lake Superior Pollution Prevention Strategy was released in October 1993 as part of the Lake Superior Binational Program.

- EPA, together with State Departments of Agriculture and local government agencies, has funded a series of "Clean Sweeps" to collect and properly dispose of existing stocks of canceled pesticides from residents in the Great Lakes area.

g. EPA will continue its work with Canada, under the Great Lakes Water Quality Agreement, on airborne toxic substances. These continuing bilateral efforts are assisting and will continue to assist in meeting Great Water program objectives during the 1990s.

h. EPA will distribute technical information to State and local air and water agencies to facilitate cooperative efforts toward common goals to further reduce human and environmental exposure to Great Waters HAPs.

i. EPA will initiate discussions about possible mechanisms that Regional EPA offices and State agencies could use for sharing information on new or renewal permit applications for sources with the potential to emit Great Waters pollutants of concern.

3. EPA will continue to support research activities and will develop and implement a strategy describing necessary research and policy assessments to address the mandates of section 112(m).

Action Items

a. EPA is developing a strategy to target research necessary to answer the scientific questions outlined in section 112(m). The strategy will be reviewed by the EPA Science Advisory Board and will influence decisionmaking on the priority and funding for future research. This strategy will focus on utilization of the mass-balance approach for determining relative loading and will acknowledge the need for a balance between monitoring, modeling, and emission inventory efforts for that work. The strategy will also consider how to better identify those persistent chemicals with the tendency to bioaccumulate that may become problematic if emissions continue. Included in the strategy will be an assessment of the need for development of tools that can be used to: (1) assess and quantify the human health and environmental risk from exposure to air toxics, especially via indirect exposure routes, and (2) quantify the social, environmental, and economic benefits and costs of pollution prevention and regulatory actions.

b. EPA will continue to work with NOAA to pursue the development and application of the appropriate technical tools to further define and estimate loadings to the Great Waters and to identify sources of atmospherically deposited pollutants.

c. Through the use of Great Waters Program funds and other resources, EPA will continue to support those research activities identified as priorities by the research communities and affirmed by the Great Waters Core Project Management Group.

   - EPA will continue work on the characterization of processes and parameters for mass balance modeling and the verification of the mass balance methodology, especially the development of the prototype mass-balance program being conducted in Lake Michigan.

   - EPA will work with State agencies to complete regional emission inventories for the Great Lakes and will complete a national screening level emission inventory for section 112(c)(6) chemicals (a group of six of the Great Waters pollutants), and will identify categories of sources of the specific pollutants listed in section 112(c)(6).

   - EPA will continue source characterization and identification activities.

   - EPA will complete and evaluate mercury screening level deposition models using screening emission inventories and will determine whether to transfer the method to other chemicals and to provide support for other more intensive regional air emission inventory efforts.

   - EPA will continue to support ongoing monitoring efforts.

d. EPA will initiate discussions among the appropriate groups to identify ongoing benefits analysis efforts and human health (cancer and noncancer) and environmental risk assessment efforts within the Agency, in other Federal programs, in other countries, in academia, and elsewhere. The goal is to define more clearly the research/data needs and to develop a long-term plan for developing tools and methods for benefits analyses and risk assessments.

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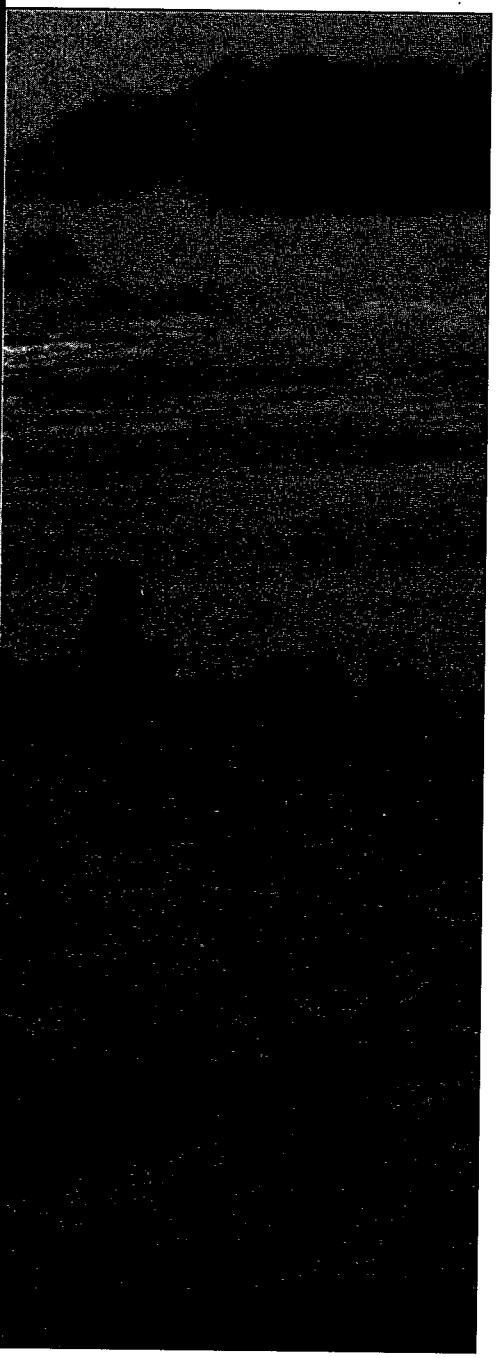
Chapter Five References



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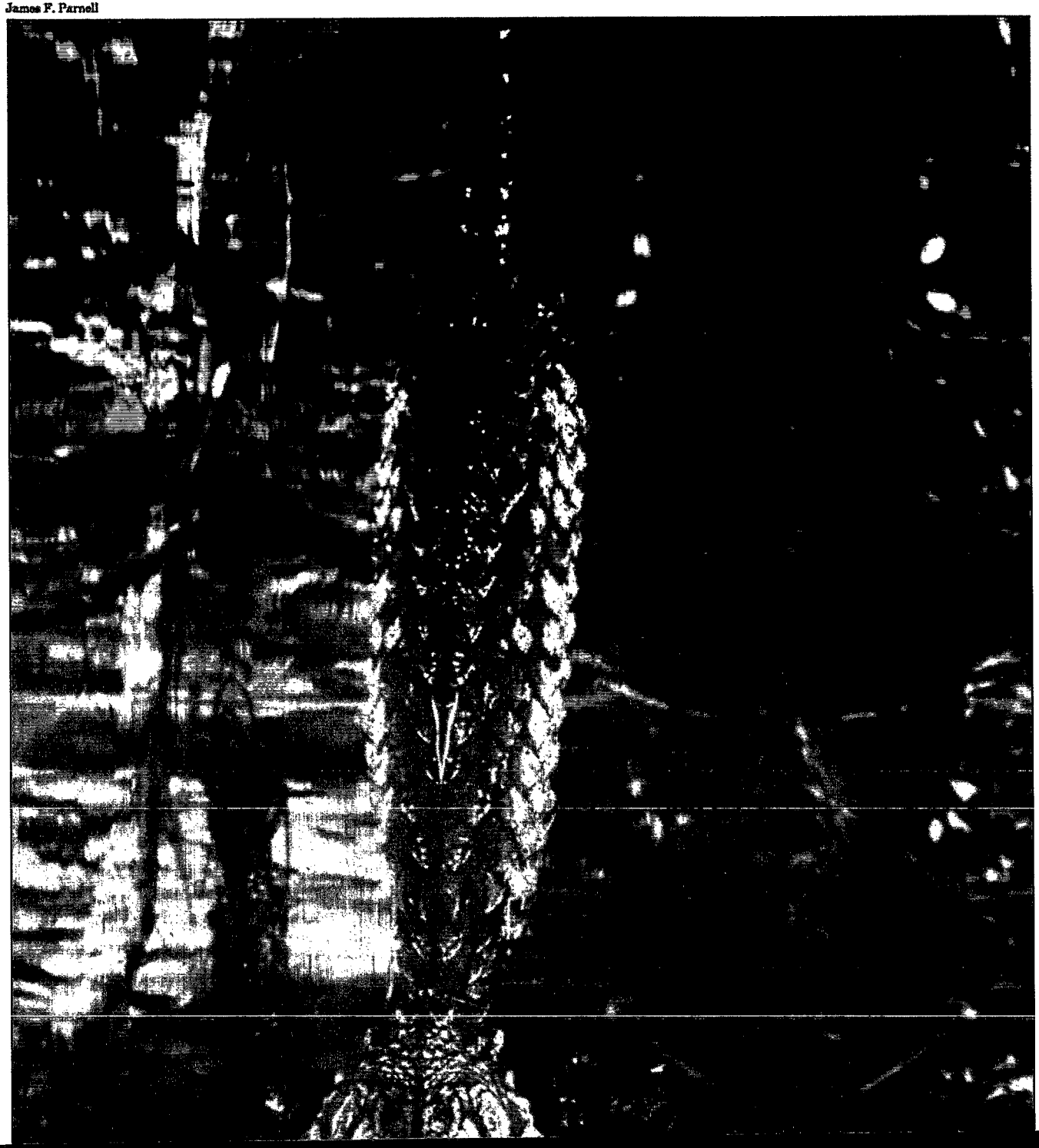

Appendices

Appendix A: Lists of Bioaccumulative Chemicals of Concern and Potential Bioaccumulative Chemicals of Concern

Appendix B: Comparison of Great Lakes Sampling Data to Various Water Quality Benchmarks

Appendix C: Historical EPA Regulations

Appendix D: Summary of Clean Air Act Section 112 Activities

Appendix E: Progress Under Section 112(m)

Appendix F: Summary of MACT Source Categories Potentially Emitting Great Waters Pollutants of Concern

Appendix G: Preliminary Summary of Research and Program Planning

Appendix A

Bioaccumulative Chemicals of Concern	Potential Bioaccumulative Chemicals of Concern
Aldrin	Benzo[*a*]pyrene; 3,4-benzopyrene
4-Bromophenyl phenyl ether	3,4-Benzofluoranthene; benzo[*b*]fluoranthene
Chlordane	11,12-Benzofluoranthene; benzo[*k*]fluoranthene
4,4-DDD; p,p-DDD; 4,4-TDE; p,p-TDE	1,12-Benzoperylene; benzo[*ghi*]perylene
4,4-DDE; p,p-DDE	4-Chlorophenyl phenyl ether
4,4-DDT; p,p-DDT	1,2:5,6-Dibenzanthracene; dibenz[*a,h*]anthracene
Dieldrin	Dibutyl phthalate; di-n-butyl phthalate
Endrin	Indeno[1,2,3-cd]pyrene; 2,3-o-phenylene pyrene
Heptachlor	Phenol
Heptachlor epoxide	Toluene; methylbenzene
Hexachlorobenzene	
Hexachlorobutadiene; hexachloro-1,3-butadiene	
Hexachlorocyclohexane; BHC	
α-Hexachlorocyclohexane; α-BHC	
β-Hexachlorocyclohexane; β-BHC	
δ-Hexachlorocyclohexane; δ-BHC	
Lindane; γ-BHC; γ-hexachlorocyclohexane	
Mercury	
Methoxychlor	
Mirex; dechlorane	
Octachlorostyrene	
PCBs; polychlorinated biphenyls	
Pentachlorobenzene	
Photomirex	
2,3,7,8-TCDD; dioxin	
1,2,3,4-Tetrachlorobenzene	
1,2,4,5-Tetrachlorobenzene	
Toxaphene	

Source: U.S. Environmental Protection Agency, Proposed water quality guidance for the Great Lakes system: Proposed rule and correction, *Federal Register* 58:20802-21047, April 16, 1993.

Appendix B:

Comparison of Great Lakes Sampling Data to Various Water Quality Benchmarks (in ppb)

Comparison of Great Lakes Sampling Data to Various Water Quality Benchmarks (ppb)

Pollutant	National AWQC: Fresh Water Aquatic Life[a]	National AWQC: Human Health[b]	Proposed Great Lakes Water Quality Criteria[c]	Great Lakes Water Quality Agreement Objectives[d]
Cadmium	1.1	10	0.78	0.2
Chlordane	0.0043	0.0046	0.0002	0.06
DDT/DDE	0.001[f]	0.00024[f]	0.00000087	0.003
Dieldrin	0.0019	0.00071	0.0001	0.001[g]
Hexachlorobenzene	—	0.0072	0.0001	—
α-HCH	—	0.092	—	—
Lindane	0.08	0.186	0.7	0.01
Lead	3.2	50	—	10-25
Mercury	0.012	0.144	0.00018	0.2
PCBs	0.014	0.00079	0.000017	—
Benzo(*a*)pyrene (indicator of POM)	—	0.028[h]	—	—
2,3,7,8-TCDF	—	—	—	—
2,3,7,8-TCDD	0.00001	0.00000013	0.0000000096	
Toxaphene	0.0002	0.0071	0.00002	0.008

[a] Values listed are for fresh water chronic criteria except for 2,3,7,8-TCDD, which is the fresh water chronic lowest observed effects level (LOEL). Values for cadmium and lead are hardness dependent (based on 100 mg/L $CaCO_3$).[58]

[b] Values listed are for human chronic exposure through both fish consumption and drinking water; values for potential carcinogens correspond to a 10^{-5} individual cancer risk level.[58]

[c] Values listed are the most stringent (i.e., lowest) among those proposed for protection of human health, aquatic life, or wildlife; values for potential carcinogens correspond to a 10^{-5} individual cancer risk level. Value for cadmium is hardness dependent (based on 50 mg/L $CaCO_3$).[29]

[d] Values listed are for protection of the most sensitive user of the water among humans, aquatic life, or wildlife.[31,59]

[e] Concentrations are the maximum post-1980 open water sampling values reported in References 31, 78, and 79 (sampling data are for 1980-1986). Values in bold indicate exceedance of at least one criterion. Sources of data are as follows: a = Reference 78; b = Strachan and Eisenreich 1988, as cited in Reference 31; c = Rossman 1984 and 1986, as cited in Reference 31; d = Reference 79.

[f] Value for DDT only.

[g] Value for aldrin/dieldrin combined.

[h] Value for polycyclic aromatic hydrocarbons.

[i] Measured water concentrations were beneath detection levels.

Maximum Open Water Concentrations[e]				
Erie	**Huron**	**Michigan**	**Ontario**	**Superior**
0.32[c]	0.061[c]	0.087[c]	0.12[b]	0.044[c]
0.0001[a]	0.00007[a]	no data	0.000074[a]	0.0006[a]
0.000096[a]	**0.000046[a]**	**0.0002[b]**	**0.00014[a]**	not detected
0.0011[a]	**0.00069[a]**	**0.0003[b]**	**0.00051[a]**	**0.00043[a]**
0.00026[a]	0.000073[a]	0.00006[b]	**0.00011[a]**	0.00004[a]
0.0065[a]	0.011[a]	0.01[b]	0.0059[a]	0.011[a]
0.0025[a]	0.0014[a]	0.0007[b]	0.0023[a]	0.0014[a]
3[c]	0.11[c]	0.48[b]	0.4[b]	0.13[c]
0.14[c]	**0.35[c]**	**0.11[b]**	**0.025[b]**	**0.12[b]**
0.0035[a]	**0.0023[a]**	**0.002[b]**	**0.0026[a]**	**0.00058[a]**
0.0003[b]	0.0001[b]	0.001[b]	0.0003[b]	0.0001[b]
nd[i]	nd[i]	nd[i]	nd[i]	nd[i]
nd[i]	nd[i]	nd[i]	nd[i]	nd[i]
nd[i]	nd[i]	nd[i]	nd[i]	nd[i]

Appendix C

Historical EPA Regulations

Authority	Action	GW Pollutants Controlled	Notes
Clean Air Act (1970 - present) Stationary Sources	National Ambient Air Quality Standards for Criteria Pollutants	Lead, Particulate Matter,[a] Nitrogen Oxides	These "health-based" standards established safe concentration levels of six criteria pollutants, three of which are not currently of concern to the Great Waters. States are responsible for implementing regulations to keep the levels of air pollution below these concentrations and are provided guidance by the EPA. States must submit plans to EPA for how areas will meet these standards. Guidance to States includes an identification of alternative control techniques for sources in various industries including incinerators, smelters, electric utilities, cement plants, and wood stoves.
	National Emission Standards for Hazardous Air Pollutants	Mercury	These standards set emission limits for various hazardous air pollutants. Mercury emissions from ore processing facilities, mercury cell chlor-alkali plants, and sludge drying plants were regulated.
Clean Air Act (1970 - present) Mobile Sources	Emissions Controls	Nitrogen Oxides, Particulate Matter, Lead	The Clean Air Act required reductions in emissions from auto exhaust, set more stringent fuel economy standards, and required inspection and maintenance (I/M) programs to locate malfunctioning emission control systems. Since 1970, lead emissions from automobiles have been reduced by approximately 90%. The 1990 Amendments require lower tailpipe standards; more stringent emissions testing procedures; expanded I/M programs; new vehicle technologies; introduction of a range of clean fuels programs; clean transportation provisions; and possible regulation of emissions from nonroad vehicles.
Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA) (1972 - present)		Mercury, Chlordane, DDT/DDE, Hexachlorobenzene, Lindane, Toxaphene	This Act provides the authority for banning and restricting the use of pesticides containing these chemicals in the U.S. according to how and where they are used. It requires registration of all pesticides and reporting of any exported pesticides.
Toxic Substances Control Act (1976 - present)		PCBs	In addition to other requirements, this Act bans the manufacture, processing, distribution in commerce, and use of PCBs except in totally closed systems and establishes rules for disposal of PCBs.
Superfund Amendments and Reauthorization Act (1976 - present)	Emergency Planning and Community Right-to-Know (EPCRA)	All except nitrogen compounds, dieldrin, DDT/DDE, 2,3,7,8-TCDD, 2,3,7,8-TCDF, and some POM.[b]	Establishes new authorities for emergency planning and preparedness, community right-to-know reporting, and toxic chemical release reporting.

[a]Particulate matter includes airborne particles made up of a variety of substances that may include the following pollutants of concern: cadmium, POM, 2,3,7,8-TCDF, and 2,3,7,8-TCDD.

[b]Reporting of releases of these pollutants is not currently required, mainly due to their low emissions. EPA is taking comment on modifications to EPCRA 313 requirements, such as lowering the reporting thresholds to ensure that release and transfer information is obtained for certain persistent pollutants. (See proposed rule: 59 FR 1788, January 12, 1994.)

Note: This table documents EPA legislation that has reduced emissions of Great Waters pollutants directly into the air. It does not account for other legislation that may have reduced these pollutants from other sources that may eventually be emitted to the air. Other such sources may include effluent released to waterbodies and runoff from agriculture.

Appendix D

Summary of Clean Air Act Section 112 Activities[a]

Subsection	Affected Pollutant	Year Due	Comments
112(c)(6)	Lead compounds, Hexachlorobenzene, Mercury, PCB, 2,3,7,8-TCDF, 2,3,7,8-TCDD, POM	2000	Requires regulations for "sources accounting for 90% of the aggregate emissions of each such pollutant."
112(d)	All GW pollutants except nitrogen compounds	1991-2000	Requires regulations for all sources emitting 10 tons of any one hazardous air pollutant[b] or 25 tons total. These sources must have "maximum achievable control technology" (MACT). Smaller sources can be regulated in certain cases.
112(g)	All GW pollutants except nitrogen compounds	1992	Requires MACT for new or modified sources.
112(f)	All GW pollutants except nitrogen compounds	Beginning in 2001	Evaluation of remaining health risk (residual risk) to public after application of the Section 112 standards. Additional standards to reduce residual risk.
112(j)	All GW pollutants except nitrogen compounds	1994	If any Section 112 standard is not promulgated in accordance with the schedule,[c] the individual sources become responsible for controlling their emissions subject to State approval.
112(k)	Not yet determined	1999	90% of emissions of 30 hazardous air pollutants that pose the greatest threat to public health must be regulated.
112(m)	All GW pollutants	1993 and every 2 years thereafter	Reports to Congress due on the status of the Great Waters program including regulatory recommendations.
112(n)(1)(A)	Potentially: Mercury, Cadmium, Lead, POM, TCDF, TCDD	1995	A report to Congress to assess the need for further regulation on this industry.
112(n)(1)(B)	Mercury	1994	A report to Congress will document the health and environmental effects of mercury emissions and the technologies available to control them.

[a]Under section 112(m), the Clean Air Act, as amended in 1990, requires a determination of whether the other provisions of this section are adequate to prevent serious adverse effects to public health and serious or widespread environmental effects. This table is designed to alert the reader to other pertinent activities that affect Great Waters pollutants. Nitrogen compounds are not addressed under Title III of the 1990 Clean Air Act, but they are addressed under Titles I, II, and IV.

[b]Section 112 of the Clean Air Act, as amended in 1990, contains a list of 189 hazardous air pollutants.

[c]National Emission Standards for Hazardous Air Pollutants Schedule for the Promulgation of Emission Standards under section 112(e) of the Clean Air Act Amendments of 1990 (58 FR 63941, December 3, 1993).

Appendix E
Progress Under Section 112(m)

Section 112(m), "the Great Waters program," was written into the Clean Air Act, as amended in 1990, to complement existing programs working to address water quality problems in the Great Lakes and other waterbodies. In fact, the requirements are similar to Annex 15 of the Great Lakes Water Quality Agreement, which the EPA is working to fulfill.

The EPA began planning for the Great Waters program during the summer of 1990. Since then, a great deal of work has taken place through the cooperation of many EPA program offices, laboratories, and regional offices. Other agencies participating actively in the Great Waters program are the National Oceanic and Atmospheric Administration (NOAA) and the appropriate States.

Below is a summary of the activities that have been undertaken by the Agency and the progress on specific monitoring requirements. There is also related work taking place by non-Federal parties, such as emission inventory efforts by State agencies, which will not be addressed in detail here. However, EPA is working closely with the State agencies, and is leveraging their efforts whenever possible and appropriate. These complementary State efforts are necessary to the accomplishment of the goals of the Great Waters program.

Progress by EPA

- Development of a screening level literature review, providing an assessment of what kind and amount of information is available on the issue of atmospheric deposition of hazardous air pollutants (HAPs) to aquatic ecosystems

- Development of an assessment of the 1990 Amendments list of 189 HAPs to determine which are most likely to be problematic when deposited into aquatic systems

- Intra-agency leveraging of relevant activities, including:
  - Lake Michigan Urban Air Toxics Study
  - Great Lakes deposition estimates for Lake Michigan
  - Metals monitoring in Chesapeake Bay area (with NOAA)
  - NO_x deposition modeling in Chesapeake Bay
  - Modification and extension of Chesapeake Bay Atmospheric Deposition Study

  - Sample analysis for Integrated Atmospheric Deposition Study, a U.S./Canadian cooperative network
  - Great Lakes regional toxics emission inventory (with the Great Lakes Commission/States)
  - Compilation of available emission inventory data on a national scale.

- Analysis of existing ambient air metals samples for Gulf of Mexico States
- Conduct of a scoping level mass-balance for nitrogen for Gulf of Mexico
- Preparation of three support documents by technical experts to address the three main scientific questions defined in Section 112(m) of the Clean Air Act: relative loading, effects, and source identification
- Sponsorship of a major workshop for peer review of support documents
- Production of a descriptive brochure of the Great Waters program and the waterbodies included
- Development of a research planning guidance document
- Preparation of a national screening level emission inventory for the specific pollutants in Section 112(c)(6)
- Prototype long-range mercury transport modeling
- Prototype indirect mercury exposure modeling
- Development of a screening level atmospheric loading assessment for Galveston Bay, using a suite of chemicals and a method to complement work in Chesapeake Bay
- Assessment of over-water versus onshore siting for samplers
- Assessment of urban contribution to atmospheric loading
- Deposition sampling for loading assessments
- Development (with Great Lake States) of Lakewide Management Plans (LaMPs), first for Lake Michigan, that have a requirement for assessment of pollutant loading and planning for elimination of water quality effects. Some relevant activities under this program are:
  - Development of a mass balance for PCBs in Green Bay
  - Lake Michigan Urban Air Toxics Study

  - Monitoring for tributary and air loads for Lake Michigan
  - Evaluation of direct loading and sediment contribution to water pollution
- Integration of air and water models for mass-balance calculations
- Working with the Agency on Toxic Substances Disease Registry (ATSDR) evaluating chemical exposure from consumption of Great Lakes fish and health effects on a variety of sensitive or highly exposed population subgroups
- Working with ATSDR to conduct an air toxics monitoring study in conjunction with EPA Region 5 and Southeast Chicago Initiative.

Progress on Specifically Mandated Monitoring Networks

- Five master (regional background) stations collecting wet and dry toxics deposition samples on each of the Great Lakes, begun in 1992 as part of the Integrated Atmospheric Deposition Network—a joint effort between the United States and Canada
- Three stations collecting toxics for the Chesapeake Bay, begun in 1990—a joint effort between EPA and the Bay States
- State-run toxics deposition programs for Lake Champlain, which are to be enhanced through the Lake Champlain Management Conference under the Lake Champlain Special Designation Act of 1990; and mercury deposition monitoring for the Lake

Progress by NOAA on Section 112(m) Issues

- Cooperation with the Lake Champlain Research Consortium and the Vermont Monitoring Cooperative to conduct mercury monitoring and research on wet and dry deposition of nutrients and HAPs and on models for meso-scale deposition in the Lake Champlain basin
- Establishment and operation of the Atmospheric Nutrient Input to Coastal Areas (ANICA) program to develop deposition sampling data for modeling of atmospheric input, especially to the Chesapeake Bay
- Establishment and operation of the Atmospheric Integrated Research Monitoring Network (AIRMoN), an activity using state-of-the-art measurement technologies that should provide quick indication of the impact of Clean Air Act emission reductions on national air quality.

Appendix F

Summary of MACT Source Categories Potentially Emitting Great Waters Pollutants of Concern[a]

MACT Source Category	Cadmium Compounds	Lead Compounds	Mercury Compounds	POM	Regulation Promulgation Schedule
Industrial Boilers	●	●	●	●	11/15/2000
Institutional/Commercial Boilers	●	●	●	●	11/15/2000
Process Heaters	●	●			11/15/2000
Primary Aluminum Production		●		●	11/15/1997
Secondary Aluminum Production	●	●			11/15/1997
Primary Copper Smelting	●		●		11/15/1997
Primary Lead Smelting	●	●	●		11/15/1997
Secondary Lead Smelting		●			4/30/1995[b]
Lead Acid Battery Manufacturing		●			11/15/2000
Coke By-Product Plants				●	11/15/2000
Ferroalloys Production		●			11/15/1997
Integrated Iron and Steel Manufacturing	●	●		●	11/15/2000
Non-Stainless Steel Manufacturing – Electric Arc Furnace (EAF) Operation	●	●	●		11/15/1997
Stainless Steel Manufacturing – Electric Arc Furnace (EAF) Operations		●			11/15/1997
Iron Foundries		●			11/15/2000
Steel Foundries	●	●	●	●	11/15/1997
Steel Pickling – HCL Process				●	11/15/1997
Asphalt Concrete Manufacturing		●			11/15/2000
Asphalt Processing				●	11/15/2000
Lime Manufacturing	●	●	●		11/15/2000
Portland Cement Manufacturing	●	●	●		11/15/1997
Petroleum Refineries – Catalytic Cracking (Fluid and Other) Units, Catalytic Reforming Units, and Sulfur Plant Units	●	●	●		11/15/1997
Petroleum Refineries – Other Sources not Distinctly Listed	●	●	●		6/30/95[b]
Gasoline Distribution (Stage 1)				●	11/23/1994[b]

(continued)

Summary of MACT Source Categories Potentially Emitting Great Waters Pollutants of Concern[a] (continued)

MACT Source Category	Cadmium Compounds	Lead Compounds	Mercury Compounds	POM	Regulation Promulgation Schedule
Auto and Light Duty Truck (Surface Coating)				●	11/15/2000
Printing, Coating, and Dyeing of Fabrics				●	11/15/2000
Hazardous Waste Incinerators	●	●	●		11/15/2000
Sewage Sludge Incinerators	●	●	●		11/15/2000
Synthetic Organic Chemical Manufacturing				●	2/28/1994
Dry Cleaning				●	11/15/1992
Plywood/Particle Board				●	11/15/2000
Pulp and Paper Production	●	●	●		11/15/1997

[a] The information in this table was extracted from the documents EPA-450/3-21-030 *Documentation for Developing the Initial Source Category List* and EPA 455/R-93-048 *National Emissions Inventory of Mercury and Mercury Compounds: Interim Final Report.* These documents represent preliminary data only. Pollutants other than those listed may prove to be present in emissions from these listed sources as more information on source categories becomes available.

[b] Court-ordered deadlines.

Note: If the EPA misses the deadlines in this schedule for promulgating Federal emissions standards by at least 18 months, section 112(j) of the 1990 Amendments requires State and local agencies to establish case-by-case emission standards. Theses case-by-case standards must be equal to the level of control that would have been required by the Federal emission standards.

Appendix G
Preliminary Summary of Research and Program Planning

With the completion of this first report and the ensuing discussions within and outside EPA, it is now appropriate to assess the future needs and direction of the program, the state of the knowledge, and the kinds of efforts needed to provide the necessary information on deposition of air pollutants to the Great Waters. Given that the problem of atmospheric deposition of toxics to aquatic ecosystems is vastly complex and that much of the research in this area is extremely expensive, EPA and other Federal agencies must now determine where efforts are best spent to collect the most important information to meet the mandate of Section 112(m) of the Clean Air Act.

The EPA is working on a program strategy to target the most effective efforts. A preliminary research planning guidance document was prepared that describes many of the efforts necessary to provide each kind of information: relative loading, emission inventory/source characterization, and ecological and human health effects. Summary charts are provided in this appendix to describe those tasks and to begin to rank subtasks. It should be noted that the charts on the following pages are a "first cut" at the research elements and program activities needed to better define and address these issues. There are undoubtedly activities that are needed that have not been listed here, and some of those listed may ultimately prove less important for decisionmaking. However, EPA, in concert with other Federal agencies, is developing an overall strategy to address Great Waters issues in the context of other environmental priorities. The strategy must recognize the realities of time (how long can we wait for an answer before we act or allow impacts by inaction) and money (what funds will continue to be available for research in this area) and define what activities are necessary to provide the important pieces of information.

The strategy, anticipated to be completed by mid-1994, is still being developed, but the essence of it is this: there must be three ongoing efforts that complement and provide information to each other:

Long-term efforts will work toward developing a more certain picture and will provide feedback on the effectiveness of maximum achievable control technology (MACT) standards and other controls.

Short-term efforts will focus on important, transferrable information, usable in an early time frame, especially for regulatory decisionmaking. For example, the Lake Michigan loading/mass balance work will provide an integrated picture of one geographic area as well as provide a transferrable mass-balance methodology.

Justified action is the goal of the program—to determine what, if any, action is needed to prevent adverse effects and to implement or recommend that action. This is not one final action, but a continuum of problem recognition and solution definition over time, as information becomes available through short- and long-term efforts.

This strategy will be developed by the EPA Office of Air Quality Planning and Standards, jointly with the offices and agencies that participate in program decisionmaking through the Great Waters Core Project Management Group. It will also undergo peer review and will be available to interested parties through the Great Waters program.

The following six charts are the summary charts of the preliminary research planning guidance document. This is a compilation of many of the tasks necessary to provide complete information on relative loading, effects, and source identification. **Again, the strategy being developed will define the priority and schedule of the work according to the resulting information's importance, the ease with which it can be obtained, and its utility in determining the need for any additional regulations.**

Ecological Effects Research and Program Needs

Technical Need	Research Issues	Preliminary Priority Ranking	Estimated Relative Costs	Short-Term Benefit	Long-Term Benefit
Mechanisms of Action	Long-term exposure studies with single or low-dose, embryonic or developmental	1	$$$	High	Above average
	Diverse mechanisms for individual effects and diverse effects by individual mechanisms	1	$$	High	High
	Interaction of chemicals	2	$$$	Average	Average
Population Effects	Bioavailability	1	$$$	High	High
	Effects in reptiles, amphibians, and chondrichthian fishes	1	$$$	Above average	Above average
	Thresholds for sensitive populations	3	$$$$	Average	Average
Ecosystem Effects	Eutrophication	1	$$$	High	High
	Ecosystem dynamics, including invader species impacts	1	$$$$	High	High
	Broaden base of studied ecosystems to include warmwater lakes, estuaries, and tropical marshes	2	$$$$	Above average	Above average
	Impact of relatively new contaminants	3	$$$	Above average	High

Source: *Great Waters Technical Planning Guidance*, July 30, 1993.

Detailed Human Health-Related Effects Research Needs

Technical Need	Research Issues	Preliminary Priority Ranking	Estimated Relative Costs	Short-Term Benefit	Long-Term Benefit
Multiple reproductive endpoint studies of men and women	Reproductive couple:				
	Fertility	1	$$$	High	High
	Reproductive behavior	2	$$	Above average	Above average
	Men:				
	Alterations in libido	2	$	Average	Average
	Alterations in spermatogenesis	1	$$	High	High
	Alterations in reproductive tissue	1	$	High	High
	Women:				
	Menarche	2	$	Above average	Above average
	Menstrual cycle	1	$$	High	High
	Menopause	2	$$	Above average	Average
	Time to pregnancy	1	$$	Above average	Above average
	Endometriosis	2	$	Average	Average
	Pregnancy intervals	1	$$	High	High
	Contaminated breast milk	1	$$	High	High
Developmental Effects	Long-term functional significance of effects	1	$$	High	High
	Diminished potential	1	$$$	High	High
	Developmental biomarkers	1	$$	Above average	Above average
	Standardized/nonstandardized testing	2	$$	Above average	Above average
	replication studies	2	$$$	Above average	Above average
Neurobehavioral Effects	Alteration of dopamine production	1	$$	High	High
	Memory and attention deficits	1	$$	High	High
	Standardized vs. nonstandardized testing:				
	Cognitive processing efficiency	1	$$	High	High
	Vigilance/sustained attention	1	$$	High	High
	Activity level	2	$	Above average	Average
Endocrinological Effects	Long-term functional significance of effects	1	$$	High	High
	Delayed effects:				
	Puberty	1	$	High	High
	Senescence	2	$$	Above average	Average
	Menopause	2	$$	Above average	Average
	Sex hormones:				
	Libido	2	$	Average	Average
	Spermatogenesis	1	$$	High	High
	Menstrual cycle	1	$$	High	High
	Thyroid hormone alterations	2	$$	Above average	Average
Immunological Effects	Long-term functional significance of effects	1	$$	High	High
	Immunosuppression	1	$$	High	High
	Immunoenhancement	2	$$	Above average	Above average
	Immunological responses and susceptibility:				
	Primary antibody response	1	$$	High	High
	Diseases of viral and bacterial origin	2	$$	Above average	Above average

Source: *Great Waters Technical Planning Guidance*, July 30, 1993.

Human Health-Related Effects Research and Program Needs

Technical Need	Research Issues	Preliminary Priority Ranking	Estimated Relative Costs	Short-Term Benefit	Long-Term Benefit
Research Compounds	Identify effects across a broader range of compounds	2	$$$	Above average	Above average
Biomarker Development		3	$$	Unknown	Unknown
Noncancer Endpoints	See detailed table for research issues	1			
Interdisciplinary Organization	Information networks	1	$$	High	High
HERL	Vehicle for multidisciplinary research	1	$$$	High	High
NIEHS	Symposia	1	$	High	High
GLIN					
RIEN					
RISP					

Source: *Great Waters Technical Planning Guidance*, July 30, 1993.

Monitoring Research and Program Needs

Technical Need and Programs	Research Issues	Preliminary Priority Ranking	Estimated Relative Costs	Short-Term Benefit	Long-Term Benefit
Atmospheric Deposition Monitoring Networks GLAD (Great Lakes) IADN (Great Lakes) L. Michigan Network Chesapeake Bay Network L. Champlain Station Narragansett, Delaware, Massachusetts, and other bays National as applied to Coastal	Siting Trace organic monitoring Quality control Data compatibility with other stations and networks Coordination among network parties	2 1 1 2 3	$$ $$$ $$$ $$ $	Above average High High High High	Average High High High High
Atmospheric Measurements (same as above)	Spatial and temporal variability Inland vs. shoreline vs. open lake sites Quality control Meteorological	1 2 1 2	$$ $$ $$$ $	High Above average High Average	High Above average High Average
Atmospheric Deposition Processes	Rain, snow, and fog scavenging Gas/particle partitioning Particle deposition velocity	1 2 1	$ $ $$	High Above average High	High Above average High

Source: *Great Waters Technical Planning Guidance*, July 30, 1993.

Atmospheric Modeling Research and Program Needs

Technical Need	Research Issues	Preliminary Priority Ranking	Estimated Relative Costs	Short-Term Benefit	Long-Term Benefit
Source Attribution	Collect ambient and source signature data for source-receptor modeling and apportionment, including source profiles and tracer compounds	1	$$$$	High	High
	Pollutant exchange process: air-soil-plant	1	$$	High	High
Transport Description	Three-dimensional windfields and diffusion coefficients	2	$$	Above average	Above average
	Precipitation field effects	1	$$	High	High
Model Interactions	Nested regional models	1	$$$	High	High
	Nested hemispheric and/or global scale models	2	$$$$	High	High

Source: *Great Waters Technical Planning Guidance*, July 30, 1993.

Emission Inventory and Source Characterization Research and Program Needs

Technical Need	Research Issues	Preliminary Priority Ranking	Estimated Relative Costs	Short-Term Benefit	Long-Term Benefit
Statewide Emission Inventory	Quality control	1	$$$$	High	High
California Air Research Board	Maximal bottom-up data	1	$$$$	High	Average
GLC (8 Great Lakes States)	Chemical focus	2	$$	Above average	Average
	Coordination of inventories	3	$	Average	Above average
Other local areas and States are also involved in developing HAP emission inventories, including Puget Sound, Louisiana, Maryland, Texas, Arizona, and New Mexico	Completeness of source types and chemical speciation	1	$$$	Above average	Average
	Define PCBs, POMs, and TCDDs/TCDFs for inventory purposes	2	$$	Above average	Average
Regional Emissions Inventory GLC	Same as above				
Development of re-emission estimates for Hg and OC pesticides that have been discontinued in the United States	Development of analytical methods to determine re-emission	2	$$	High	Low
	Development of re-emission estimates	2	$$$	High	Above average
	Increased availability of pesticide manufacturing and usage data	1	$	High	High
Definition and Characterization of Urban Air Plume	See emission inventory priorities (above) and modeling section of report				
Lake Michigan and Chesapeake Bay Cooperative Agreement (LMCB)					
Southwest Lake Michigan Urban Area Air Toxic Inventory					

Source: *Great Waters Technical Planning Guidance*, July 30, 1993.

Atmospheric Component of Mass Balance Studies Research and Program Needs

Technical Need	Research Issues	Preliminary Priority Ranking	Estimated Relative Costs	Short-Term Benefit	Long-Term Benefit
Loading Estimates Lake Michigan Mass Budget/ Mass Balance Chesapeake Bay Basin-Wide Toxics Reduction	Increase understanding of atmospheric deposition processes Improve accuracy of loading estimates, particularly for atmospheric deposition	1 1	These relative costs are discussed in the specific research areas within the planning guidance.	———	———
Wet Deposition	Speciation of trace elements, Hg, and N (DON)	2	$$$	Above average	Above average
	Gas/aerosol distribution of SOCs	2	$$	Above average	Above average
	Aerosol scavenging coefficient of SOCs	2	$$	Above average	Above average
Dry Deposition	Aerosol deposition velocity of trace elements and SOCs	1	$$$	High	High
	SOC speciation in water	1	$$$	High	High
	Hg aerosol reactivity	1	$$$	High	High
	DON aerosol concentration	1	$$	High	High
	Nitrogen gas/water partitioning	1	$$$	High	High
	Hg gas exchange (flux)	1	$$	High	High
Microlayer	Improved sampling methodologies	2	$$	High	———
	Development of assay techniques	1	$$$	High	Above average
	Biologic effects research	2	$$$	Above average	Above average
	Process research and integration into air and mass balance models	2	$$$$	———	High
	Microlayer symposium	1	$	High	———

Source: *Great Waters Technical Planning Guidance,* July 30, 1993.

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15. SUPPLEMENTARY NOTES

16. ABSTRACT

This report provides an assessment of the following: (1) the contribution of atmospheric deposition to pollutant loadings to the Great Lakes, Chesapeake Bay, Lake Champlain, and coastal waters (i.e., "the Great Waters"), (2) the environmental and human health effects caused by the deposited pollutants, (3) the sources of these pollutants, and (4) whether atmospheric loadings cause or contribute to exceedances of water quality standards or criteria. The report also includes recommendations for actions to be taken to address this air and water quality problem. Recommendations include EPA committing to do the following: to propose certain emission standards early for some sources of Great Waters pollutants, to consider further regulation of some area sources that emit Great Waters pollutants, to propose a revised Municipal Waste Combustor rule by summer 1994, and to publish an advance notice of proposed rulemaking to establish lesser-quantity emission rates for sources emitting less than 10 tons annually of Great Waters pollutants. Additional recommendations, addressing authorities beyond the Clean Air Act and also focusing on further research efforts, are included in the report.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Air Pollution Atmospheric Deposition Air Toxics Great Waters		

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