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EPA

DEPOSITION OF AIR POLLUTANTS TO THE GREAT WATERS

SECOND REPORT TO CONGRESS

JUL - 2 1997
EPA AIR DOCKET

REVIEW DRAFT

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APPENDICES

LIST OF ABBREVIATIONS AND ACRONYMS

Ah	Aryl hydrocarbon
AQSM	Air quality simulation model
ATSDR	Agency for Toxic Substances and Disease Registry
AWQC	Ambient water quality criterion or criteria
CAA	Clean Air Act
CBADS	Chesapeake Bay Atmospheric Deposition Study
CBP	Chesapeake Bay Program
CCMP	Comprehensive Conservation and Management Plan
CMB	Chemical mass balance
CWA	Clean Water Act
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
dL	Deciliter
EMAP	Environmental Monitoring and Assessment Program
EPA	U.S. Environmental Protection Agency
FDA	Food and Drug Administration
g	Gram
GLWQA	Great Lakes Water Quality Agreement
GLWQB	Great Lakes Water Quality Board
GLWQC	Great Lakes water quality criteria
GLWQO	Great Lakes Water Quality Objective
HAP	Hazardous air pollutant
HCB	Hexachlorobenzene
HCH	Hexachlorocyclohexane
IADN	Integrated Atmospheric Deposition Network
IJC	International Joint Commission
kg	Kilogram
L	Liter
LaMP	Lakewide Management Plan
LCBP	Lake Champlain Basin Program
LQER	Lesser-quantity emission rates
MACT	Maximum achievable control technology
MCL	Maximum contaminant level
m^3	Cubic meters
µg	Microgram
mg	Milligram
NADP	National Atmospheric Deposition Program
NAPAP	National Acid Precipitation Assessment Program
NEP	National Estuary Program
NERRS	National Estuarine Research Reserve System
ng	Nanogram
NOAA	National Oceanic and Atmospheric Administration
NO_x	Oxides of nitrogen
NS&T	National Status and Trends
OTC	Ozone Transport Commission
PAH	Polycyclic aromatic hydrocarbon
PCA	Principal component analysis
PCB	Polychlorinated biphenyl
pGLWQC	Proposed Great Lakes water quality criteria
POM	Polycyclic organic matter

LIST OF ABBREVIATIONS AND ACRONYMS
(continued)

ppb	Parts per billion
ppm	Parts per million
ppt	Parts per trillion
RADM	Regional Acid Deposition Model
RAPIDS	Regional Air Pollutant Inventory Development System
RELMAP	Regional Lagrangian Model of Air Pollution
SAB	Science Advisory Board
SETAC	Society of Environmental Toxicology and Chemistry
SOLEC	State of the Lakes Ecosystem Conference
TCDD	Tetrachlorodibenzo-p-dioxin
TCDF	Tetrachlorodibenzofuran
TSCA	Toxic Substances Control Act
y	Year

CHAPTER I
OVERVIEW OF THE GREAT WATERS PROGRAM

Section 112 of the Clean Air Act (CAA) provides the legislative basis for EPA's hazardous air pollutant (HAP) programs. In response to mounting evidence that air pollution contributes to water pollution, Congress included section 112(m), *Atmospheric Deposition to Great Lakes and Coastal Waters* (also called the Great Waters program), in the 1990 Amendments to the CAA to establish research, reporting, and regulatory requirements related to atmospheric deposition of HAPs to the Great Waters.

This chapter provides a basic introduction and overview of the Great Waters program, emphasizing the role and objectives of the Great Waters Report to Congress. It contains background information related to five main topics: (1) the statutory requirements of section 112(m) of the CAA, including the specific points to be covered by the Report to Congress; (2) the waterbodies comprising the Great Waters; (3) the pollutants of concern in the Great Waters; (4) the findings and recommendations of the First Report to Congress; and (5) the objectives and preparation of the Second Report to Congress.

I.A Statutory Requirements

Activities Under Section 112(m)

Section 112(m) directs EPA, in cooperation with the National Oceanic and Atmospheric Administration (NOAA), to identify and assess the extent of atmospheric deposition of toxic pollutants to the Great Waters. As part of the assessment, EPA supports the following activities:

- Monitoring of atmospheric deposition, including the establishment of monitoring networks in the Great Lakes, Chesapeake Bay, Lake Champlain, and coastal waters;
- Investigation of sources and deposition rates of air pollutants;
- Research for developing and improving monitoring methods and for determining the relative contribution of atmospheric pollutants to total pollutants in the Great Waters;
- Evaluation of adverse human health and environmental effects;[1]
- Identification of exceedances of water quality standards;
- Sampling of fish and wildlife for atmospherically-deposited pollutants and characterizing the sources of such pollutants; and
- Determination of whether regulatory programs under section 112 are "adequate to prevent serious adverse effects to public health and serious or

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[1] For the purposes of this report, environmental effects include both ecological health effects and other effects, such as the commercial impacts of fish advisories or lost recreational opportunities.

> 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 such adverse effects to human health and the environment.

This report responds to the requirements in section 112(m)(5), which directs EPA, in cooperation with NOAA, to periodically submit, 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 atmospheric deposition 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, water quality standards, or Great Lakes water quality objectives; and
- Regulatory changes needed to assure protection of human health and the environment.

Highlights of Progress on Section 112(m) Mandates

Since the First Report to Congress, much progress had been made under section 112(m) through the cooperation of many EPA program offices, laboratories, and regional offices, in addition to NOAA and a number of states. Some of the activities undertaken under section 112(m) include the following:

- Since implementation of the Integrated Atmospheric Deposition Network (IADN) (a joint U.S./Canadian monitoring network), quantitative data have been gathered on atmospheric deposition of pollutants including PCBs, DDT, dieldrin, and lindane in each of the Great Lakes. Recent data have been incorporated into deposition estimates for 1994 and have been compared to 1992 results.
- Pollutants have been sampled in the atmosphere, tributaries, open (lake) water, sediments, and biota for the Lake Michigan Mass Balance Study. Modeling of data will improve understanding of key environmental processes that govern environmental cycling and bioavailability of contaminants in an ecosystem.
- Atmospheric mercury concentration and deposition have been monitored continuously in the Lake Champlain region.
- A large-scale airshed model for Chesapeake Bay has been developed to determine the general geographical location and type of sources of nitrogen emissions, and the patterns of nitrogen deposition within the Bay watershed.

Other Subsections of Section 112 of the Clean Air Act

The requirements under many other CAA section 112 programs are relevant to the atmospheric deposition of air toxics, as highlighted below. Three of these requirements (sections 112(d), 112(g), and 112(j)) focus on the development and implementation of maximum achievable control technology (MACT) standards for the control of emissions from stationary sources of hazardous air pollutants (HAPs). Other sections, such as sections 112(f) and 112(n)(1), evaluate public health concerns related to HAPs, including some pollutants of concern for the Great Waters.

Subsection	Requirement
112(c)(6)	Identify and regulate, under 112(d), the sources responsible for at least 90 percent of total air emissions of alkylated lead compounds, polycyclic organic matter, hexachlorobenzene, mercury, polychlorinated biphenyls, 2,3,7,8-tetrachlorodibenzofurans, and 2,3,7,8-tetrachlorodibenzo-p-dioxin.
112(d)	For source categories listed in 112(c), install maximum achievable control technology (MACT) on "major" sources of HAPs. Includes consideration of a margin of safety where health thresholds are available.
112(f)	Evaluate and, if necessary, take additional actions to address the public health risk remaining after application of section 112(d) standards.
112(g)	Install case-by-case MACT on new and modified sources until 112(d) or 112(j) standards are developed.
112(j)	Control major source emissions if a 112(d) standard does not meet its promulgation schedule.
112(k)	Regulate 90 percent of emissions from urban area sources for at least 30 HAPs that, through research, are shown to pose the greatest threat to public health.
112(n)(1)(A)	Study the potential hazards to public health from emissions of mercury and a few other Great Waters pollutants of concern from electric steam-generating units.
112(n)(1)(B)	Study emissions of mercury to the air from electric utilities, municipal waste combustors, and other sources, considering rate and mass of emissions, health and ecological effects, control technologies, and costs of controls.
112(n)(1)(C)	Determine "threshold" level for human health effects from mercury, including a level for mercury concentrations in fish that may be eaten by humans.
112(r)	Regulate and prevent accidental releases of 77 toxic substances (plus certain flammables and explosives), including furan and some nitrogen compounds (e.g., ammonia).

- Measurements of atmospheric nitrogen have been collected within the Chesapeake Bay watershed to further quantify the linkage with resultant nitrogen runoff from lands surrounding the tidal Bay waters.

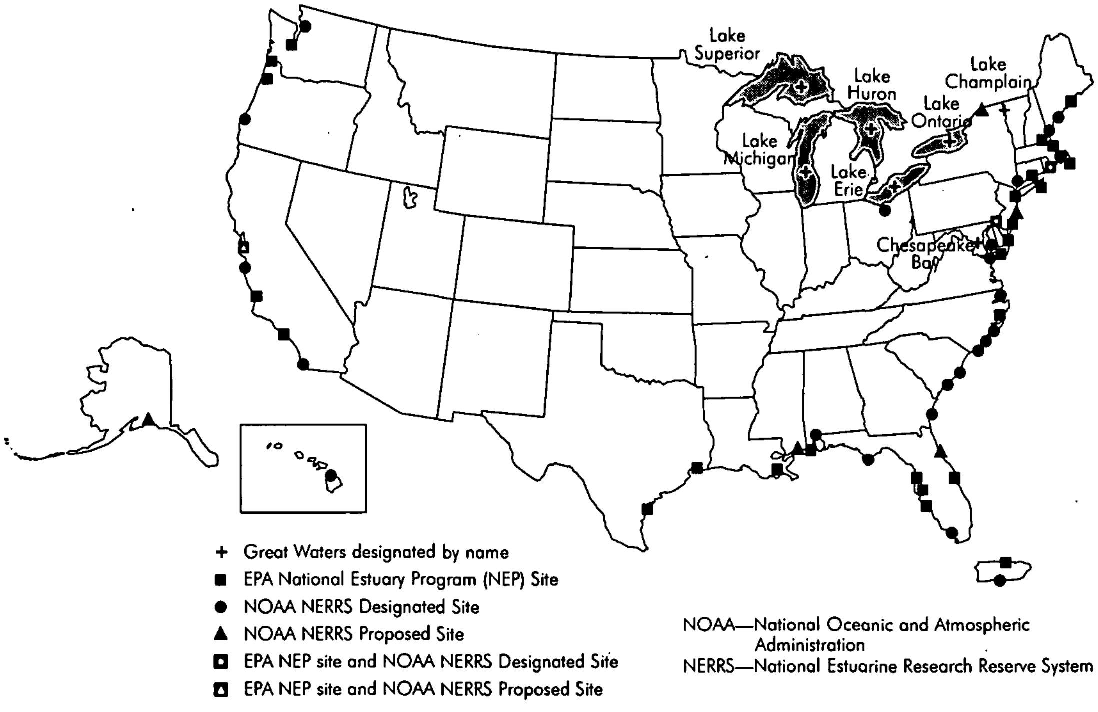
- The Chesapeake Bay Atmospheric Deposition Study (CBADS) network was established to help quantify atmospheric loadings and depositional fluxes of toxic contaminants to the Bay.

- Research stations have been established to measure atmospheric deposition of nitrogen and other selected pollutants in Tampa Bay and Galveston Bay.

I.B The Great Waters

The Great Lakes, Lake Champlain, Chesapeake Bay, and specific coastal waters (i.e., defined in the statute as coastal waters designated through the National Estuary Program and the National Estuarine Research Reserve System) are the waterbodies collectively referred to as the "Great Waters" in this report (see Figure I-1).

FIGURE I-1
Locations of the Great Waters



Much of the available scientific information on deposition of air pollutants is taken from studies on the Great Lakes and Chesapeake Bay. The Great Waters Report to Congress has focused on the Great Lakes because, with their importance as the largest freshwater system in the United States and the observations reported for decades of toxic contamination in organisms living in the Great Lakes, there exists the best base of information on which to build. In addition, studies from the 1980s show atmospheric deposition to be a significant route of introducing pollutants to the Lakes. Knowledge gained of the Great Lakes is useful in evaluating atmospheric deposition in other fresh waters as well. For example, researchers at Lake Champlain have developed scientific programs to determine the role of atmospheric deposition, particularly mercury, in water pollution. This report highlights Chesapeake Bay because accelerated eutrophication and its effects on the Bay have been recognized for over a decade. Accelerated eutrophication in the Bay is attributed, in part, to nitrogen loadings deposited from the atmosphere to the surrounding watershed, as well as directly onto the Bay itself. Similar circumstances affect other U.S. estuaries, and information collected and applied in Chesapeake Bay will be useful for these waterbodies. For example, EPA has sponsored studies to refine the methodology used for estimating sources of nitrogen in Chesapeake Bay and to apply the methodology to estuaries in Galveston Bay, Texas, and Tampa Bay, Florida. A discussion of atmospheric deposition specific to each of the Great Waters -- Great Lakes, Lake Champlain, Chesapeake Bay, and other coastal waters -- is presented in Chapter IV.

I.C Pollutants of Concern

Great Waters Pollutants of Concern and Reasons for Inclusion

As did the First Report to Congress, this report focuses on selected pollutants of concern (see sidebar). These pollutants are potentially of concern for atmospheric deposition in the Great Waters.[2] The general types of sources and uses (and use restrictions) of these pollutants are presented in Table I-1.

The list of 15 Great Waters pollutants of concern has not expanded since the First Report to Congress. Three pesticides, atrazine, hexachlorobutadiene, and methoxychlor, mentioned in the First Report, continue to be flagged by EPA as potential future additions to the Great Waters list of pollutants of concern. Atrazine warrants continued attention as a potential pollutant of concern because of its widespread occurrence (e.g., commonly used in the Great Lakes basin), its at-least moderate persistence, and its potential to cause a variety of effects on biota. For these reasons, atrazine is also one of the chemicals of focus for the Lake Michigan Mass Balance Study (discussed in Chapter IV). The other two pesticides that are flagged by EPA, hexachlorobutadiene and methoxychlor, are both on the CAA HAPs list and have the potential to bioaccumulate in the food web. Additional information suggests that atrazine and methoxychlor are potential endocrine disruptors, a group of chemicals that mimic hormones in the body, resulting in various adverse biological effects.

Great Waters Pollutants of Concern

Cadmium and cadmium compounds
Chlordane
DDT/DDE
Dieldrin
Hexachlorobenzene (HCB)
α-Hexachlorocyclohexane (α-HCH)
Lindane (γ-hexachlorocyclohexane; γ-HCH)
Lead and lead compounds
Mercury and mercury compounds
Polychlorinated biphenyls (PCBs)
Polycyclic organic matter (POM)
Tetrachlorodibenzo-p-dioxin, 2,3,7,8- (TCDD; dioxins)
Tetrachlorodibenzofuran, 2,3,7,8- (TCDF; furans)
Toxaphene
Nitrogen compounds

Under Evaluation for Addition to Great Waters List
Atrazine
Hexachlorobutadiene
Methoxychlor

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[2] This report addresses pollutants from the section 112(b) list of 189 hazardous air pollutants, plus other pollutants of concern. These pollutants are not considered to be inclusive of *all* chemicals that may, now or in the future, be an important component of atmospheric deposition to the Great Waters. Nitrogen's contribution to eutrophication is examined, but acidification or "acid rain" is not discussed because it is addressed in a separate Clean Air Act program.

TABLE I-1
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 cancelled 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. By-product of chlorinated compound and pesticide manufacturing. Also a by-product 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 is used widely in other countries.
Lindane (γ-Hexachlorocyclohexane) (γ-HCH)	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 cancelled 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 a paint additive in U.S. in both interior (1990) and exterior (1991) paint.
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 the United States.
Polycyclic organic matter (POM)[c]	Naturally occurring substances that are by-products of the incomplete combustion of fossil fuels and plant and animal biomass (e.g., forest fires). Also, by-products from steel and coke production and waste incineration.
TCDD	By-product of combustion of organic material containing chlorine and of chlorine bleaching in pulp and paper manufacturing. Also a contaminant in some pesticides.
TCDF	By-product 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 cancelled in 1987.
Nitrogen compounds	By-products of combustion processes and motor vehicles. Also, compounds used in fertilizers.

[a] Data for this table taken from U.S. EPA and ATSDR documents.

[b] Applicable restrictions (including bans) on use or manufacture in 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.

The 15 pollutants of concern for the Great Waters were selected based on available data on their effects and deposition. Reasons for selecting these pollutants include:

- All the pollutants, except for nitrogen compounds, persist in the environment and/or have a high potential to accumulate in living organisms. All the pollutants can cause adverse effects in humans and the environment.
- Data indicate that these pollutants are present in the waters and biota of the Great Waters and that one route of pollutants to these waterbodies is atmospheric deposition.
- All 15 pollutants are known air pollutants and are known to be present in atmospheric deposition (e.g., rainfall).
- These pollutants overlap substantially with the toxic air pollutants that ranked highest in a 1991 EPA study (ICF 1991) to identify chemicals having characteristics that lead to potential problems in the Great Waters.
- With the exception of dieldrin and nitrogen compounds, all of these pollutants are listed as HAPs in the 1990 Amendments to the CAA.[3]
- With the exception of 2,3,7,8-TCDF and nitrogen compounds, these pollutants are included on the list of pollutants that were the initial focus of the EPA/state Great Lakes Water Quality Initiative.[4] They are considered to be potentially significant as air pollutants deposited to the Great Lakes.
- Ten of the 15 pollutants are designated as bioaccumulative chemicals of concern under the Great Lakes Water Quality Guidance.[5]
- Six of the 15 pollutants (cadmium, chlordane, lead, mercury, PCBs, and POM) are on the Chesapeake Bay Toxics of Concern List,[6] and two more pollutants, dieldrin and toxaphene, are listed as potential future additions to this list.
- Nitrogen compounds play an important role in excessive nutrient enrichment in estuaries and coastal waters, and several studies indicate that atmospheric loadings of nitrogen to the Chesapeake Bay and other coastal waters are a significant portion of total nitrogen loadings.

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[3] Hexachlorobutadiene and methoxychlor are also found on the CAA HAPs list.

[4] Established in 1989 to provide consistent level of environmental protection for the Great Lakes ecosystem, and supported principles and goals of the 1986 Great Lakes Toxic Substances Control Agreement (Governors' Agreement).

[5] The Final Water Quality Guidance for the Great Lakes System was released in 1995 (Federal Register 60:15366) and resulted in the deletion of six chemicals (including aldrin, endrin, methoxychlor) from the proposed 1993 list of bioaccumulative chemicals of concern. The final guidance also eliminated the list of 10 pollutants considered potential bioaccumulative chemicals of concern. Although furans (2,3,7,8-TCDF) are not specified in the 1995 guidance, criteria for furans may eventually be set.

[6] Atrazine is also found on the Chesapeake Bay Toxics of Concern List.

◆ The pollutant list overlaps substantially with several sets of Great Lakes chemicals of concern selected by other scientific and regulatory groups, including the Great Lakes Water Quality Board (GLWQB) of the International Joint Commission (IJC), a cooperative committee comprised of U.S. and Canadian representatives.

Pollutant Groups

In some sections of this report, discussion of the Great Waters pollutants of concern is organized by pollutant group. The five pollutant groups used in this report are described below, followed by the rationale for choosing these groupings. Many of the pollutants may fit into more than one group, but have been placed in the most appropriate category (see rationale discussion below).

◆ **Mercury and mercury compounds.** Mercury is released as an air pollutant from a variety of natural and anthropogenic area and point sources (including combustion and manufacturing sources). Although mercury is a metal, it is treated in this report as a separate pollutant group because it behaves differently in the environment than other metals and produces different types of effects, as well as because of the comprehensive data that are available for it. In addition, special emphasis is given to mercury emissions in the CAA. Several subsections of section 112 require studies to be conducted on mercury as a toxic air pollutant; a review draft of an EPA report related to atmospheric emissions of mercury was submitted to the Science Advisory Board (SAB) in 1996 (see sidebar).

SAB Review Draft of the Mercury Study Report to Congress

In June 1996, EPA submitted to the EPA's Science Advisory Board (SAB) a review draft of a comprehensive study report addressing the impact of air emissions of mercury from a variety of sources. This draft report is available to the public, but has not yet been submitted to Congress. When submitted to Congress, this Mercury Study Report will fulfill a mandate under CAA section 112(n)(1)(B).

EPA designed the Mercury Study Report to address many different (but related) types of information:

- Data on type, sources, and trends in emissions;
- Evaluation of the atmospheric transport of mercury to locations distant from emission sources;
- Assessment of potential impacts of mercury emissions close to the source;
- Identification of major pathways of exposure to humans and non-human biota;
- Identification of the types of human health consequences of mercury exposure and the level of exposure likely to result in adverse effects;
- Evaluation of mercury exposure consequences for ecosystems and for non-human species;
- Identification of populations especially at risk from mercury exposure due to innate sensitivity or high exposure; and
- Estimates of control technology efficiencies and costs.

This assessment included judgments as to the potential hazard to humans and wildlife of methylmercury exposure which is largely through the consumption of contaminated fish.

Source: U.S. EPA 1996a.

◆ **Other metals.** Cadmium and lead compounds comprise this group. The metal compounds are released from various combustion and production processes.

- **Combustion emissions.** The pollutants under this group include PCBs, POM, 2,3,7,8-TCDD, and 2,3,7,8-TCDF.[7] These pollutants generally are released during combustion of fossil fuels and/or combustion during manufacturing or incineration processes. PCBs, though historically used in electrical equipment, are included in this group because they may be released to the atmosphere in combustion gases when PCB-containing materials are burned.

- **Pesticides.** The use of these pollutants is significantly restricted, but pesticides continue to be of concern in the Great Waters because of their persistence in the environment. The group includes chlordane, DDT/DDE, dieldrin, α-HCH, hexachlorobenzene, lindane, and toxaphene. Atrazine, hexachlorobutadiene, and methoxychlor are potential future additions to this group.

- **Nitrogen compounds.** This group includes nitrogen oxides and reduced nitrogen compounds (such as ammonia and ammonium); these pollutants are released through both natural and anthropogenic pathways. Nitrogen oxides are combustion by-products, but are treated as a separate pollutant group because: (1) other measures are being taken to control nitrogen through programs related to ground-level ozone and to acid precipitation; (2) nitrogen, unlike the other selected pollutants of concern, is an essential nutrient; and (3) when present in excessive amounts, nitrogen is the nutrient driving the accelerated eutrophication of most estuarine waters, resulting in significant adverse ecosystem effects.

EPA has organized the pollutants of concern in these five groups for several reasons. First, the pollutants in each group generally originate from similar sources or are released through similar mechanisms. Thus, action proposed to reduce emissions of individual pollutants may be applied more broadly to the entire group. Second, pollutants in each group may have similar chemical characteristics, allowing for generalizations related to deposition and cycling within the environment. Third, separating the pollutants into various groups allows for pollutants with unique regulatory concerns, such as mercury and nitrogen, to be highlighted and emphasized in the Report to Congress. Finally, grouping the pollutants helps decision-makers develop conclusions about pollutants with similar chemical/physical behavior or sources, where there are limited data.

Relationship of Pollutants of Concern to Section 112 Requirements

Table I-2 presents the section 112 requirements that could be used to regulate emissions of each pollutant of concern. Pollutants are grouped in Table I-2 by similar requirements and are presented in decreasing order of section 112 coverage. As shown, mercury is the pollutant of concern covered most comprehensively by section 112 requirements, followed by TCDF, lead, POM, TCDD, hexachlorobenzene, and PCBs. (Lead also is regulated as a criteria air pollutant under another CAA program.) For cadmium and several pesticides, the development of MACT standards is the main section 112 requirement that is expected to control emissions of these pollutants. Only section 112(r) applies to

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[7] 2,3,7,8-TCDD (or TCDD) and chemically-related compounds (i.e., similar in molecular structure and in mechanism of action) such as 2,3,7,8-TCDF (or TCDF) are collectively known as dioxins. 2,3,7,8-TCDD is the most widely studied of these compounds.

TABLE I-2
Great Waters Pollutants of Concern and Section 112 Requirements

Pollutant	Applicable Section 112 Activities[a]
Mercury and compounds	• Included on HAP list (112(b)); • Sources accounting for 90 percent of emissions to be subject to regulation per 112(c)(6) under 112(d); • Emissions from major sources subject to regulation by MACT standards under 112(d), 112(g), and 112(j); • Emissions from electric utility steam generating units evaluated for regulation under 112(n)(1)(A); • Emissions from electric utilities, municipal waste combustors, and other sources to be studied (112(n)(1)(B)); and • "Threshold" level for human health effects from mercury to be determined (112(n)(1)(C)).
2,3,7,8-TCDF	• Included on HAP list (112(b)); • Sources accounting for 90 percent of emissions to be subject to regulation per 112(c)(6) under 112(d); • Emissions from major sources subject to regulation by MACT standards under 112(d), 112(g), and 112(j); • Emissions from electric utility steam generating units evaluated for regulation under 112(n)(1)(A); and • Accidental releases evaluated for regulation under 112(r).
Lead and compounds,[b] POM, 2,3,7,8-TCDD	• Included on HAP list (112(b)); • Sources accounting for 90 percent of emissions to be subject to regulation per 112(c)(6) under 112(d); • Emissions from major sources subject to regulation by MACT standards under 112(d), 112(g), and 112(j); and • Emissions from electric utility steam generating units evaluated for regulation under 112(n)(1)(A).
Hexachlorobenzene, PCBs	• Included on HAP list (112(b)); • Sources accounting for 90 percent of emissions to be subject to regulation per 112(c)(6) under 112(d); and • Emissions from major sources subject to regulation by MACT standards under 112(d), 112(g), and 112(j).
Cadmium and compounds	• Included on HAP list (112(b)); • Emissions from major sources subject to regulation by MACT standards under 112(d), 112(g), and 112(j); and • Emissions from electric utility steam generating units evaluated for regulation under 112(n)(1)(A).
Chlordane, DDT/DDE, α-HCH, lindane, toxaphene	• Included on HAP list (112(b)); and • Emissions from major sources subject to regulation by MACT standards under 112(d), 112(g), and 112(j).
Nitrogen compounds[c]	• Not included on HAP list; • Accidental releases of certain compounds (e.g., ammonia) evaluated for regulation under 112(r).
Dieldrin	• Not included on HAP list

[a] Does not include section 112(f), which is intended to address the public health risks remaining after implementation of 112(d) standards.

[b] Also regulated under the criteria air pollutant program.

[c] Regulated under several other CAA programs, such as those that control criteria air pollutants, mobile source emissions, and acid rain.

nitrogen compounds; however, nitrogen compounds are regulated under several CAA programs other than section 112. Dieldrin is the one pollutant of concern that is not currently addressed by section 112 requirements.

I.D Findings and Recommendations from the First Report

Findings and Conclusions

For most of the Great Waters, water quality conditions have improved substantially over the past two or three decades, demonstrating the effectiveness of federal, state, and local legislation and regulations, as well as industry and public cleanup efforts. However, these aquatic ecosystems are far from fully recovered. In order to attain water quality goals and ecosystem protection, the air contribution to water pollution must be addressed. The First Report to Congress, referred to throughout this report as the "First Report," assembled information available at that time to assist EPA in addressing this issue. The most important conclusions of the First Report are related to scientific concepts concerning characteristics of the pollutants, their transport through air, and their impacts after being deposited to waterbodies. Some basic information was quite clear and very important in any discussion of these pollutants. From the data compiled, three main conclusions were drawn:

- Adverse effects possible from exposure to pollutants in the Great Waters (e.g., cancer, developmental effects) are fairly well understood. However, data are insufficient to establish a quantitative link between atmospheric deposition of these pollutants and their related effects.

- Atmospheric deposition can be a significant contributor of toxic chemicals to the Great Waters, although the relative importance of atmospheric loading for a particular chemical in a given waterbody depends on characteristics of the waterbody, properties of the chemical, and the kind and amount of airborne and waterborne sources. Therefore, only some of the data can be generalized to other pollutants and waterbodies.

- Many sources and source categories of pollutants of concern to the Great Waters have been identified. However, atmospheric loadings can originate from local, regional, and global sources and determining the particular sources responsible for the deposited pollutants in a specific waterbody is very difficult.

Specific conclusions from the First Report to Congress, based on scientific data available at that time, included:

- Persistence and the tendency to bioaccumulate, critical characteristics of the Great Waters pollutants of concern, cause potentially greater human and ecological exposure to a pollutant in the environment.

- Exceedances of water quality criteria and standards occurred for some of the pollutants.

- Adverse effects on human health and wildlife have been observed due to exposure to persistent pollutants that bioaccumulate (especially through fish consumption).

- Noncancer effects (e.g., nervous system damage, immunological effects) caused by the pollutants are a significant human health concern, and will impact some individuals exposed to levels above certain thresholds. Many of the pollutants of concern may be toxic to developing embryos and fetuses and breast-fed infants, who are more susceptible than the general population to the adverse effects of these chemicals.

- Ecological effects on animal populations due to the pollutants of concern are significant, such as immune function impairment, reproductive problems, and neurological changes that affect survival. Sometimes the effects on wildlife may be delayed and/or the symptoms subtle so that the effects are easily overlooked.

- Eutrophication, caused by excess nitrogen inputs, is a major problem in U.S. estuarine and coastal waters, and the relative contribution from atmospheric deposition of nitrogen to this problem can be significant. Ecological effects, ranging from nuisance algal blooms to oxygen depletion and fish kills, and economic impacts to the waterbody region may result.

- Case studies have shown atmospheric deposition to be a major contributor of mercury, POM, PCBs, and nitrogen with information on relative loadings estimates. Attention should be given to the absolute quantity of the loadings because even small amounts of pollutants that bioaccumulate can produce a significant burden in fish and, ultimately, in humans and other fish-eating animals.

- Airborne emissions from both local and distant sources contribute to atmospheric deposition of pollutants to waterbodies. Deposition patterns can be influenced by characteristics of the pollutants, source, and weather patterns.

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

Based on the scientific conclusions in the First Report to Congress, EPA's principal policy conclusion was that ***reasonable actions are justified to further reduce atmospheric pollutants, based on evaluation of the scientific data available and given the significant uncertainties in these data, and that these actions should be taken now and that research should continue.***

Progress on Recommendations and Action Items

After evaluating the available scientific information and requesting expertise from scientists, EPA identified action items to carry out the policy conclusion. The recommendations for action were divided into three strategic themes (see sidebar on next page). Some activities that have occurred in recent years to address these themes are highlighted below. Appendix A provides specific information on the status of each of the recommended actions presented in the First Report.

The first strategic theme focused on developing policy to reduce emissions of the Great Waters pollutants of concern by EPA's efforts to continue implementing section 112 and other sections of the CAA, as well as to use the results of the First Report to Congress. Under this theme, the Agency identified 14 specific action items to implement section 112 and other parts of the CAA. Progress has been made on many of the action items:

- A successful partnership with EPA, states, industry, and environmental and tribal interests resulted in the development of the primary aluminum MACT regulation under CAA section 112(d) standards, with promulgation expected in late 1996. The Great Waters program provided funding for the development of this regulation in 1994. The standard focuses on fluoride emissions, but includes limits for POM, a Great Waters pollutant.

- A draft advanced notice of proposed rulemaking for establishment of lesser-quantity emission rates (LQERs) was developed to notify the public of EPA's intent to evaluate the HAPs identified in the Great Waters report, as well as other HAPs listed in section 112(b)(1), to determine whether they warrant the establishment of LQERS. This notice is currently being reviewed within the Agency.

- A pilot study was recently completed on the use of Great Waters impacts analyses in the development of section 112(d) standards.

Three Strategic Themes For Recommended Actions Developed Under the First Report to Congress

1. EPA will continue ongoing efforts to implement section 112 and other sections of the CAA, as amended in 1990, and use the results of the First Report to Congress in the development of policy that will reduce emissions of the pollutants of concern to the Great Waters.

2. EPA recognizes the need for an integrated multimedia approach to the problem of atmospheric deposition of pollutants to waterbodies and, therefore, will utilize authorities beyond the CAA to reduce the human and environmental exposure to 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).

In the second strategic theme, EPA recommended utilizing authorities beyond the CAA to reduce the human and environmental exposure to Great Waters pollutants of concern. The Agency recommended nine action items to assist in the implementation of this theme, and progress has been made on several of these items:

- EPA is continuing its work on toxic air pollutants with Canada, as parties of the Great Lakes Water Quality Agreement, and with the activities of the International Joint Commission. Canada has also provided input on major Great Waters planning and reporting activities.

- EPA continues to work in international forums on the development of protocols for persistent organic pollutants, pesticides, and nitrogen oxides.

Lastly, EPA recommended continued support on research activities and to develop and implement a strategy describing necessary research and policy assessments to address the mandates of section 112(m). In the First Report, the Agency recommended four action items to help implement this theme. Progress on these action items continues:

- Data collection of pollutant levels in atmosphere, water, and biota was completed in 1996 for the Lake Michigan Mass Balance Study. This work also provided valuable data on basic deposition and flux processes, to be incorporated into models as parameters. Calculations and modeling of the data should be completed in 1997.

- The Great Waters program continues to support the development of NOAA-assisted transport and deposition models, which includes determining the modeling parameters of important atmospheric processes. The findings will subsequently be applied to further define and estimate loadings to the Great Waters and to identify sources of atmospherically deposited pollutants.

- Since the First Report, EPA has worked with the Great Lakes States to continue development of regional emission inventories for the Great Lakes and a data storage and retrieval system. Initial data collection was recently completed, and the data base will be updated annually thereafter. Work will continue to characterize mobile source emissions and to add to the emissions inventory.

I.E Highlights of the Second Great Waters Report to Congress

The main objectives of this Report to Congress are to build on the information presented in the First Report by: (1) updating EPA's understanding of atmospheric deposition of toxic air pollutants to the Great Waters based on data available since 1993; (2) reporting on activities in the geographic areas of each of the Great Waters; and (3) presenting further recommendations based on the currently available scientific information.

The information for this report was collected from several sources. To collect recent scientific information about atmospheric deposition to the Great Waters, EPA organized two symposia at the annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC) in Denver, Colorado, from October 31-November 4, 1994. Invited researchers presented findings from current research relevant to the Great Waters program, which was then assembled in a book entitled *Atmospheric Deposition of Contaminants to the Great Lakes and Coastal Waters* and published through SETAC (Baker 1996). To collect additional information in certain critical areas, such as human health and ecological effects, the Agency conducted a literature search to identify recent articles in peer-reviewed literature related to the toxicity and observed effects of the pollutants of concern. In addition, EPA incorporated findings from recent research that has been funded and/or conducted under the Great Waters program, and has integrated findings from other significant EPA projects such as a study of mercury emissions in the United States (U.S. EPA 1996a) and the reassessment of dioxin and dioxin-like compounds (U.S. EPA 1994a, 1994b).

CHAPTER II
EXPOSURE AND EFFECTS

Section 112(m) of the CAA Amendments requires EPA to assess the environmental and human health effects attributable to atmospheric deposition to the Great Waters and to determine whether atmospheric pollutant loadings to the Great Waters cause or contribute to exceedances of drinking water or water quality standards. At this time, it is not possible to distinguish between effects caused by airborne pollutants and the same pollutants delivered by a waterborne route. In the absence of data to the contrary, EPA must assume that the contribution of airborne pollutants to adverse effects corresponds to relative air contribution to pollutant loads. This chapter addresses adverse effects of the Great Waters pollutants of concern through the aquatic food web.

Adverse effects on human and environmental health and exceedances of drinking water or water quality standards that result from the pollutants of concern are pieces of a larger puzzle of what happens to the pollutants of concern after they are deposited to the Great Waters. Once deposited to water, the pollutants can bind to particles, concentrate at the water surface, or dissolve in the water. To determine what happens next requires an assessment of the following:

- *Exposure routes*: how humans and ecosystems are exposed to the pollutants of concern (e.g., food consumption);
- *Extent of contamination*: levels of the pollutants of concern in water and biota to which humans and ecosystems are exposed; and
- *Effects*: ways that humans and ecosystems are negatively affected following exposure to the pollutants of concern.

This chapter addresses these topics in six sections that discuss:

- Exposure and extent of contamination in the Great Waters (Section II.A);
- The level of contamination in Great Waters biota (Section II.B);
- Potential human health effects that may result from exposure to the pollutants of concern (Section II.C);
- Potential ecological effects that may result from exposure to the pollutants of concern (Section II.D);
- Other actual and potential effects, such as recreational fishing losses, attributable to the pollutants of concern (Section II.E); and
- A summary of the information presented on exposure and effects (Section II.F).

Each section (except Section II.B) begins with a brief summary of the information introduced in the First Great Waters Report to Congress (U.S. EPA 1994a) followed by recent information available for the Second Report.

II.A Exposure and Extent of Contamination

This section presents information on the exposure routes of concern for humans and ecosystems and the extent of contamination in the Great Waters. The measures used to assess pollutant exposure and the extent of contamination are exceedances of water quality criteria and the issuance of fishing restrictions and fish consumption advisories. Levels of contamination in biota are discussed separately in Section II.B.

As noted earlier, the relationship between exposure and resulting adverse effects of toxic pollutants and atmospheric deposition is not well understood. As described in the First Report, some correlations and linkages have been established between specific pollutants of concern and exposure and effects in the Great Waters. Many pollutants that are a concern due to atmospheric deposition also have a long history of direct water discharges to the Great Waters. In addition, current pollutant levels may include the contribution of pollutants that are recycled from sediments or that are resuspended, following deposition, and redeposited at other locations. There currently is no evidence available to suggest that the pollutants deposited from the air will have effects on biota any different from these same pollutants carried in water or found in sediment.

Conclusions from the First Report to Congress

The research findings and studies presented in the First Report led to the following conclusions concerning exposure and extent of contamination in the Great Waters:

- For water pollutants that are derived from atmospheric deposition, the major routes of exposure are fairly well understood. The main exposure routes of concern for animals and plants are intake of food, intake of drinking water, and direct contact with water. For humans and fish-eating birds and mammals, intake of food (e.g., contaminated fish) is the main exposure route of concern for pollutants that are persistent in the environment and that tend to bioaccumulate.
- The pollutants of concern generally are persistent in the environment and tend to accumulate in fat or muscle tissue and, as a result of food web interactions, reach the highest concentrations in animals at the top of the food web, including humans. These characteristics allow the pollutants to remain in the environment and animal tissue for long periods of time, increasing the opportunity for exposure and resulting in greater exposures to animals at the top of the food web.

Distinguishing Common Terms Describing Bioaccumulation

Bioaccumulation is the uptake and retention of a chemical by a living organism as a result of intake of food, intake of drinking water, direct contact, or inhalation.

Bioconcentration is the phenomenon by which chemicals become more concentrated in an organism than in its surrounding environment.

Biomagnification is the phenomenon by which chemicals become more concentrated in animals at higher levels in the food web.

- The tendency of the pollutants of concern to bioconcentrate and biomagnify was supported by numerous studies summarized in the First Report. (See sidebar for definition of terms.) Evidence presented by these studies included (1) documented cases of elevated levels of persistent toxic pollutants in various fish species compared to levels in water and, in many cases, levels in sediment; (2) data showing that predators (e.g., the herring gull, bald eagle, and turtle) in the Great Lakes region have had some of the highest reported concentrations of persistent toxic chemicals in their tissues; and (3) data indicating 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 tissues compared with those who did not consume fish.

- Due to levels of pollutants in fish, portions of all of the Great Lakes and many associated waterbodies, Chesapeake Bay, and Lake Champlain had some kind of advisory on fish consumption.

- Understanding of the contribution of atmospheric deposition to overall exposure was limited because: (1) overall exposure to toxic water pollutants had not been adequately quantified; (2) sufficient and accurate information on all pollutant inputs and outputs was not available at that time; and (3) the difficulty in distinguishing the origin of a contaminant (e.g., originated from the air) once it is in the water made it difficult to link exposure, and resultant effects, to particular pathways (e.g., atmospheric deposition).

- Although the exposure routes of concern have been identified, the concentrations of pollutants in water to which humans, animals, and plants are exposed (i.e., the extent of contamination) were not easily determined given available data at that time.

- Few violations of existing drinking water standards (i.e., maximum contaminant levels or MCLs) were found in Great Lakes drinking water systems; for the pollutants that exceeded their MCLs, much of the problem was thought to be caused by the distribution system rather than the water source.

- When maximum open water concentrations from Great Lakes sampling data taken between 1980-1986 were compared to current water quality criteria, six pollutants of concern (cadmium, dieldrin, DDT/DDE, hexachlorobenzene, mercury, and PCBs) potentially exceeded at least one criterion out of three sets of water quality criteria in at least one of the Great Lakes. Maximum concentrations of most of the remaining pollutants in most of the lakes approached levels of concern.

- In Lake Champlain, limited sampling data indicated that lead was the only pollutant of concern that exceeded applicable water quality criteria. In Chesapeake Bay, a limited number of measured concentrations of cadmium and lead in the tidal tributaries to the Chesapeake Bay exceeded EPA water quality criteria and state water quality standards prior to 1993.

Current Understanding of Exposure and Extent of Contamination

As indicated above and in the First Report to Congress, the exposure routes of concern for humans and ecosystems are fairly well understood. Exposures can occur through intake of drinking water, direct contact with water, and, especially important for humans and fish-eating birds and mammals, intake of food. This section presents a few measures for assessing the extent of contamination in the Great Waters.

COMPARISON TO WATER QUALITY CRITERIA

One means of assessing the extent of contamination in the Great Waters caused by the pollutants of concern is to compare available water sampling data to various water quality criteria. Such comparisons are consistent with the requirement in section 112(m) of the 1990 CAA Amendments for EPA to assess the contribution of atmospheric deposition to exceedances of certain water quality standards and criteria. For the purposes of this report, available Great Waters sampling data are compared with three sets of relevant water quality criteria: EPA's national ambient water quality criteria (AWQC); the U.S.-Canadian Great Lakes water quality objectives (GLWQOs); and Great Lakes water quality criteria (GLWQC) developed by EPA and Great Lakes states. The first two sets of criteria are the same as those used in the First Report, while the third set, GLWQC, was released in 1995. Proposed GLWQC (pGLWQC) were used in the First Report, but these criteria have since been finalized (U.S. EPA 1995a); see Chapter V for more discussion on the development of GLWQC. The three sets of criteria are briefly defined in Table II-1.

TABLE II-1
Summary of Water Quality Criteria Used For Comparison in This Report

Criteria Set[a]	Summary
Ambient water quality criteria (AWQC)	Designed to protect humans, freshwater and saltwater animals and plants from harmful effects resulting from chronic and acute exposures. Reflect current knowledge on health and welfare effects, dispersal of pollutants across media, and effects on animal and plant reproduction and communities. Derived entirely with risk-based data (not cost or technology considerations). Provided by EPA as guidelines to states for developing regulations.
Great Lakes water quality objectives (GLWQOs)	Developed through joint U.S.-Canadian agreement. Set for certain chemicals to protect the most sensitive user of the water among humans, aquatic life, and wildlife. For chemicals with no specific GLWQO, concentrations in water (does not specify whether ambient water) and in aquatic organisms should be lower than detection levels.
Great Lakes water quality criteria (GLWQC)	Developed by EPA and Great Lakes States. Specific to the Great Lakes system. Form basis for new state water quality standards for ambient waters of the Great Lakes. Provided as guidelines 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). Includes consideration of bioaccumulation.

[a] References: U.S. EPA 1986, IJC 1978, and U.S. EPA 1995a, respectively.

Water sampling data are compared with water quality criteria, rather than comparing sediment contamination data or biological contamination data to appropriate standards, for two main reasons: (1) the specific requirement in section 112(m) to report exceedances of 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. Because many of the pollutants of concern bioconcentrate and biomagnify, water concentrations may understate the full potential for fish and wildlife to contain high concentration levels; only the GLWQC account for the potential for biomagnification. Therefore, the absence of water quality criteria exceedances for pollutants that have a strong tendency to bind to sediments and to bioaccumulate does not necessarily indicate the absence of contamination levels of potential human health or ecological concern. Contamination levels in biota and sediments in the Great Waters are discussed in Section II.B.

Table II-2 (on next page) compares recent estimates of total water column concentrations in the Great Lakes for the seven pollutants of concern for which sampling data exist. The data are taken from studies conducted between 1986 and 1991 by Environment Canada, EPA, and researchers funded by EPA. As shown in Table II-2, total water column concentrations of dieldrin and PCBs exceed their GLWQC in all of the Great Lakes. In addition, the concentrations of PCBs in Lakes Erie, Huron, and Ontario are above the AWQC for human health. For the pollutants with sampling data reported in both the First Report to Congress and this Report, the total water column concentrations presented in Table II-2 are generally lower than the concentrations reported in the First Report.

Meaningful trend data on contaminant concentrations in the Great Lakes are limited mainly because the technology required to measure contaminants at the trace concentrations found in the water column of the Great Lakes has become widely available only in the last few years. However, recent studies conducted for EPA and NOAA provide insight into water column trends for PCBs (De Vault et al. 1995). As shown in Table II-3, total PCB concentrations in the Lake Superior water column show an overall decline from 1978 to 1992, though there is some variation from year to year.

TABLE II-3
Concentration of Total PCBs in Lake Superior from 1978 to 1992

Year	Total PCB Concentration[a] (ppb)
1978	0.00173
1979	0.00404
1980	0.00113
1983	0.0008
1986	0.00056
1988	0.00033
1990	0.00032
1992	0.00018

[a] Reference: Jeremiason et al. 1994.

TABLE II-2
Comparison of Water Quality Criteria to Pollutant Concentrations in the Great Lakes (ppb)

Pollutant	National AWQC: Fresh Water Aquatic Life[a]	National AWQC: Human Health[b]	Great Lakes Water Quality Agreement Objective[c]	Great Lakes Water Quality Criterion[d]	Total Water Column Concentration[e]				
					Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario
DDT/DDE	0.001	0.00024	0.003	0.000011	<0.00006	NA	<0.00006	<0.00006	<0.00006
Dieldrin	0.0019	0.00071	0.001[f]	0.0000065	0.00026	NA	0.00032-0.00035	0.00038	0.00028-0.00032
HCB	—	0.0072	—	0.00045	<0.00004	NA	0.000072	0.000047	0.000036
alpha-HCH	—	0.092	—	—	0.0011	0.0016	0.0015	0.0011	0.0008-0.0009
Lindane	0.08	0.186	0.01	0.47	0.0004	0.00034	0.00038	0.00049	0.00036
Total PCBs	0.014	0.00079	—	0.0000039	0.00018	0.00020-0.00036	0.0007-0.0009	0.00122	0.0012
POM[g]	—	0.028	—	—	<0.00046	NA	<0.00046	<0.00046	<0.00046

NA=No data available.

Highlighted boxes indicate exceedances of water quality criteria or standards.

[a] Values are for freshwater chronic criteria (EPA 1986).

[b] Values are for human chronic exposure through both fish consumption and drinking water (EPA 1986).

[c] Values are for protection of the most sensitive user of the water among humans, aquatic life and wildlife (IJC 1978).

[d] Values are the most stringent (i.e., lowest) among those for protection of human health, aquatic life, or wildlife (EPA 1995a).

[e] Concentrations are taken from: Jeremiason et al. 1994; U.S. EPA, Great Lakes National Program Office, unpublished data; L'Italien 1993; De Vault 1992; Stevens and Neilson 1989; and Ontario Environmental Quality Branch 1993.

[f] Value for aldrin and dieldrin combined.

[g] AWQC for human health is for polycyclic aromatic hydrocarbons (PAHs), a subset of POM; sampling data are for benzo(a)pyrene, a PAH.

FISHING RESTRICTIONS AND FISH CONSUMPTION ADVISORIES

An additional measure of contamination of the Great Waters caused by selected pollutants of concern is to monitor the existence of fishing restrictions or fish consumption advisories. These advisories are established as a means of limiting human exposure when fish taken from a particular body of water are found to contain levels of contaminants that exceed recommended intake levels (see sidebar). Such advisories have immediate significance to the general public by providing concrete examples of health concerns and impacting the public use of waters and aquatic resources. States issue several different types of advisories for the waterbodies in an effort to reduce health risks associated with exposure to pollutants in certain freshwater fish and shellfish species:

Interpreting Fish Advisory Data

Individual states are responsible for issuing fish advisories. Generally, an advisory is issued for a particular waterbody (or portion of water body), pollutant, fish species, and advisory type. In many advisories, the size of the fish affects the type of advisory issued (e.g., for walleye < 22", restricting meals in the general population may be advised, while for walleye > 22" not consuming the fish may be advised). For several reasons, it is difficult to express advisories quantitatively (e.g., count the number of advisories per water body) and therefore, this Report does not do so. For example, a waterbody may appear to have more fish advisories than another waterbody, but it may be that: (1) more states are involved (e.g., advisories in Lake Ontario are issued only by New York, while four states issue advisories for Lake Michigan; (2) states have different methods or use different standards for identifying fish species affected by advisories, some of which may be more comprehensive than others; or (3) a state may issue an advisory for "all fish" making it difficult to count this advisory with advisories for particular fish species.

- *Informational health advisories*: advisories that fish tissue contains contaminants but not at levels high enough to warrant advising people to limit consumption.
- *Advisories to limit fish consumption*: advisories to either the general population or subpopulations potentially at greater risk (e.g., pregnant or nursing women) to restrict the size and frequency of meals of fish and shellfish;
- *Advisories against fish consumption*: advisories to either the general population or subpopulations potentially at greater risk (e.g., pregnant or nursing women) against consuming fish and shellfish;
- *No-kill zones*: notification that it is illegal to take, kill, or possess any fish from the specified waters; and
- *Commercial fishing bans*: bans on the commercial harvest and sale of fish and shellfish from the specified waterbody.

EPA collects the state advisory data in a national data base (U.S. EPA 1995b). According to the information in this data base for the Great Waters, no active no-kill zones were in effect in any of the Great Waters in 1994 (U.S. EPA 1995b). Commercial fishing bans were in effect for only a few coastal waters, including bans on various species in the New York/New Jersey Harbor related to chlordane, dioxins, and PCBs, and bans on one fish species in Rhode Island waters (which include Narragansett Bay) for PCBs.

However, there are many active advisories to limit or avoid fish consumption in the Great Waters (U.S. EPA 1995b). Table II-4 (the Great Lakes and Lake Champlain) and Table II-5 (selected coastal waters, including Chesapeake Bay) indicate the type of advisories in effect for the relevant pollutants of concern, in increasing order of severity of advisory (i.e., from advisories to at-risk subpopulations to restrict fish consumption up to advisories to the general population to not eat certain fish). For each waterbody presented in these tables, advisories for the entire waterbody (e.g., Lake Huron), portions of the waterbody (e.g., Saginaw Bay), and major tributaries (e.g., Saginaw River) are included. Consequently, the advisories shown may not apply to the entire waterbody. The advisories shown in the tables have been issued for at least one fish species and, in many cases, have been issued for several fish species. For more detail on the fish species affected by the advisories, the states that issued the advisories, and the waterbody portions and tributaries included in the tables, refer to Appendix B.

As shown in these tables, fish advisories in the Great Waters are most commonly associated with PCBs, followed by dioxins. Chlordane and mercury are associated with several advisories, and a few advisories related to DDT, hexachlorobenzene (HCB), polycyclic aromatic hydrocarbons (PAHs), and toxaphene also have been issued. Current fish advisories generally are associated with the same pollutants of concern as in the First Report. Furthermore, in this Report, data are available for many more tributaries and areas of concern for each waterbody than were reported in the First Report. Therefore, the increase in number of advisories presented in Appendix B does not necessarily reflect a higher level of contamination in the Great Waters.

TABLE II-4
Fish Consumption Advisories in the Great Lakes and Lake Champlain

Pollutants of Concern	Great Lakes						Lake Champlain
	Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario	Connecting Channels	
Chlordane	■	◆ • ■	◆ • ■	■			
Dioxins	◆ • ■	▲ ◆ ■	▲ ◆ • ■		◆ • ■	◆ • ■	
HCB				■			
Mercury	▲ ◆ • ■	▲ ◆ •				▲ ◆ ■	◆ •
PAHs				■			
PCBs	◆ • ■	▲ ◆ • ■	▲ ◆ • ■	◆ • ■	◆ • ■	◆ • ■	◆ •
Toxaphene	■						

TABLE II-5
Fish Consumption Advisories in Selected Coastal Waters

Pollutants of Concern	Chesapeake Bay	Other Coastal Waters					
		Narragansett Bay	Long Island Sound	New York/New Jersey Harbor	Delaware Bay	Galveston Bay	San Francisco Bay
Chlordane	◆			◆ • ■	■		
Dioxins				◆ • ■		◆ •	▲ ◆ • ■
DDT							▲ ◆ • ■
Mercury							▲ ◆ • ■
PCBs	◆	◆ •	◆ •	◆ • ■	▲ ◆ ■		▲ ◆ • ■

KEY: ▲ Advisories to subpopulations potentially at greater risk (e.g., pregnant women) to restrict the size and frequency of meals of fish and shellfish
◆ Advisories to the general population to restrict the size and frequency of meals of fish and shellfish
• Advisories to subpopulations potentially at greater risk (e.g., pregnant or nursing women) against consuming fish and shellfish
■ Advisories to the general population against consuming fish and shellfish

Source: U.S. EPA 1995b.

II.B Contamination of Biota

Measurements of pollutant levels in biota provide information about the extent of contamination in the waterbody, as well as potential bioaccumulation in the food web. Section 112(m)(1)(E) of the CAA requires EPA to sample biota, fish, and wildlife in the Great Waters for hazardous air pollutants and to identify the sources of these pollutants. Because studies are already being performed under national programs such as the National Status and Trends Program, Great Lakes National Program Office, Chesapeake Bay Program, Lake Champlain Basin Program, as well as state programs, the Great Waters program relies largely on these studies to fulfill the mandate under the CAA. These programs provide information on the extent of contamination in the waterbodies, although the contribution of atmospheric deposition is generally not evaluated. This section presents a brief overview of biota sampling approaches and how the sampling data provide useful information for assessing the extent of pollutant contamination in a waterbody. This overview includes a summary of two large-scale studies that are designed to assess national pollutant levels in aquatic biota, and how the results from these studies can apply to the Great Waters. This discussion is followed by a summary of research efforts addressing biota contamination specific to the Great Waters.

Sampling Biota for Contamination

Different sampling approaches are used to determine pollutant levels in biota because each monitoring study has its own objectives, such as to identify "hot spots" or characterize a waterbody's general condition. Therefore, caution must be taken in the interpretation of data, as well as statistical analyses applied to these data. To estimate spatial or temporal patterns of contamination, sophisticated sampling designs are often used. Figure II-1 illustrates the importance of sampling various components of an aquatic ecosystem when attempting to characterize the condition of the system.

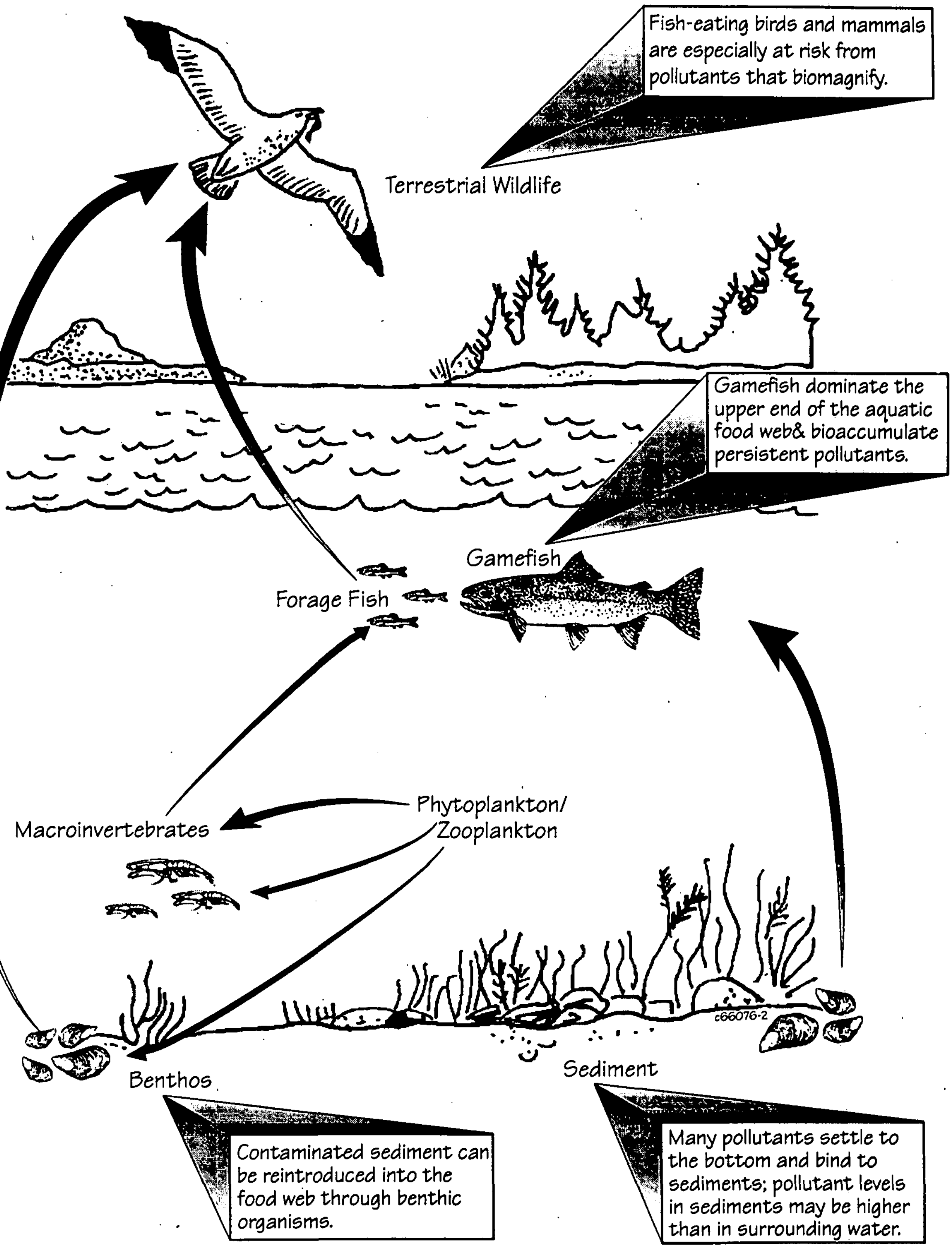
In assessing contamination in a waterbody, aquatic and terrestrial species are often collected and analyzed. Game fish are useful bioindicators because they have long life spans, dominate the upper end of the aquatic food web, and can bioaccumulate many of the persistent pollutants (see sidebar). Their population levels can be affected by continued exposure to environmental stresses, such as eutrophication and pollutant contamination, and people become aware of changes in abundance of game fish. Reduced dissolved oxygen concentrations in the water can affect growth, survival, or structural development of fish, while pollutant contamination

Mercury in the Food Web

Of the pathways by which ecosystems and components of ecosystems may be exposed to atmospheric mercury, exposure of high trophic level (i.e., food chain) wildlife to mercury in food is particularly important. Mercury biomagnifies in aquatic food chains, with the result that mercury concentrations in tissue increase as trophic levels increase. Therefore, the trophic level and feeding habits of an animal influence the degree to which that species is exposed to mercury. Predatory animals primarily associated with aquatic food chains accumulate more mercury than those associated with terrestrial food chains. Thus, fish eaters and their predators generally have the highest exposure to mercury. Species with high tissue levels of mercury include otter and mink, which are top mammalian predators of aquatic food chains. Top avian predators of aquatic-based food chains include raptors such as the osprey and bald eagle.

Source: U.S. EPA 1996a

FIGURE II-1
Assessing Contamination in a Waterbody



Fish-eating birds and mammals are especially at risk from pollutants that biomagnify.

Gamefish dominate the upper end of the aquatic food web& bioaccumulate persistent pollutants.

Contaminated sediment can be reintroduced into the food web through benthic organisms.

Many pollutants settle to the bottom and bind to sediments; pollutant levels in sediments may be higher than in surrounding water.

can result in decreased growth, reproduction, or survival of fish. Contamination in fish can enter the diet of humans and other animals (see sidebar). Therefore, terrestrial wildlife, such as fish-eating birds or mammals, are often monitored. These animals are especially at risk from pollutants that biomagnify because they are frequently exposed to high levels of these pollutants.

FDA Criteria for Chemical Concentration in Fish, Shellfish, or Other Foods

The U.S. Food and Drug Administration (FDA) issues 3 distinct levels of criteria, and each carries different regulatory weight. FDA takes into account both magnitude of health risks to consumers and economic impacts of banning food from any particular source. These criteria are then used by states to develop health advisories.

Advisories: criteria that are considered opinions and have no regulatory weight.

Action Levels: minimum concentrations of a chemical in food that may be cause for FDA to take enforcement action. Although no action levels have been developed by FDA since 1990, levels developed prior to 1990 are still considered applicable.

Tolerance Levels: maximum permissible concentrations; FDA's most rigorously developed numbers and carry the most regulatory weight.

Contaminated sediments pose a potential hazard to human health due to biomagnification up the food web. Many pollutants in the water settle to the bottom and bind to sediments or remain in solution in the water between the sediment particles. Although some pollutants can be degraded by bacteria, many persist in sediments for years, even after the original source of contamination has been removed. Therefore, pollutant levels in the sediment may be higher than the concentrations in the surrounding water and/or biota, and should be interpreted cautiously when extrapolating to potential levels in biota. Pollutants in the contaminated sediment can also be reintroduced into the water column and can enter the food web through benthic (bottom-dwelling) organisms (e.g., clams, crustaceans, and worms), which are prey to larger fish. Benthic community structure, as well as pollutants in tissues of benthic organisms, may also serve as useful indicators of sediment contamination.

Wide-scale monitoring studies have been conducted on pollutant contamination of biota to assess large-scale regional and national impacts in U.S. waters. These programs provide valuable information about major pollutants (e.g., where they are found and where they bioaccumulate) and the extent of contamination in the waterbodies, although the source of contamination (or contribution of atmospheric deposition) is generally not evaluated.

Two large-scale programs that assess the contamination of biota in major U.S. waterbodies or coastal areas are the NOAA National Status and Trends (NS&T) Program and the EPA Environmental Monitoring and Assessment Program (EMAP). The NS&T Program monitors trends of more than 70 chemical contaminants (organic compounds and trace metals) in bottom-feeding fish, shellfish, and sediments at almost 300 coastal and estuarine locations throughout the United States. A well-known project of NS&T is the Mussel Watch. EMAP has carried out regional-scale studies, both terrestrial and aquatic. The data collected for EMAP-Estuaries, to quantify conditions in coastal regions of the United States, are useful to the Great Waters. Sampling and subsequent analyses in EMAP studies are generally not directed at a specific waterbody, but encompass a larger geographic region, which may include waterbodies of the Great Waters. These two projects provide information on assessing the extent of pollutant contamination in U.S. waterbodies; however, care must be taken in applying their findings to individual waterbodies since these projects were not designed to address concerns specific to the Great Waters.

NS&T MUSSEL WATCH PROJECT

Initiated in 1986, this continuing project is directed at tracking temporal trends in concentrations of pollutants (e.g., PAHs, PCBs, pesticides, metals) found in whole soft-parts of mussels and oysters at about 255 coastal and estuarine sites in the Atlantic, Pacific, and Gulf coasts (including Great Waters such as Chesapeake Bay and Galveston Bay).

Biota sampled include two species of mollusks, mussels (*Mytilus edulis*) from the North-East and West Coast, oysters (*Crassostrea virginica*) from the Mid-Atlantic and Gulf coasts, and zebra mussel (*Dreissena polymorpha*) from the Great Lakes. Mussels and oysters are useful for monitoring changes in the pollutant levels because they remain at fixed sites, and concentrations in their tissues reflect in general, changes in the concentrations in the surrounding water. There are species differences, so data need careful interpretation. Sampling for this project is performed during the same season each year to reduce the influence of seasonal cycles on natural factors (e.g., salinity, reproductive state). Sites that were selected support adequate populations of these mollusks such that sufficient samples are available annually over many years (O'Connor 1992; O'Connor and Beliaeff 1995).

Systematic Sampling Approach

The Mussel Watch project is designed to provide long-term and large-scale monitoring of pollutant distribution, looking for temporal (not spatial) trends. The sampling design is set up to be *representative of large areas* rather than small-scale patches of contamination. The objective is to get a representative picture of the general or "average" conditions of the U.S. coastal waters. Therefore, a systematic sampling approach is used. Also, it is useful for estimating statistically the average concentration of pollutants when general trends or patterns in concentration are known from other sources of information.

Table II-6 is a general representation of the pollutant trends in mollusks at Great Waters sites according to the Mussel Watch study; however, magnitudes of the changes were not indicated by O'Connor and Beliaeff (1995). The overall level of chemical contamination in mussels at some Great Water sites has seen a general decrease between 1986 and 1993. Levels of contamination in mollusks from these "average" beds reportedly have not exceeded FDA warning or action criteria for human consumption of shellfish (wet-weight basis).

- Levels of most pollutants of concern (cadmium, PCB, DDT, PAHs, chlordane, and dieldrin) have either decreased or not changed at mussel watch sites in the Great Waters over the eight-year period. Decreases are thought to be the result of bans on the use of chlorinated hydrocarbons and the reduced economic use of certain pollutants.

- Mercury concentrations stayed the same or increased between 1986 and 1990 (data not shown in Table II-6). By 1993, however, the trend over the eight-year period showed that mercury levels had generally stabilized, although an increasing trend was observed at one site at Galveston Bay. Lead levels also have increased recently in some sites at Galveston Bay and Tampa Bay (although no change was evident between 1986 and 1990). The high concentrations of mercury and lead were found, for the most part, in the vicinity of population centers and are, therefore, attributed to human activities.

- Sport-fish and other "fin-fish" species are much higher in the food web than mussels and oysters, and may show different patterns of pollutant concentrations in their tissues.

Although trends shown on Table II-6 are statistically significant, quantitative information is not presented, and consequently, a comparison of the increases or decreases cannot be assessed between sites (e.g., cannot determine which sites showed the most improvement).

TABLE II-6
Eight-Year Trends of Pollutant Concentrations in Mussel Watch Program (1986-1993)

Waterbodies (number of sites examined)	Contaminant Trend (number of sites affected)[a]							
	Mercury	Lead	Cadmium	PCB	DDT	PAH	Chlordane	Dieldrin
Chesapeake Bay (5 sites)			↓ (1)	↓ (3)	↓ (3)		↓ (4)	↓ (3)
Delaware Bay (4 sites)			↓ (1)	↓ (2)			↓ (1)	
Long Island Sound (9 sites)		↓ (1)	↓ (4)	↓ (5)	↓ (2)	↓ (1)	↓ (3)	↓ (1)
Narragansett Bay (2 sites)		↓ (1)						
Tampa Bay (3 sites)		↑ (1)		↓ (1)	↓ (1)		↓ (2)	
Galveston Bay (6 sites)	↑ (1)	↑ (1)		↓ (2)	↓ (1)		↓ (4)	↓ (3)

[a] Represents trends in annually measured concentrations of contaminants in mollusks; blank cell indicates no change in contaminant levels over 8-year period; trend indicated by arrow (i.e., ↓ = decreasing trend; ↑ = increasing trend). Number of sites showing trend within each water body is indicated in parentheses. Sites were sampled in at least 6 of the 8 years. All trends shown are statistically significant.

Adapted from O'Connor and Beliaeff (1995).

EMAP-ESTUARIES

One goal of EMAP is to quantitatively evaluate the condition of coastal estuaries, by investigating several environmental conditions: hypoxia (low oxygen levels), sediment contamination, coastal eutrophication, and habitat loss. A probability-based approach is used, which allows estimates to be made of the uncertainty associated with assessments and improves the ability to identify ecological responses to pollution. Of interest to Great Waters are the EMAP-Estuaries results obtained for the Virginian Province (Cape Cod to the Chesapeake Bay) and Louisianian Province (Texas to west coast of Florida). Statistical data collected provide primarily quantitative information on a regional scale. Comparing among specific waterbodies within the regional area are limited because the density of sampling points was not designed to thoroughly characterize each waterbody separately.

EMAP is a national program initiated in 1989 in response to the EPA Science Advisory Board's recommendation to monitor the status and trends of the U.S. ecological resources -- terrestrial, freshwater, and marine. The program is directed by EPA's Office of Research and Development, with participation by other federal agencies (e.g., NOAA, U.S. Forest Service, U.S. Fish and Wildlife Service).

Results are available for the Virginian Province for 1992 (trend data for 1990-1993 are currently being analyzed), which include sampling results for two relevant Great Waters sites, the Chesapeake Bay (as well as connecting tributaries and small water systems) (53 sampling stations) and Long Island Sound (14 stations). Together, the two waterbodies represent approximately 63 percent of the surface area of the entire province. The 1992 data demonstrate that Chesapeake Bay exhibits a eutrophication effect (i.e., high percentage of bottom-waters with dissolved oxygen concentrations ≤2 mg/L). Metal concentrations in the sediment for the Chesapeake Bay were similar to the concentrations for the overall Province (≈24 µg/g for lead; ≈0.054 µg/g for mercury; ≈0.206 µg/g for cadmium). In Long Island Sound, there was no evidence of eutrophication. The concentrations of some metals in the sediment (44.2 µg/g lead; 0.088 µg/g for mercury) were higher than those reported for the Chesapeake Bay and the overall Virginian Province. Considerable uncertainty exists in this level of analysis of these two waterbodies due to the small number of samples for comparison and short (one year) data collection period (Strobel et al. 1994).

Discussion of Biota Contamination by Major Waterbody

GREAT LAKES

Contaminant concentrations in gamefish from the open waters of the Great Lakes have been monitored for over 20 years and provide one of the most extensive databases on trends of environmental contaminants in organisms at the upper end of the food web. These data come from three monitoring efforts: lake trout monitored by Fisheries and Oceans Canada; lake trout and walleye cooperatively monitored in U.S. waters by EPA's Great Lakes National Program Office, U.S. National Biological Service, and the Great Lakes States; and coho salmon fillets cooperatively monitored in U.S. waters by the Great Lakes States, FDA, and EPA's Great Lakes National Program Office.

These monitoring efforts have demonstrated that, while significant declines in PCB and DDT concentrations in lake trout, walleye, and coho salmon have been observed over the past two decades, the amount of residues of PCBs and DDT in these fish have leveled off or even increased in the last few years (De Vault et al. 1995) (see sidebar). This change in trend has occurred despite declining ambient water concentrations of PCBs. A similar trend has been noted in the levels of PCBs and DDT in herring gull eggs, which have been monitored by the Canadian Wildlife Service since 1974.

PCB Contamination in Great Lakes Biota

Lake Trout/Walleye: During the period 1977-1990, PCB concentrations in lake trout, as well as walleye in Lake Erie, declined significantly, but in recent years, concentrations have generally not changed in Lakes Michigan, Huron, Superior, and Erie.

Coho Salmon: PCB concentrations collected from Lake Michigan declined from 1.9 µg/g (1980) to 0.38 µg/g (1983), but then increased to 1.09 µg/g (1992). A similar pattern was observed in Lake Erie and the upper reaches of the Saint Lawrence River.

Herring Gull Eggs: Monitored since 1974, the greatest decline in PCB contamination occurred between 1974 and 1981. Since then, the rate of decrease has leveled off, and by 1991, slight increases were reported in the levels of some PCBs.

Source: De Vault et al. 1995.

The strong correlation between trends in DDT and PCBs suggests that changes in the composition of the food web (or trophic structure) may be partly responsible for increases in contaminant concentrations at the upper end of the food web (i.e., gamefish) (De Vault et al. 1995). Recent research on the exposure dynamics of organic pollutants in Lakes Erie and Michigan supports the theory that changes in the food web could be the cause of the observed increase in PCB contamination in biota (Haffner 1994; Stow et al. 1995). Large, regional pools of PCBs can be reintroduced from sediments by benthic organisms. Changes in the species composition at the mid-trophic levels of the food web biomagnify greater amounts of PCBs to higher trophic levels. Increased PCB levels in certain predator fish also may be due to reductions in their growth rate.

Evidence of changes in the exposure dynamics of organic contaminants has been observed in the western basin of Lake Erie with the invasion of zebra and quagga mussels (Haffner 1994). These mussels increase biomagnification of pollutants in the benthic food web by consuming significant amounts of phytoplankton that are contaminated with pollutants. The major predator of the zebra mussel is the drum (a low trophic level fish), which in turn is a preferred prey of the herring gull. By mobilizing contaminants in sediments and phytoplankton, zebra and quagga mussels can cause PCB levels in certain fish and in herring gulls to increase even though ambient water concentrations of contaminants are decreasing.

The three monitoring programs discussed above also provide information on the levels of dieldrin and toxaphene in upper trophic-level fish from the Great Lakes. Dieldrin concentrations have exhibited a general pattern of decline in the Great Lakes since the 1970s (De Vault et al. 1995). Peaks were noted in 1979 in Lakes Michigan, Superior, Huron, and Ontario and again in 1984 in Lakes Superior, Huron, Ontario, and Erie. Toxaphene concentrations are highest in lake trout from Lakes Michigan and Superior and lowest in those from Lakes Erie and Ontario (De Vault et al. 1995). Toxaphene is currently the dominant contaminant in Lake Superior lake trout, and it is second to PCBs in Lake Michigan lake trout. Toxaphene levels in lake trout have not been monitored long enough to detect trends.

In contrast to the monitoring studies of gamefish, monitoring forage fish provides an indication of contamination at lower levels of the food web. Rainbow smelt have been routinely monitored in Lakes Superior,,Huron, Erie, and Ontario by Fisheries and Oceans Canada since 1977. During this time, concentrations of PCBs, mercury, and total DDT have declined significantly in smelt from these lakes (De Vault et al. 1995).

Sediment Data in the Great Lakes

Atmospheric pollutant loadings into the Great Lakes region are estimated from mass balance studies and modeling data, although indirect measures of contaminant loadings, such as sediment core data, are also desirable. Recent measurements of sediment core data have shown declining concentrations of PCB and DDT in Lakes Michigan and Ontario and both lead and mercury in Lakes Superior, Michigan, and Ontario. Sediment concentrations plus atmospheric deposition data from the International Atmospheric Deposition Network (IADN) have been used to estimate the atmospheric contribution of metal loadings. Comparison of sediment data between the Great Lakes provides additional information on sources of loadings. For example, toxaphene has long been thought to result from long-range atmospheric transport from southeastern U.S.; however, sediment cores from northern Lakes Michigan and Superior show no significant decline in toxaphene (contrary to trends observed for PCB, DDT, mercury, and PCBs in these lakes). Efforts are underway to examine this issue.

Source: De Vault et al. 1995.

Smelt from Lake Ontario consistently have the highest tissue concentrations of PCBs and total DDT, while those from Lake Superior have the highest mercury levels.

Contaminant concentrations in young-of-the-year spottail shiners are useful indicators of local, recent pollutant inputs into aquatic ecosystems because they do not travel extensively during their first year of life. Surveillance of these fish by the Ontario Ministry of Environment and Energy (primarily in Canadian waters) has shown a general decline in tissue PCB and DDT concentrations (De Vault et al. 1995). Contaminant levels also have been assessed in young-of-the-year fish from the New York waters of the Great Lakes (Skinner et al. 1994). Elevated concentrations of PCBs were found mainly in fish from the St. Lawrence River drainage area below the Moses-Saunders Dam in Masena; these levels were attributed to industrial activities in the area. Levels of mercury in these fish were low (<100 ng/g).

LAKE CHAMPLAIN

Current efforts to monitor toxic pollution in Lake Champlain have focused on fish and sediment contamination by metals and organic compounds. The Vermont Department of Environmental Conservation implemented a study to analyze soft tissue from mussels (*Elliptio complanata*) as a bioindicator of the Lake. Mussels were collected at mouths of several Lake Champlain tributaries; chlordane and PAHs were detected in the mussels (LCBP 1994). In 1987-1988, the States of Vermont and New York analyzed fish tissue collected from Lake Champlain for 17 contaminants. Elevated levels of PCBs were found in large lake trout and in American eel and brown bullhead. The findings of this study led, in part, to health advisories being issued against eating these fish species in Lake Champlain (LCBP 1994).

Due to elevated levels of pollutants in Lake Champlain, the Lake Champlain Sediment Toxics Assessment Program was initiated (McIntosh 1994). Pollutants that were measured included trace metals (cadmium, mercury, lead) and organic compounds (PCBs, PAHs, dioxins/furans). Pollutant levels were measured at nine sites during 1991 and 1992. Findings after the end of the first phase (May 1993) provided little evidence of widespread high-level contamination (although high levels of PCBs and PAHs were measured in sediment near two dock sites). The study did find widely varying patterns of contamination. It appears that, for some pollutants, extremely high concentrations were present only in deeper layers of the sediments, with the upper layers of sediment showing less contamination. Other pollutants showed a reversal of this pattern (McIntosh 1994). The local and/or regional source of the contamination is not known.

As part of the Lake Champlain Sediment Toxics Assessment Program, a biological assessment of the contaminated sediments was also performed (McIntosh 1994). Most of the year, lake trout do not inhabit bottom waters near the sediment-water interface. However, concerns exist for the mechanisms that may link lake trout to PCB-contaminated sediment. This issue was evaluated by looking at one possible link, the freshwater shrimp *Mysis relicta* (or mysids). Mysids are believed to be a major component in the Lake Champlain food web, and the high lipid content of these organisms make them potential accumulators of PCBs. Laboratory experiments demonstrated that exposure to PCB-contaminated sediment results in high levels of PCBs in the mysids. However, there was no attempt to predict the potential of mysids to redistribute PCBs within the sediments in Lake Champlain.

CHESAPEAKE BAY

Adverse effects, such as reduced growth, reproduction effects, and tumor development, have been reported in aquatic organisms in a variety of habitats in the Bay from the 1980s to the early 1990s, which could be related to pollutants accumulating in the tissues of organisms. The Chesapeake Bay Program has sponsored forums to assess contaminant levels in biota and to reach a consensus regarding the trends in the pollutants found in biota and in sediment. While significant declines in metal contamination of fish tissue have been observed over the past two decades, elevated metal concentrations have been measured in specific, more industrialized areas of Chesapeake Bay (CBP 1994b).

Studies collected from 1970 to 1992 show that chemical contamination has caused various effects to wildlife in the Chesapeake Bay (CBP 1994b). The most recent effort to assess contamination in the Chesapeake Bay was the *Status and Assessment of Chesapeake Bay Wildlife Contamination Forum* held in 1991 to critically review data and information on the effects of exposure and uptake of pollutants on Chesapeake Bay basin birds, mammals, reptiles, and amphibians. The committee concluded that there was little evidence to suggest pollutants were posing a serious *direct* hazard to birds in the early 1990s. Instead, it is more likely that indirect effects on wildlife habitats and food sources (e.g., excessive nutrients, suspended sediments, herbicides) have greater impacts on bird populations. Similarly, the forum found that the data available at the time did not indicate any adverse impacts of chemical contamination on mammal populations; however, the data were not specific to the Chesapeake Bay area. For reptile and amphibian populations in the Chesapeake Bay basin, the available studies were insufficient for the committee to draw conclusions regarding threats to these organisms (CBP 1994b).

In 1993, the *Chesapeake Bay Finfish and Shellfish Tissue Contamination Critical Issues Forum* was held to address the following issues: (1) magnitude and extent of fish and shellfish contamination in the Chesapeake Bay and its basin; (2) impact (i.e., bioaccumulation, toxicity) of the contamination at basinwide, baywide, regional, or local scales; and (3) comparison of the contamination to that of other waterbodies (e.g., Puget Sound, Great Lakes) (CBP 1994b). The data compiled by the forum indicate that finfish and shellfish tissue contaminant concentrations declined significantly after the 1970s for several metals, pesticides, and organic chemicals. For fish species combined, concentrations of PCBs and DDT in fish liver tissue are in the low range relative to national data. However, at the species-specific level, atlantic croaker and spot collected from 1984 to 1987 had levels of chlordane, PCBs, dieldrin, and total DDT in the liver above the national average and national median for these species. Lead and mercury concentrations in croaker livers were generally above the national average and national median, while lead concentrations in spot livers were sporadically high. The concentrations of PCBs, chlordane, dieldrin, DDT, cadmium, and mercury in oysters in the Chesapeake Bay have declined between 1986 and 1991. Levels of mercury, chlordane, toxaphene, and PCBs in finfish from the Chesapeake Bay Basin "hot spots" (e.g., Baltimore Harbor, Back River) are generally well below those found at other areas considered contaminated (e.g., New York/New Jersey Harbor, Lake Michigan, Boston Harbor).

OTHER COASTAL AREAS

Monitoring studies of biota contamination also have been performed in other coastal waterbodies of the Great Waters Program. Some recent findings are highlighted below:

- *Galveston Bay.* There is little information about historical trends and concentrations of pollutants in aquatic organisms from Galveston Bay. For this reason, the Galveston Bay National Estuary Program initiated a study to characterize pollutant contamination in edible fish and shellfish in the bay. Between 1991 and 1993, 14 fish species, two shellfish species, and three bird species were sampled for numerous pollutants, including several Great Waters pollutants of concern. No "hot spots" of biota contamination were detected and the fish tissue concentrations rarely exceeded FDA criteria for these contaminants (Brooks et al. 1992). The study did not evaluate dioxins, but currently, fish consumption advisories to protect the general population exist for dioxins in Galveston Bay (see Table II-5).

- *Tampa Bay.* Total PCB concentrations in Tampa Bay oysters (C. *Virginica*) sampled between 1986 and 1989 were high enough (average for four years was 0.15 ppm) to create some potential for adverse biological effects in these organisms (e.g., reproductive effects, cellular damage, and biochemical changes). Mercury concentrations in Tampa Bay oysters indicated a moderate potential for adverse effects; the overall average concentration of mercury in the collected oysters over four years was 0.27 ppm dry weight (Long et al. 1991).

- *New York Bight.* PCB levels in blue crabs collected from the New York Bight in 1989 averaged 5.5 ppm, which exceeds the FDA PCB tolerance level of 2 ppm. Fish collected from the Bight contained tissue levels of chlordane, DDT, DDE, dieldrin, hexachlorobenzene, and lindane that exceeded recommended fish tissue criteria for these contaminants (U.S. EPA Region II et al. 1990).

- *New York/New Jersey Harbor.* Aquatic biota monitoring data from the New York/New Jersey Harbor Estuary show a significant decline in the concentrations of 2,3,7,8-TCDD and total PCBs in striped bass since 1983. However, 1987 and 1993 data show that striped bass collected from some areas of the Estuary still exhibit PCB levels that exceed the FDA tolerance level of 2 ppm (New York-New Jersey Harbor Estuary Program 1995).

II.C Human Health Effects

Much of the information presented in this section on human health effects is based on effects observed in animals, mainly in laboratory studies, that are suggestive of potential human health effects. Unlike the documented effects of certain pollutants of concern on ecological health in the Great Waters (see Section II.D), data on adverse health effects observed in humans from non-occupational exposure to the pollutants of concern are limited.

Conclusions from the First Report to Congress

The research findings and studies presented in the First Report led to the following conclusions regarding human health effects and the pollutants of concern:

Potential Human Health Effects Caused by Pollutants of Concern

- Cancer
- Reproductive effects
- Developmental effects, including effects on embryos, fetuses, and children
- Neurological (i.e., brain and nervous system) effects
- Immune system effects
- Endocrine system effects, including effects on hormone synthesis and function
- Other noncancer effects, including liver and kidney damage

- ◆ Numerous studies indicated potential human health effects associated with the pollutants of concern (see sidebar).
- ◆ Though many of the pollutants of concern are probable carcinogens, the noncancer effects of these pollutants are also a significant concern and may be as detrimental as cancer, or more so, to individuals and populations.
- ◆ It is possible for low-level exposure to several of the pollutants to have little or no measurable effect on an adult, yet alter the formation and function of critical physiological systems and organs in children of the exposed adult, especially when the child is exposed at critical developmental stages *in utero.*
- ◆ Some human health effects caused by the pollutants of concern are subtle, result from long-term exposures to low levels of pollutants, and may be delayed in onset and occur across multiple generations. For example, long-term exposure to low levels of mercury may result in kidney or nervous system damage only after gradual exposure and bioaccumulation in the body.

Current Understanding of Human Health Effects

Since the First Report, new information on the pollutants of concern and the health effects to humans and animals has become available. This section summarizes the health effects data in the current literature. First, a discussion of some health effects research relevant to some Great Waters pollutants of concern is presented, including the scientific efforts on assessing effects of endocrine disruptors and the release of multi-volume reports by EPA on mercury and dioxins. Second, a brief overview of recent data on the Great Waters pollutants of concern is presented, followed by a more detailed summary of the effects data on each pollutant of concern.

As introduced in the First Report to Congress, the role of endocrine disruptors in causing adverse human health effects is an emerging and controversial issue. Endocrine disruptors were termed "environmental estrogens" in the First Report; however, because the interference with hormone action was found not to be limited to estrogen, these pollutants are now more generally referred to as "endocrine disruptors." For example, DDE has been shown to inhibit the binding of androgen, a male hormone, to receptors, among other androgen actions (Kelce et al. 1995). Research to date has identified 11 of the 15 pollutants of concern to the Great Waters as possible endocrine disruptors: chlordane, dieldrin, DDT/DDE, hexachlorobenzene, lead, lindane, mercury, PCBs, TCDDs, TCDFs, and toxaphene (e.g., Cassidy et al. 1994; Chowdhury et al. 1993; Colborn et al. 1993; McKinney 1994; Soto et al. 1994; U.S. EPA 1994c). Two of the potential pollutants of concern, atrazine and methoxychlor, also have been identified as possible hormone disruptors (ATSDR 1993b; Colborn et al. 1993; Fail et al. 1994; Kniewald et al. 1994; Payne et al. 1992). Since the First Report to Congress, scientific research of endocrine disruptors has continued to provide evidence of their adverse effects and has attempted to determine their mechanisms of action (i.e., how the pollutant causes the effect within the body).

The existence and effects of the hormone-like action of environmental pollutants were first hypothesized in the late 1980s by scientists concerned with noncancer effects of toxic pollutants. In July 1991, the Wingspread Conference brought together many scientists whose diverse research interests touched on some aspect of endocrine system disruption. The conference helped identify future research needs for improving the understanding of endocrine disruptors, their mechanism of action, and their effects (NWF 1994). Many of the adverse effects in humans and in ecosystems (e.g., reproductive, developmental, and immunological effects) associated with the pollutants of concern are now thought to be associated with the endocrine-disrupting action of the pollutants. Recent articles published in the mass media have brought this issue widespread attention (Begley and Glick 1994; Suplee 1996; Weiss 1994; Weiss and Lee 1996).

Effects of endocrine disruptors have been seen mostly in wildlife and laboratory experiments; however, there are a few known cases of human exposure to endocrine disruptors (see sidebar for example). Humans are especially susceptible to exposure to endocrine disruptors during periods of high hormonal activity, such as during embryo and fetal development (Hileman 1994). As an example, the chemical hormone diethylstilbestrol (DES) was given to millions of women to prevent miscarriages between the 1940s and 1970s. Though the women were largely unaffected by their exposure to DES, their children exhibited reproductive deficiencies and physical abnormalities, such as decreased fertility in both sexes, testicular cancer in males, and abnormal pregnancies in females (Colborn et al. 1993; Hileman 1994). Scientists use the DES incident as a model of how endocrine disruptors may affect humans.

Human Exposure to Endocrine Disruptors

Accidental exposure to cooking oil contaminated with PCBs occurred in Japan in 1968 and Taiwan in 1979, affecting 3,000 to 5,000 people overall. Children born to women who consumed the oil when pregnant exhibited birth defects and developmental and behavioral deficiencies that scientists believe may be related to hormonal changes caused by exposure (Lai et al. 1993; Rogan and Gladen 1992). Upon reexamination of the Taiwanese children in the early 1990s, researchers found that the children scored lower than controls on IQ tests, girls were shorter than average, and boys' penises were significantly smaller than normal (Chen and Hsu 1994; Guo et al. 1993).

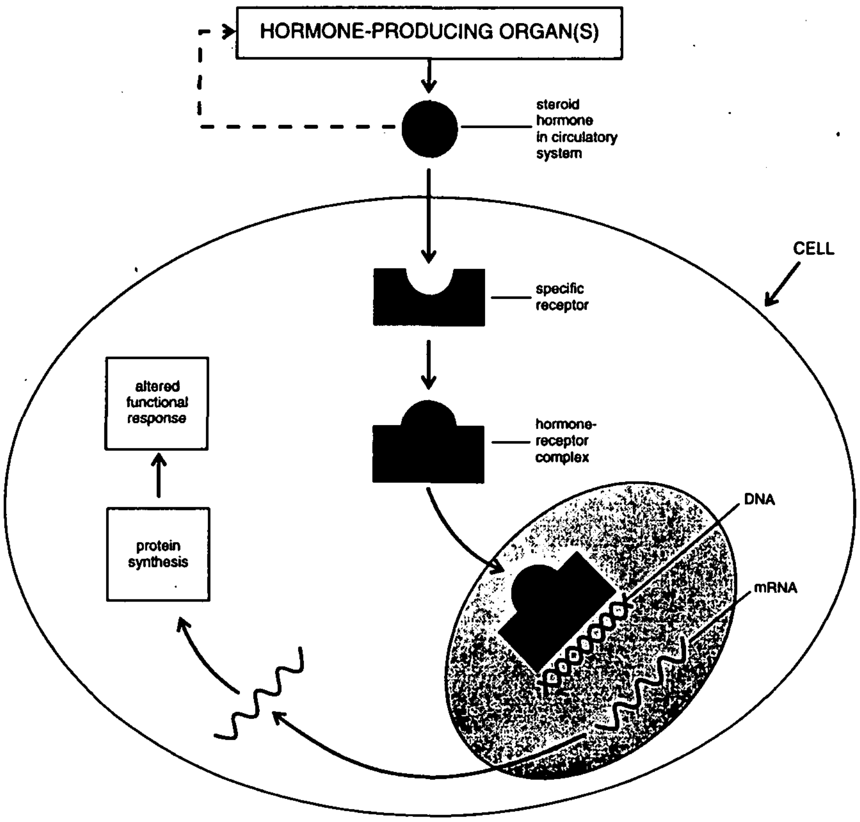
Endocrine Disruptors and the Endocrine System

The endocrine system controls hormone levels in hormone-producing organs, the central nervous system, and the target organs that perform physiological functions. Hormones influence activities in a cell by travelling to various parts of the body through the circulatory system and binding to hormone receptors on cells of the target organ. During prenatal and early postnatal development, a specific ratio of estrogens (female hormones) to androgen (male hormones) is necessary for sexual differentiation and proper formation of reproductive organs (Hileman 1994; Vander et al. 1985). Endocrine disruptors can interfere with the operation of the endocrine system in many ways:

- Binding to cell receptors and mimicking the effect of natural hormones to cause cell activity to occur without being signalled by the brain or endocrine organs;
- Occupying receptors and blocking natural hormones from reaching their destination, thus disrupting endocrine signals to the target cells;
- Reacting directly or indirectly with hormones to alter hormone function and change cell activity;
- Altering the feedback mechanisms that maintain hormone balance; and
- Influencing cells to promote an increase or decrease in the number of receptor sites on individual cells, which magnifies the impact of natural or synthetic hormones on the body.

The interference in the endocrine system by endocrine disruptors can potentially disrupt the reproductive and immune systems and adversely affect metabolism, growth, and behavior.

Hormone Action in Cell



Source: Adapted from Vander et al. (1985).

Some researchers have cautioned that because of the complex interactions involved, proving a cause and effect link between adverse effects in humans and endocrine disruptors will be difficult. Some have doubts about the severity of the effects of endocrine disruptors. They note that these chemicals are only weak hormones; binding of the body's main estrogen, estradiol, to estrogen receptors is up to thousands of times stronger than that of endocrine disruptors (Birnbaum 1994; Stone 1994). In addition, some researchers theorize that the net effect of natural and chemical endocrine disruptors may be zero, especially given that some environmental estrogens are found in plants and have been shown to have beneficial effects, such as inhibiting tumor formation (Stone 1994).

However, recent work on mixtures of endocrine disruptors raises possible answers to these arguments. Researchers found that combinations of two weak endocrine disruptors (e.g., dieldrin and toxaphene) were 10 to 1,600 times more potent than individual compounds in eliciting binding and activation of the estrogen receptor (Arnold et al. 1996). This research is preliminary, but has important implications, especially because pollutants in the environment are typically found in mixtures.

Research Planning Workshop on Endocrine Disruptors (April 1995)

Purpose: To begin developing a national research strategy on endocrine disruptors, in response to growing public concern over their adverse effects.

Findings: The hypothesis that endocrine disruptors cause a variety of adverse effects in wildlife and humans is of sufficient concern to warrant a concerted research effort. Research priorities include: identifying and characterizing effects on developing reproductive systems; and refining exposure assessments and research on toxicology of mixtures.

Outcomes: At a minimum, EPA will publish workshop findings and recommendations in scientific literature, implement some of the identified research initiatives, and form an Endocrine Disruptor Research Coordination Committee.

Source: U.S. EPA 1995c.

Given the potentially broad nature of the problem, the White House has given EPA the task of developing a national research strategy by 1998. EPA recently held a workshop to plan research in this area (see sidebar). In addition, Chemical Industry Institute of Toxicology (CIIT) shifted 10 percent of its total resources in 1995 to endocrine toxicology research and initiated a comprehensive research program to evaluate the potential for selected chemicals to affect the human endocrine system.

In addition to the recent focus on endocrine disruptors, other research efforts relevant to the Great Waters include the release of comprehensive documents on the exposure and health effects of mercury compounds and dioxins:

- EPA has recently submitted the Draft Mercury Study Report to the Science Advisory Board for review (U.S. EPA 1996a). This report addresses the impacts of mercury air emissions from a variety of sources. When submitted to Congress, this report will fulfill CAA section 112(n)(1)(B).[8] One of the volumes focuses on the human health effects of mercury compounds. The human health effects volume was not intended to be an exhaustive survey of the extensive health effects literature available for mercury. Rather, the purpose was to present a brief survey of the studies relevant

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[8] The interim SAB review draft is available through the National Technical Information Service (NTIS).

for assessing potential human health effects and to present more detailed information of those studies that form the basis for EPA's hazard identification and dose-response assessments for three forms of mercury: elemental mercury, mercuric chloride (inorganic mercury), and methylmercury (organic mercury). The three forms of mercury were selected based on data indicating that these are the predominant forms of mercury to which humans are exposed. In addition, examination of the published literature indicates that most health data are on these forms. Additional discussion on the current health effect issues related to mercury exposure, particularly with methylmercury compounds, are discussed later in this section.

- In response to growing scientific controversy regarding the biological, human health, and environmental effects of dioxins and related compounds, EPA began a scientific reassessment of the health risks from exposure to TCDD and chemically-related compounds (known collectively as dioxins and including TCDFs) in April 1991. Activities under the dioxin reassessment included updating and revising the health assessment and exposure assessment documents and performing research to characterize ecological risks in aquatic ecosystems. The dioxin reassessment document was published in draft form in June 1994 and is found in two reports (each 3 volumes): the health assessment document (U.S. EPA 1994c) and the exposure document (U.S. EPA 1994d). More detailed information on the health effects related to TCDDs and TCDFs are discussed later in this section.

Based on the above research areas of interest plus human health effects data from other recently published studies, Table II-7 presents the potential human health effects associated with the Great Waters pollutants of concern, except nitrogen. Nitrogen compounds are not included in this table because nitrogen compounds that are atmospherically deposited generally are not a hazard to human health. (Nitrites in drinking water from wells are of concern in many areas, but are not linked to atmospheric deposition.) For balance in understanding the importance of nitrogen oxides and other nitrogen emissions to the atmosphere, effects on human health due to ozone formation resulting from nitrogen compounds are briefly discussed later in this section. The data in Table II-7 generally are based on a compilation of results from *in vivo* animal laboratory studies, *in vitro* toxicity/cellular studies, and human epidemiological studies describing occupational or accidental exposure to high concentrations of chemicals. Although serious potential effects have been identified for humans, it is difficult to estimate the adverse effects in humans of chronic and low-level exposure to which humans are actually exposed in the environment. New additions to Table II-7 since the First Report are the potential endocrine-disrupting effects of DDT/DDE and lindane, and the "possible human carcinogen" classification for mercury (organic and inorganic forms).

The current information on the health effects associated with each of the pollutant groups is highlighted in the remainder of the section. Recent studies on health effects of the Great Waters pollutants of concern continue to support the findings presented in the First Report. In general, the information presented in this section represent data published from 1992 to 1995. Most of the data describe effects that may occur in humans with long-term exposure to pollutants of concern, which may differ from effects caused by acute, high-level exposures (e.g., accidental spills). In addition, the information presented in this report, with the exception of Table II-7, covers only recent studies, and therefore, cannot be used alone to determine whether these effects are widespread.

TABLE II-7
Potential Human Health Effects Associated With Pollutants of Concern[a]

Pollutant	Potential Effects on Human Health[b]					
	Cancer[c]	Reproductive/ Developmental	Neurological/ Behavioral	Immunological	Endocrine	Other Noncancer[d]
Cadmium and compounds	Probable	●	●	●		Respiratory and kidney toxicity
Chlordane	Probable	●	●	●	●	Liver toxicity
DDT/DDE	Probable	●	●	●	●	Liver toxicity
Dieldrin	Probable	●	●	●	●	Liver toxicity
Hexachloro-benzene	Probable	●	●	●	●	Liver toxicity
α-HCH[e]	Probable					Kidney and liver toxicity
Lead and compounds	Probable	●	●	●	●	Kidney toxicity
Lindane	Probable[f]	●	●	●	●	Kidney and liver toxicity
Mercury and compounds	Possible[g]	●	●	●	●	Kidney toxicity
PCBs	Probable	●	●	●	●	Liver toxicity
Polycyclic organic matter	Probable	●		●		Blood cell toxicity
2,3,7,8-TCDF	Not classifiable	●		●	●	Liver toxicity
2,3,7,8-TCDD	Probable[f]	●	●	●	●	Chloracne
Toxaphene	Probable	●	●	●	●	Cardiovascular effects; liver toxicity

[a] Sources: Cassidy et al. 1994; Chowdhury et al. 1993; Clayton and Clayton 1994; Colborn et al. 1993; Howard 1991; Linder et al. 1992; Soto et al. 1993; applicable ATSDR Toxicological Profiles; and U.S. EPA 1987a, 1987b, 1988a, 1988b, 1990, 1991a, 1993d, 1994c, and 1996.

[b] For purposes of this table, a pollutant 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).

[c] Cancer classifications: (1) "probable human carcinogen" when there is limited or no evidence of human carcinogenicity from epidemiological studies but sufficient evidence of carcinogenicity in animals (corresponds to EPA weight-of-evidence category B); (2) "possible human carcinogen" when there is limited evidence of carcinogenicity in animals and inadequate or lack of human data (corresponds to EPA weight-of-evidence category C); and (3) "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). Data on cancer classifications are obtained from EPA's Integrated Risk Information System, unless otherwise noted.

[d] This column reports 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.

[e] Toxicity data are available primarily for lindane and technical-HCH (a mixture of several HCH isomers); limited data are available for α-HCH.

[f] Data from Health Effects Assessment Summary Tables (HEAST), which classify these chemicals as probable human carcinogens; however, these carcinogenic evaluations are currently under review by EPA.

[g] Inorganic (mercuric chloride) and organic (methylmercury) forms are classified as "possible," whereas elemental is "not classifiable."

MERCURY AND COMPOUNDS

Mercury, a metal, is discussed separately from other metals because of the significant emphasis placed on mercury in section 112 of the CAA and the difference in its behavior and effects compared to other metals. As noted above, a significant amount of information is available in a draft report recently prepared by the EPA on mercury emissions in the United States.[9] One volume of the draft report is devoted to the health effects of mercury and compounds. It has long been known that organic mercury (methylmercury) bioaccumulates in fish, and can biomagnify in the food web. In the air, mercury exists primarily as elemental (Hg^0) and inorganic (Hg^{2+}) mercury. Most of the mercury in water, soil, sediments, or biota is in the form of inorganic mercury salts and organic forms of mercury.

Humans are most likely exposed to mercury indirectly as methylmercury through a diet containing contaminated fish, instead of directly from inhalation of mercury in air. Ingestion of mercury-contaminated fish can result in various health effects, particularly toxicity to the nervous system in adults and in children exposed as fetuses. As shown in Table II-7, mercury can also affect the reproductive and immune systems. Since the First Report, EPA has recommended the current rating of the scientific weight-of-evidence regarding the human carcinogenicity of mercury (U.S. EPA 1996a):

- Elemental mercury: not classifiable as to human carcinogenicity;
- Inorganic (mercuric chloride) and organic (methylmercury) mercury: possible human carcinogens.

In recent years, researchers, including EPA, have focused on several issues related to the health effects of methylmercury: (1) improving the quantitative estimate of the relationship between mercury levels in air and exposure levels; (2) using refined statistical approaches and the application of physiologically-based pharmacokinetic models to evaluate the critical dose levels at which health effects occur from mercury; and (3) effects on nervous system development in populations that consume mercury-contaminated fish. New data are available from a recently-published study investigating neurological effects in children belonging to a fish-consuming population in the Seychelles Islands. Data from evaluation of these children (up to six years of age) have been collected and are being evaluated; subtle neurologic effects are expected to be detected more often in older children. Data from a similar study in the Faroe Islands have been published in abstract form; however, further investigation is being conducted to determine if the study subjects also may have been exposed to PCBs. Smaller scale studies evaluating effects in populations around the Great Lakes also are in progress. The above data and methodologies have not yet been incorporated into an EPA risk assessment because a majority of the new data are either not yet published or have not yet been subjected to rigorous review (U.S. EPA 1996a).

OTHER METALS

Cadmium. As indicated in Table II-7, cadmium has been linked to numerous adverse human health effects, including respiratory and kidney toxicity, probable carcinogenicity, reproductive and developmental effects, and immunological effects. Recent literature

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[9] EPA submitted a review draft of the Mercury Study Report to the SAB in June 1996 (U.S. EPA 1996a). This comprehensive report will fulfill the mandate under CAA section 112(n)(1)(B) when it is submitted to Congress.

discussing the adverse human health effects of exposure to cadmium is limited and generally is focused on how cadmium alters the function of the kidney, which is known to be the critical organ for cadmium exposures (WHO 1992). Cadmium exposure has been found to cause deficient vitamin D metabolism in the kidney, which subsequently affects the calcium balance and bone density and may result in osteoporosis or osteomalacia (both bone diseases characterized by a change in the mineral and matrix phases of bone tissue). Exposure to cadmium has also been linked to developmental effects (e.g., particularly low birth weight).

Lead. Recent literature on human health effects from lead exposure generally supports the findings presented in II-7: lead may affect reproduction and development, alter the immune, nervous, and endocrine systems, and damage the kidney. Lead is also considered a probable carcinogen by EPA. While the correlation between high levels of lead in blood and adverse human health effects is relatively well known, current research has focused on the adverse effects associated with low levels of lead in blood (<30 micrograms of lead per deciliter of blood, or µg/dL) (see sidebar). Low levels of lead may be found in blood of a significant portion of the general public (U.S. EPA 1991b). Effects from low level exposure range from subtle cellular changes, such as effects on red blood cell metabolism, to pronounced effects on physical and mental development (Graziano 1994; Hovinga et al. 1993; Huseman et al. 1992; Kim et al. 1995). Blood lead levels as low as 10 µg/dL (and possibly lower) may result in adverse human health effects (U.S. EPA 1991b). The EPA has also noted that infants and children up to the age of two years may be most susceptible to adverse effects resulting from lead exposures.

Low-Level Lead Exposure

Because of the adverse human health effects associated with low-level lead exposure coupled with the numerous exposure pathways for lead, EPA has abandoned its standard approach for evaluating chemical toxicity in favor of a site-specific modeling approach. In 1994, EPA released a revised version of the *Integrated Exposure Uptake Biokinetic Model for Lead in Children* as its recommended methodology for assessing lead exposure. The model considers the principal lead exposure pathways (e.g., drinking water, diet, inhalation) to estimate lead levels in blood of children. Overall lead intake is then integrated in the model to estimate blood lead concentration. This concentration can then be used to predict: (1) the percentage of the exposed population that will have blood lead levels greater than 10 µg/dL; or (2) the probability that a child exposed to this level will have a blood lead level greater than 10 µg/dL. EPA defines 10 µg/dL blood lead as the lower bound of the range that is known to cause adverse effects on behavior in young children.

Source: U.S. EPA 1994b.

COMBUSTION EMISSIONS

The combustion emissions of present concern are 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD), 2,3,7,8-tetrachlorodibenzofuran (TCDF), and polycyclic organic matter (POM). Polychlorinated biphenyls (PCBs) are also discussed under this pollutant group; they were historically produced for specific uses but are often now released into the environment in combustion gas emissions when PCB-containing materials are burned. The potential human health effects associated with these pollutants are discussed below.

TCDD and TCDF. As shown in Table II-7, the potential human health effects from exposure to TCDD include probable carcinogenicity, and reproductive and developmental, neurological and behavioral, immunological, and endocrine system effects. For TCDF, the potential human health effects are reproductive and developmental, immunological, and endocrine system effects, and liver toxicity. The dioxin reassessment document (U.S. EPA 1994c) discusses the effects of TCDD and related (compounds collectively referred to as dioxins) in detail. A summary of some of the research presented in the reassessment is discussed below.

- *Carcinogenicity.* A number of new studies provide further evidence that dioxins are probable human carcinogens. Because available human studies alone cannot confirm whether there is a causal relationship between dioxin exposure and increased cancer incidents, assessors have extrapolated from available animal data to potential human cancer. Recent animal studies have demonstrated dioxins to be carcinogenic in hamsters and small fish. All of these data have contributed to the weight-of-evidence that dioxins and related compounds may be carcinogenic, under certain circumstances, in humans.

- *Reproductive and Developmental Effects.* Research suggests that effects on reproduction and development may occur at concentrations lower than originally thought. A study of monkeys found that chronic exposure to TCDD increased the risk of endometriosis, a female reproductive system disorder that can produce infertility or even death. Human epidemiologic studies on the relationship between dioxin exposure and endometriosis are planned as part of follow-up research to a 1976 dioxin release in Seveso, Italy.

- *Immune System Effects.* Recent animal studies confirm that dioxins may cause immunological effects and suggest that some effects may occur after low-level exposure. There is conflicting evidence, however, concerning the effects of these compounds on humans. A developing human immune system is believed to be particularly sensitive to the effects of exposure to dioxins compared to adults, but additional research is necessary to confirm this hypothesis.

- *Endocrine System Effects.* Two human epidemiologic studies linked exposure to TCDD with changes in male reproductive hormone levels. Two of three studies found decreased testosterone levels and one of two studies observed an increase in a female ovulation hormone in males. Animal studies have produced similar results.

- *Other Noncancer Effects.* Chloracne is a severe acne-like disorder that develops within humans after a few months of exposure to dioxin. It may disappear in some individuals after exposure is discontinued, or persist in others for many years. Limited data exist to determine the doses at which chloracne is likely to occur, but long lasting, high-intensity exposures that begin at an early age are believed to increase an individual's chances of contracting this skin disorder. Another potential effect is enzyme induction which has occurred in animals exposed to TCDD compounds. The biochemical alteration may either benefit the animal or result in adverse effects to the animal (i.e., alter metabolism of certain chemicals by increasing or decreasing toxicity). Studies linking this effect directly to humans are not available.

One of the main issues of current interest is the mechanism of action of dioxins (i.e., how they bring about the effects). The unusual potency of dioxins in eliciting toxic effects suggested to researchers that a dioxin receptor existed. Based on a substantial amount of biological and genetic evidence, an intracellular protein called the aryl hydrocarbon receptor (Ah receptor) is believed to mediate biological responses to TCDD and related compounds.

Figure II-2 presents a simplified diagram of how biological responses to dioxin-like compounds are mediated by the Ah receptor. Although our understanding of the Ah receptor is limited, mammals (including humans), birds, and fish all have exhibited detectable concentrations of the Ah receptor in a number of different tissues. In laboratory mammal studies, binding of dioxins to the Ah receptor has been associated with weight loss, edema, liver damage, immune system toxicity effects on reproduction, and promotion of tumors (Fox 1993). Researchers believe that responses to TCDD are likely to vary within and between species, as well as between tissues in individual species, based on differential responses to Ah-receptor binding. Several proteins are believed to contribute to TCDD's gene regulatory effects; the response to TCDD probably involves a complex relationship between multiple genetic and environmental factors.

FIGURE II-2
Role of Ah Receptor in Biological Responses to Dioxin Exposure

Dioxin Exposure
↓
Free Dioxin in Tissues
↓
Dioxin Binding to the
Ah Receptor in Cells
↓
Ah Receptor-Dioxin Complex
Binding with DNA
↓
Gene Regulation
↓
m-RNA Regulation
↓
Protein Synthesis
↓
Biochemical Alterations
↓ Interactions of
↓ Multiple Target
↓ Genes
Early Cellular Responses
(cell growth stimulation)
↓
Late (irreversible) Tissue Response
(cancer, deformity)

Source: Adapted from EPA 1994c.

PCBs. PCBs represents a class of halogenated aromatic compounds with similar chemical structure (i.e., contains 12 to 68 percent chlorine). This section presents health effects that are generally common to all PCB compounds, although the levels at which these effects may occur vary between compounds. In addition, specific compounds may produce their own range of effects. As shown in Table II-7, PCBs are classified as probable carcinogens, and potentially affect reproductive, neurological, immunological, and endocrine processes in the human body. Recent research suggests that PCBs may be able to act as endocrine disruptors in ways similar to many pesticides and dioxins (McKinney and Waller 1994).

PCBs remain a significant concern because, although they have not been produced in this country for over a decade, appreciable concentrations are still found in water, and animal and human tissue, milk, and blood. As reported in Section II.A, the majority of fish advisories in the Great Waters are due to the presence of PCBs.

One recent study comparing Great Lakes fish eaters to a control population showed that PCB levels, as shown in Table II-8, did not change substantially over a seven-year period (Hovinga et al. 1992). The study proposes a number of reasons for the static PCB levels in the fish-eater population: (1) restrictions on PCB production alone may not ensure decreasing levels of PCB exposure in human populations; (2) other sources of PCB contamination may be major sources of exposure; or (3) because of the persistent nature of PCBs, seven years may not be long enough time to see a decrease in body burdens of PCBs in human populations.

Changes in Serum PCB Levels in Great Lakes Populations

Hovinga et al. (1992) compared data from a 1982 study that examined 115 individuals who consumed Great Lakes fish and 95 who did not eat fish (i.e., controls) to a similar study in 1989 that reevaluated these individuals. Comparison of the data from the two studies (Table II-9) shows a *significant decrease in mean serum DDT levels in both fish-eaters (almost 40 percent) and controls* (approximately 30 percent). In contrast, serum PCB levels decreased only slightly in fish-eaters and remained relatively constant in controls. These results support other studies that have found stable PCB levels over time when DDT levels have been decreasing and suggest that, despite their ban, restrictions on PCBs have not been as effective in reducing levels of contamination in humans as those placed on DDT.

TABLE II-8
Mean Serum PCB and DDT Levels in Fish Eaters and Controls (1982 vs. 1989)[a]

	Fish Eaters		Controls	
	1982	1989	1982	1989
Serum PCB (ppb)	20.5	19.0	6.6	6.8
Serum DDT (ppb)	25.8	15.6	9.6	6.8

[a] Values represent 111 fish eaters and 90 controls (out of original 115 and 95, respectively).

Potential sources of PCBs other than through fish consumption are disposal of previously manufactured products containing PCBs; atmospheric transport from other countries where PCBs are still produced and used; and cycling of PCBs in the environment. The widespread use of PCBs in electrical equipment is of concern, but there has been an increased awareness of the combustion of waste containing PCBs. The leveling off of PCB concentrations in the environment suggests that there is a reservoir of PCBs from "old" pollution that cycles between sediments, water, and air.

In view of this evidence of continuing persistence of.PCBs in the environment and in the human population, effects on humans related to this group of chemicals remain a concern and many studies assessing their effects are currently underway. Recent findings include:

- Delayed puberty, fertility impairment, and irregular cycle patterns were observed in female rat pups given doses of PCBs (Sager and Girard 1994). The investigators suggested that these reproductive effects may be related to impairment of endocrine function.
- The effects of prenatal exposure to PCBs on neurological function in children has been investigated in a follow-up study of children from the contaminated rice oil incident in Taiwan. Researchers concluded that PCB exposure to the fetuses may impair the psychological functions of the brain in the child and that PCBs may persist in the brain for a long period (Chen and Hsu 1994).
- In response to evidence that the development of cancer may involve miscommunication between cells, a recent study investigated the effect of PCB

exposure on intercellular communication in human breast cells and found that cell communication was inhibited with increasing doses (Kang et al. 1996).

- One study found that consumption of PCB-contaminated sport fish from Lake Ontario did not increase the risk for spontaneous fetal death, which has been observed in various mammalian species after PCB exposure (Mendola et al. 1995).

As discussed in the recently published health effects by reassessment for dioxin, some PCBs have been found to produce similar effects as dioxins.

POM (Polycyclic Organic Matter). POM refers to a class of chemicals containing many organic cyclic structures. As shown in Table II-7, adverse human health effects associated with POM include reproductive and immunological effects, as well as probable carcinogenicity. Health effects data on POM are available primarily for polycyclic aromatic hydrocarbons (PAHs), one group of POM chemicals. Recent literature on health effects of PAHs corroborates previous findings and investigates the mechanism of action for PAH effects.

- *Carcinogenicity.* In animals, PAHs have been found to induce skin, stomach, lung, and mammary gland tumors (Beland and Poirier 1993). There is some speculation that ultraviolet light acts as a cofactor in the development of PAH-associated skin cancer, although this hypothesis requires further evaluation (Saboori and Newcombe 1992).

- *Reproductive Toxicity.* It has been demonstrated that short-term *in vitro* exposure to two PAHs (benzo[a]pyrene and 3-methylcholanthrene) increases the secretion of placental human chorionic gonadotrophin (HCG) (a human birth hormone) in the first trimester. This effect was maintained after the PAHs' application was discontinued and it may be involved in adverse reproductive effects observed with these pollutants (Barnea and Shurtz-Swirski 1992).

- *Immunological Toxicity.* Exposure to PAHs in young experimental animals during development of the immune system can cause severe and sustained postnatal

Workshop on Developmental Neurotoxic Effects Associated With PCB Exposure

In September 1992, EPA sponsored a workshop on the developmental nervous system effects of PCBs. At this meeting, experts addressed whether currently available health effects data are sufficient to support developmental neurotoxicity data in risk assessment. Some current findings regarding human health effects associated with PCBs were discussed and are summarized below.

- Women exposed to PCBs in the Yusho incident experienced irregularities in menstrual cycles, which may suggest alteration in ovarian function.

- Nondevelopmental nervous system effects include headaches, numbness, altered peripheral nerve function, and decreased neurobehavioral function measured through visual memory and problem solving ability. In animals, PCB exposure has been shown to affect the actions of dopamine (a brain neurotransmitter).

- PCB exposure is associated with atrophy of the thymus and immunosuppression in animals (i.e., inhibition of immune cells required for tumor resistance).

- Chloracne and liver dysfunction are associated with occupational exposure to PCBs in humans. In animal studies, increased mortality, skin ailments, hepatotoxicity, and weight loss have been demonstrated.

Source: U.S. EPA 1993b.

immunosuppression (i.e., inhibition of immune cells necessary for tumor resistance) (Holladay and Luster 1994).

PESTICIDES

The pesticides of concern for the Great Waters are chlordane, DDT/DDE, dieldrin, hexachlorobenzene, α-HCH, lindane, and toxaphene. As shown in Table II-7, these pesticides are probable human carcinogens and potentially result in toxic effects to reproductive, immune, and endocrine systems, as well as other noncancer effects.

Much of the recent literature concerning adverse health effects from exposure to the pesticides of concern in the Great Waters discusses their estrogenic and other hormonal effects (see earlier discussion of endocrine disruptors), as well as mechanisms of action of these effects (Chowdhury et al. 1993; Foster et al. 1992a, 1992b; Johnson et al. 1992; Juberg and Loch-Caruso 1992; McNutt and Harris 1993). New additions to Table II-7 in this Report include the recognition of DDT/DDE and lindane as potential endocrine disruptors.

Current research that has further explored the mechanisms of action of the pesticides in producing their known effects, other than endocrine disruption, are summarized below.

- *Carcinogenicity.* In response to evidence that the development of cancer may involve miscommunication between cells, a few recent studies have investigated the effects and mechanism of action of cancer-promoting chemicals, including dieldrin, DDT, lindane, and toxaphene, on intercellular communication (Kang et al. 1996; Leibold and Schwarz 1993; Rivedal et al. 1994; Tateno et al. 1994). They have found that cell communication is inhibited with increasing doses of these chemicals.

- *Reproductive and Developmental Effects.* In addition to studies of reproductive effects associated with endocrine disruption, recent studies discuss the effects and accumulation of the pesticides in reproductive organs (e.g., ovary) (Bourque et al. 1994; Lindenau et al. 1994; Singh et al. 1992). One recent study concluded that DDT is not toxic to sperm (Linder et al. 1992).

- *Nervous System Effects.* Studies on the adverse effects from exposure to neurotoxic pesticides provided further support of their adverse neurological effects, investigated the mechanisms of action producing these effects, the specific regions of the central nervous system affected (e.g., the motor primary cortex and hippocampus in the brain), and whether there is a critical exposure period for these effects (Barrón et al. 1993; Eriksson et al. 1992, 1993; Gilbert and Mack 1995; Goldey and Taylor 1992; Gopal et al. 1992; Johannson et al. 1993; Kilburn and Thorton 1995; Nagata et al. 1994; Rivera et al. 1992). Many of these studies evaluated nervous system effects during development in young animals, and determined that progression of the effects, as well as severity of these effects, is influenced by the time of exposure (e.g., formation of the nervous system is a sensitive stage of development). Data also suggest that some effects of early exposure to pesticides on certain regions of the central nervous system may be reversible.

- *Immune System Effects.* Studies investigating the mechanisms of action for producing immunological effects and the effects on specific organs in the immune system (e.g., spleen) have confirmed the potential adverse immunological effects related to these pesticides. Immune system effects (suppression, autoimmunity) were observed in humans with elevated levels of chlordane (McConnachie and Zahalsky 1992). Animal data have demonstrated that immune system stimulation (i.e., increased antibodies) and/or suppression (e.g., decreased production of certain antibody-forming cells) may result with exposure to DDT, dieldrin, HCB, and lindane (Flipo et al. 1992; Meera et al. 1992; Rehana and Rao 1992; Saboori and Newcombe 1992; Saha and Banerjee 1993; Schielen et al. 1993).

- *Other Noncancer Effects.* A review of literature on human health effects from long-term exposure to pesticides concluded that (1) chlordane may cause disorders in bone marrow; (2) DDT can cause chloracne, chromosome aberrations, tremors, muscular weakness, and high levels of cholesterol and triglyceride; and (3) hexachlorobenzene may cause metabolism abnormalities and liver cancer (Maroni and Fait 1993). Another recent study demonstrated that lindane may impair development of blood cells in the bone marrow (Parent-Massin and Thouvenot 1993).

NITROGEN COMPOUNDS

Nitrogen in the molecular form N_2 is the most abundant gas in the earth's atmosphere, and is essentially chemically inert under normal conditions. Other compounds of nitrogen, particularly oxides of nitrogen, are common in the air due to industrialization, and produce a variety of effects on human health and the environment. EPA has several programs, in addition to the Great Waters program, that are evaluating and attempting to reduce the threat to human health from atmospheric nitrogen dioxide, ozone, and acidic precipitation or particulates (U.S. EPA 1995d, 1995e, and 1995f). These other programs also examine general environmental effects of the nitrogen compounds, while the Great Waters Program concentrates on the over-fertilization, or eutrophic, effects in waterbodies from atmospheric deposition of nitrogen compounds (see Section II.D on ecological effects).

Oxides of nitrogen are produced abundantly by many modern combustion processes. Of the oxides of nitrogen, nitrogen dioxide (NO_2) is the most abundant in emission plumes or vehicle exhaust. At times, NO_2 itself can reach ambient concentrations associated with a variety of acute and chronic health effects. A more common problem is that, in the presence of sunlight, nitrogen oxides in the atmosphere react with volatile organic compounds (VOCs) to form ozone. Although ozone in the stratosphere is essential for protecting the earth from harsh ultraviolet rays, lower level ozone (or photochemical smog) contributes to a variety of health problems. Additional problems are posed by fine particles in the atmosphere. Under certain conditions in the air, oxides of nitrogen can undergo additional reactions resulting in fine particles or contribute to acids in aerosol droplets. EPA has set health-based National Ambient Air Quality Standards (NAAQS) for NO_2, for ozone, and for fine particulates (which have many sources in addition to nitrogen oxides). Acidic precipitation is also affected by nitrogen oxide emissions, and is the focus of another EPA program. Recent publications from those programs and NAAQS reviews should be consulted for details on the human health effects of nitrogen. The Great Waters program evaluates the reductions in nitrogen compounds that these programs have achieved and have proposed, and considers the net effects on waterbodies, but does not analyze direct human health effects from inhalation.

II.D Ecological Effects

Information presented in this section is based on effects observed in laboratory and field studies. Some of these observed ecological effects of certain pollutants of concern were investigated in the Great Waters.

Conclusions from the First Report to Congress

In the First Report to Congress, EPA identified many adverse ecological effects, at both the individual species level and the ecosystem level, associated with the selected pollutants of concern. In addition, the First Report discussed eutrophication of estuarine waters, which is the main ecological effect relevant to this Report associated with nitrogen loading. Research findings and studies presented in the First Report led to the following general conclusions concerning ecological effects:

- The selected pollutants of concern have been linked to a broad range of effects at the individual species level in aquatic organisms and other wildlife, including effects on the reproductive, nervous, immune, and endocrine systems, and changes in enzyme functioning.

- Reproductive effects of certain pollutants of concern include reduced fertility, increased embryo toxicity, reduced hatchability, reduced survival of offspring, abnormalities in offspring, parental behavior change, and changes in mating behavior (e.g., impaired hormone activity, changed adult sexual behavior). For example, eggshell thinning in a number of bird species and associated reproductive loss are linked to exposure to DDT (and its metabolite DDE) in the 1960s and 1970s. Recent decreases in environmental concentrations of reproductive pollutants of concern, such as DDT and PCBs, are correlated with population recoveries in many bird and other wildlife species; however, some populations in certain regions of the Great Lakes still exhibit higher rates of reproductive failure than in other areas.

Pollutants of Concern for Reproduction

Pollutants of concern that have been linked with reproductive impairment in aquatic and terrestrial wildlife include cadmium, DDT/DDE, dieldrin, lead, lindane, mercury, PCBs, and 2,3,7,8-TCDD.

- Effects on the nervous and endocrine systems may occur at very low exposure levels. For example, wild populations of Great Lakes herring gulls, Forster's terns, and ring-billed gulls exposed to various pollutants of concern have exhibited behavioral changes such as female-female pairings, which result in abnormal incubation activities and nesting behavior, including nest abandonment.

- Several of the pollutants of concern cause changes in enzyme functioning. Studies reported that the activity of enzymes responsible for the breakdown of foreign compounds is greatly increased by most of the pollutants of concern. In fish, the increased activity of these enzymes has been shown to result from exposure to PCBs and PAHs. 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 pollutants such as TCDD.

- Exposure of communities of bottom-dwelling aquatic species in the Great Lakes to toxic chemicals has resulted in significant changes in species diversity and populations. In addition, fish-eating birds such as bald eagles, herring gulls, and Forster's terns in the Great Lakes region have undergone significant population declines since the 1960s. Only in recent years, as water concentrations of pollutants in the Great Lakes have declined, have some species begun to recover. Certain current population recoveries of fish-eating birds are still dependent on migration to Great Lakes breeding colonies from other areas.

- Eutrophication[10] is one of the most serious pollution problems facing estuarine waters of the United States. Atmospheric deposition of various nitrogen compounds (mostly nitrates and ammonium) can contribute significantly to eutrophication in coastal waters where productivity is usually limited by nitrogen availability. Accelerated eutrophication results in severe ecological effects such as nuisance algal blooms, dieback of underwater plants (due to reduced light penetration), reduced oxygen levels in the water, 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.

Current Understanding of Ecological Effects

Since the First Report, new information on the pollutants of concern and their effects to aquatic and terrestrial wildlife has become available. This section first discusses some notable research on ecological effects relevant to some Great Waters pollutants of concern (i.e., emerging field of endocrine disruptors, EPA's Mercury Study Report and dioxin reassessment). Next, a brief overview of recent data gathered on the Great Waters pollutants of concern is presented, followed by a more detailed summary of effects data specific for each pollutant of concern.

The role of endocrine disruptors in causing adverse effects is an emerging and controversial issue. As discussed in Section II.C, many of the pollutants of concern are suspected endocrine disruptors. Recent articles demonstrating endocrine disruption by Great Waters pollutants of concern in aquatic and terrestrial wildlife are summarized below.

- Scientists observing alligators in a Florida lake found that 75 percent of their eggs were dead or infertile and that almost 60 percent of juvenile male alligators had severely shrunken penises; the researchers linked these effects to a large spill of a pesticide containing DDT, which has been identified as a endocrine disruptor (Gross and Guillette 1991). More recent research found that in eggs and young alligators, females had an estrogen to testosterone ratio twice as high as normal and males had a ratio similar to normal females, but with abnormal concentrations (i.e., almost no testosterone and no estrogen). Females also exhibited deformed ovaries that contained abnormal eggs (Raloff 1994). A recent study comparing the alligators in this lake to a control lake further supported the hypothesis linking these effects to

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[10] Eutrophication is an overabundance of nutrients (e.g., nitrogen) in a water body. It is a natural process that typically takes place over hundreds of years, but can be greatly accelerated by additions of nutrients from human activities.

endocrine disruption (Guillette et al. 1994). Studies into the mechanism of action of these effects are ongoing (Guillette et al. 1995).

- Researchers have been studying the mechanism of action of endocrine disruption in a common turtle species (the red-eared slider) and the African clawed frog (Palmer and Palmer 1995; Raloff 1994). Their work has focused on a potential biomarker for exposure to endocrine disruptors called vitellogenin (egg-yolk protein in the blood of egg-laying vertebrates). When stimulated by estrogen, the liver produces this protein and dumps the protein into the bloodstream, where it then circulates to the ovaries and is deposited into an egg. Usually only females possess a sufficient amount of estrogen to produce vitellogenin; however, DDT has induced vitellogenin production in male turtles and frogs. In another study, PCBs and lindane induced estrogen receptor and vitellogenin accumulation in rainbow trout liver (Flouroit et al. 1995).

- Many egg-laying reptiles depend on the temperature of the incubating egg to determine the sex of the offspring. PCBs applied to the shells of turtle eggs during the period of sexual differentiation counteracted male-producing temperatures and induced ovarian development (Bergeron et al. 1994). Further study of the mechanism of action and synergistic effects of different PCB congeners are ongoing (Crews et al. 1995).

- Researchers have recently been testing the hypothesis that endocrine disruptors, such as DDE, mercury, and PCBs, are playing a role in the decline of the endangered Florida panther population (Facemire et al. 1995; Raloff 1994). (Many have considered inbreeding the main reason up to this point (Facemire et al. 1995)). A large percentage of males have exhibited abnormal reproductive organs, sterility, and production of abnormal or deformed sperm. Both males and females exhibit abnormal hormone ratios, with little difference in estradiol levels between males and females (i.e., evidence that males have been demasculated and feminized).

In addition to the recent focus on endocrine disruptors and their impact on ecological health, other research efforts relevant to ecological effects of pollutants of concern include the release of the Mercury Study Report and an interim EPA report on TCDD exposure:

- EPA has recently submitted the Draft Mercury Study Report to the Science Advisory Board for review (U.S. EPA 1996a). This report addresses the impacts of mercury air emissions from a variety of sources. When submitted to Congress, this report will fulfill CAA section 112(n)(1)(B).[11] Volume 5 of the Mercury Study Report provides an overview of the ecological effects of mercury in plants, invertebrates, fish, birds, and mammals, and reviews available toxicity and bioaccumulation data for the purpose of developing a criterion for the protection of sensitive wildlife species. Furthermore, the Mercury Report presents a conceptual model describing how airborne mercury accumulates in aquatic biota, biomagnifies in aquatic food chains, and is consumed by wildlife that eat contaminated fish.

- Dioxin has been classified as the most potent known animal carcinogen, and as a probable human carcinogen, since 1985. Increased concerns that dioxins in aquatic environments may be a major contributor to overall human dioxin exposure through

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[11] The interim SAB review draft is available through the National Technical Information Service (NTIS).

fish and shellfish consumption, as well as increased evidence of its hazard to fish and wildlife, prompted EPA to reassess dioxin's effects on aquatic ecosystems. Work on characterizing ecological risks is in progress at EPA's Mid-Continent Ecology Division of the National Health and Ecological Effects Research Laboratory (NHEERL) in Duluth, Minnesota. EPA recently published the *Interim Report on Data and Methods for Assessment of 2,3,7,8-Tetrachlorodibenzo-p-dioxin Risks to Aquatic Life and Associated Wildlife* (U.S. EPA 1993a) on this research. The goal of the report was to review and evaluate relevant published and unpublished data and models currently available for analyzing dioxin exposure to and effects on aquatic life and wildlife. Information on related compounds, such as TCDF and PCBs, is not discussed in detail in the interim report; however, it is expected that the final report will assess the contribution of these related compounds to the risk for aquatic life and wildlife. The interim report findings are presented with the discussion of TCDD in this section.

Based on new information from the above research on ecological effects of pollutants of concern, as well as findings from recently published data, a summary of the potential effects of the pollutants of concern is presented in Tables II-9 and II-10. For each effect attributable to a pollutant of concern, the tables identify the organism type(s) in which the effect has been observed (i.e., plants, invertebrates, fish, amphibians/reptiles, birds, and mammals). Table II-9 presents data on the following types of effects: cancer, reproductive/developmental, immunological, metabolic/enzyme, and neurological/behavioral effects. Table II-10 presents data on other noncancer effects: death; damage to the kidney, liver, heart, lungs or gills, or gastrointestinal tract; exterior changes; and decreased growth or biomass. The information in these tables is based on both field and laboratory studies. This Report does not attempt to assess how the observed effects on individual animals contributes to changes in species population within an ecosystem.

Pollutants of Concern and Health Effects in St. Lawrence Estuary Beluga Whales

A 9-year epidemiological study in an isolated population of beluga whales documented higher levels of many pollutants of concern, including dieldrin, DDT, HCB, lead, mercury, PAHs, and PCBs, in these whales compared to Arctic belugas (Beland et al. 1993). The St. Lawrence whales exhibited high prevalence of tumors; high incidence of lesions to the digestive system, mammary glands, and other glandular structures; some evidence of immune system suppression; and frequent tooth loss and gum disease. No such lesions were observed in Arctic belugas or in other St. Lawrence aquatic mammals (e.g., seals). Researchers propose this case as a model for the potential long-term consequences of pollutants in the environment on human health (De Guise et al. 1995).

Following the tables, recent information on the ecological effects of the pollutants of concern is discussed by pollutant group. The ecological effects of nitrogen (i.e., eutrophication and its consequences) also are discussed in this section. In general, the information presented in the following section represents data published from 1992 to 1995. Most of the data describe effects that may occur in wildlife with long-term exposure to pollutants of concern, which may differ from ecological effects caused by acute, high-level exposures (e.g., accidental spills). In addition, the information presented in this report covers only recent studies, and therefore, cannot be used alone to determine whether these effects are widespread in the environment. Note that the adverse effects on ecological health caused by exposure to toxic contaminants are not often easy to distinguish from other stresses. For example, fish populations in the Great Lakes suffer from habitat loss, overfishing, and the

TABLE II-9
Potential Ecological Effects of the Pollutants of Concern

Pollutant of Concern	Potential Ecological Effects[a]					Other Noncancer Effects
	Cancer	Reproductive/ Developmental[b]	Immunological	Metabolic/ Enzyme	Neurological/ Behavioral	
Cadmium and compounds		I F B		F B M	I B	See Table II-10
Chlordane	M	I F A B M	M	I M	F B M	
DDT/DDE		I B A M	M		F B	
Dieldrin		A			B	
HCB		I F			I B	
α-HCH						
Lead and compounds		I F A B M	F A	I F B M	F A B M	
Lindane		I	F		I	
Mercury and compounds	M	I F A B M	B	P F B	I F B M	
PCBs	M	I F B M	F M	F B M	F B M	
POM (PAHs)	F A M	I F B M	F B M	F	I F	
TCDDs		F B M	M		B	
TCDFs						
Toxaphene	M	I F B M			A F B	

[a] Blank areas indicate that no data were found correlating the effect with the pollutant of concern (as opposed to data found indicating no correlation between the effect and the pollutant of concern).

[b] Includes endocrine-disrupting effects.

Key: P = Plants I = Invertebrates F = Fish A = Amphibians/Reptiles B = Birds M = Mammals

Sources: Arkoosh et al. 1994; Baturo et al. 1995; Constable and Orr 1994; Di Pinto et al. 1993; Doust et al. 1994; Dunier and Siwicki 1994; Eisler 1985, 1986a, 1986b, 1987a, 1987b, 1988, 1990; Eisler and Jacknow 1985; Ferrando et al. 1995; Fitchko 1986; Geyer et al. 1993; Government of Canada 1994; Hermsen et al. 1994; Hill and Nelson 1992; Hugget et al. 1992; Huggett et al. 1993; Johnson et al. 1993; Lahvis 1995; Malbouisson et al. 1994; Schulz and Liess 1995; Tidou et al. 1992; Trust et al. 1994; and U.S. EPA 1993a, 1993b, 1993c.

TABLE II-10
Potential Other Noncancer Ecological Effects[a]

Pollutant of Concern	Potential Noncancer Ecological Effects[b]							
	Death	Kidney Damage	Liver Damage	Heart Damage	Lung/Gill Damage	Gastrointestinal Damage	External Damage	Decreased Growth/Biomass
Cadmium and compounds	I F	B		B				I F B
Chlordane	I F A B M	M	M	M	M	M		I M
DDT/DDE	P I							P
Dieldrin	B						B	
HCB	P I F B		F B					P I B
α-HCH								
Lead and compounds[c]	I F A B M	F B	F B		F	A M	F B	I M
Lindane	I							I
Mercury and compounds[c]	I F A B M	B M	B		M			I F B
PCBs	P I F B M	F M	B M			B M	F B M	P I F B
POM (PAHs)[c]	I F M	M	F M			M		F B
TCDDs	F B M	M	F M		M	M	F M	F
TCDFs								
Toxaphene	I F A B M	F B				F	F B	I F B

[a] This table uses the same references as those cited in Table II-9.

[b] Blank areas indicate that no data were found correlating the effect with the pollutant of concern (as opposed to data found indicating no correlation between the effect and the pollutant of concern).

[c] Also causes other noncancer effects in plants, such as leaf and root damage, chlorophyll decline, and reduced photosynthesis.

Key: P = Plants I = Invertebrates F = Fish A = Amphibians/Reptiles B = Birds M = Mammals introduction of non-native species, in addition to the effects discussed below (U.S. EPA 1995a). In addition, studies on ecological effects generally do not determine the exposure pathway of the pollutants (e.g., atmospheric deposition).

MERCURY AND COMPOUNDS

The data presented in Tables II-9 and II-10 summarize the effects of mercury exposure on plants, invertebrates, fish, birds, and mammals. EPA's draft report on mercury emissions in the United States includes a discussion of the ecological effects of mercury (U.S. EPA 1996a);[12] a summary of the research findings is presented below:

- The effects of mercury on plants include death, growth inhibition, leaf or root damage, chlorophyll decline, and reduced photosynthesis.
- In fish and aquatic invertebrates, mercury can cause death, reduced reproductive success, impaired growth and development, and behavioral abnormalities.
- Effects of mercury on birds include death, liver and kidney damage, neurobehavioral effects, impaired growth and development, and reproductive effects. Reproductive effects are the primary concern for avian mercury poisoning and can occur at dietary concentrations well below those that cause overt toxicity.
- Extensive research on the toxicity of mercury to mammals indicates that effects vary depending on the form of mercury ingested, with methylmercury being the most toxic form. Methylmercury ingestion by mammals can cause death, neurological and behavioral effects, and damage to the heart, lung, liver, kidney, and stomach.

Population-level Effects of Mercury

Studies conducted on various communities have shown mercury to: (1) reduce species diversity of freshwater, brackish-water, and soil microbial communities; (2) reduce carbon fixation in phytoplankton communities; and (3) change the species composition of phytoplankton in an aquatic community. Although clear causal links between mercury contamination and population declines in various wildlife species have not been established, mercury may be a contributing factor to population declines of the endangered Florida panther and the common loon. Other researchers have concluded, however, that mercury levels in most areas are not sufficient to adversely affect bird populations.

Source: U.S. EPA 1996a.

OTHER METALS

Cadmium. As shown in Tables II-9 and II-10, adverse effects from exposure to cadmium have been demonstrated in invertebrates, fish, and birds. Recent studies have focused on the toxicity of cadmium to aquatic species (Kraak et al. 1992; Weinstein et al. 1992) and the identification of potential indicator species for cadmium exposure (Naimo et al. 1992). For example, researchers concluded that accumulation of metals, including cadmium, causes lesions to the shell of blue crabs in the Albemarle-Pamlico Estuarine System (Weinstein et al. 1992).

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[12] EPA submitted a review draft of the Mercury Study Report to the SAB in June 1996 (U.S. EPA 1996a). This comprehensive report will fulfill the mandate under CAA section 112(n)(1)(B) when it is submitted to Congress.

Lead. Many noncancer ecological effects are associated with lead exposure, as shown in Tables II-9 and II-10. However, the availability of recent studies on the ecological effects of lead is limited because of the strong interest currently focused on the neurological and behavioral effects in infants and children. One recent study demonstrated that exposure to waterborne lead may result in multiple effects on reproductive behavior and overall reproductive success in fathead minnows (Weber 1993). A recent review of wildlife contamination in Chesapeake Bay cited older articles that found lead to adversely affect embryo development and neurological behavior in the green and bull frogs (Heinz et al. 1992).

COMBUSTION EMISSIONS

TCDD. As shown in Tables II-9 and II-10, dioxins are associated with effects in fish, birds, and mammals, including reproductive effects and other noncancer effects (e.g., kidney and liver damage). EPA's assessment of ecological effects in aquatic life and associated wildlife discusses the effects of TCDD in detail (U.S. EPA 1993a); a summary of the research findings is presented below:

- Most dioxins are thought to affect animals and humans by binding with the Ah receptor. See Figure II-2 in Section II.C and the accompanying discussion for further explanation of the Ah receptor's mechanism of action. The Ah receptor has been detected in mammals (including humans), avian wildlife, and certain fish species, but valid data are not yet available to confirm its presence in amphibians, reptiles, or aquatic invertebrates.

- Available laboratory toxicity information indicates that aquatic invertebrates, plants, and amphibians are substantially less sensitive to TCDD than fish. The report emphasized that the data are limited and are not always comparable between species.

- The data indicate that young fish are more sensitive to TCDD than older fish. Fish fry were most sensitive to effects from TCDD following exposure of eggs before or shortly after fertilization.

- In mammals and birds, the primary effect of concern is reproductive impairment. In addition, the species most at risk from adverse effects from TCDD exposure are those species whose diet consists mostly of fish.

Ecological Effects of TCDD in Lake Trout Population in Lake Ontario

Commercial over-fishing and sea lamprey predation were originally thought to be the primary causes of lake trout decline in Lake Ontario. However, several findings suggest that TCDD toxicity alone could have caused the widespread lake trout reproductive failure in Lake Ontario in the post-1950s: (1) the strong association between the occurrence of blue-sac disease (a stress syndrome) in sac-fry from TCDD-exposed fish in laboratory studies and from eggs collected from Lake Ontario; (2) the historical record of lake trout exposure to TCDD and related chemicals in Lake Ontario; and (3) the consistency of predicted toxicity with the field sampling record of no natural reproduction until 1986. Attainment of a self-sustaining population of lake trout in Lake Ontario depends on further reduction of TCDD and related chemicals, in addition to sea lamprey control, maintenance of adequate spawning beds and water quality, and introduction of lake trout strains appropriate for Lake Ontario's conditions and food web.

Source: U.S. EPA 1993a.

In a recent study, nesting wood ducks in an Arkansas estuary were found to have reduced productivity (e.g., embryo anomalies, decreased hatching success) related to dioxin exposure (White and Hoffman 1995). In another study, the brains of great blue heron hatchlings exposed to TCDD developed asymmetrically, an indicator of potential neurotoxic effects (Henschel et al. 1995).

PCBs. As shown in Tables II-9 and II-10, PCBs are associated with adverse effects in plants, invertebrates, fish, birds, and mammals. These effects include reproductive/developmental, metabolic/enzyme, and neurological/behavioral effects. The specific effects and the levels at which these effects are seen may vary among the different PCB congeners and mixtures. Recent articles on the ecological effects of PCB exposure are summarized below. These studies focused on reproductive effects in invertebrates, fish, birds, and mammals, immune effects in fish and aquatic mammals, and other effects in plants and fish.

- *Reproductive and Developmental Effects.* A group of related studies investigated the reproductive effects of commercial PCB and combinations of PCB congeners in minks. Two of these studies found that commercial PCBs and combinations of PCB congeners adversely affected reproduction, but that exposure to single PCB congeners did not produce these same effects (Kihlström et al. 1992; Bäcklin and Bergman 1992). One of these studies showed a difference in adverse reproductive effects (e.g., late fetal death versus early fetal death) between two different commercial PCBs, Clophen A50 and Aroclor 1254 (Bäcklin and Bergman 1992). In another study, PCBs were found to reduce egg production and total reproductive capacity in a small crustacean under laboratory conditions (DiPinto et al. 1993). A study that investigated the developmental toxicity of PCB congeners in minnows determined that several congeners caused severe teratogenic effects (e.g., inhibition of yolk absorption) and early hatching (Silberhorn et al. 1992). Recent studies of Great Lakes bird populations (e.g., common terns) provided further evidence of the developmental problems linked to PCBs during the 1980s (Becker et al. 1993; Hoffman et al. 1993; Yamashita et al. 1993). A literature review of wildlife contamination in Chesapeake Bay cited historical evidence of elevated post-hatch mortality in the leopard frog and in unspecified toad species related to PCB exposure (Heinz et al. 1992).

- *Immune System Effects.* In a recent study, production of antibodies was suppressed in juvenile chinook salmon exposed to the commercial PCB mixture, Aroclor 1254 (Arkoosh et al. 1994). In another study, delayed immune system responses in harbor seals fed herring from the Baltic Sea were associated with PCB exposure (Ross et al. 1995). In a study of dolphins, decreased immune system response was associated with elevated levels of DDT and PCBs in their blood (Lahvis et al. 1995).

- *Other Noncancer Effects.* Researchers have correlated detrimental effects on the quality and quantity of growth, as well as increased mortality, in aquatic plants with concentrations of PCBs (Doust et al. 1994). One study of winter flounder did not find a strong association between exposure to PCBs and the development of particular liver lesions (Johnson et al. 1993). Some recent studies have shown that effects from PCB exposure are amplified in the presence of other chemicals. For example, a study in quail found that high levels of cadmium can amplify both the quantitative and qualitative retention of PCB congeners in muscle tissue, especially some of the congeners that are most toxic and resistant to metabolic degradation (Leonzio et al. 1992).

POM. The data in Tables II-9 and II-10 indicate that POM is associated with a range of adverse effects to aquatic and terrestrial wildlife. Studies on the adverse ecological effects from exposure to PAHs (a subset of POM) that have been completed in the last few years are summarized below. These studies examined immune effects in fish and birds as well as various other effects in algae, aquatic invertebrates, fish, and birds.

- *Immune System Effects.* In a recent study, production of antibodies was suppressed in juvenile chinook salmon exposed to PAHs (Arkoosh et al. 1994). Another study found that PAH exposure suppressed the immune system in both adult and young starlings, although only young starling showed overt signs of general toxicity (e.g., decreased body weight and blood hemoglobin concentration) (Trust et al. 1994). A study of effects on fish from PAH exposure found that macrophage activity, an important component of the cellular immune system (i.e., protects the host by eliminating foreign material), was markedly reduced (Huggett et al. 1992). The study results suggested that these effects may be reversible.

- *Other Noncancer Effects.* Recent research has investigated the increased toxicity of PAHs following exposure of the pollutants to solar radiation. One study that investigated the photoinduced toxicity of anthracene (a PAH compound) to a species of green algae showed that the combination of UV-A radiation and anthracene produced significant toxic effects. The study also concluded that algae may be slightly more resistant to photo-induced toxicity than fishes and invertebrates (Gala and Giesy 1992). Two recent studies on fish suggest a strong association between exposure to PAHs and the development of internal lesions, commonly found in the liver and kidney, as well as external lesions, such as lens cataracts (Huggett et al. 1992; Johnson et al. 1993). One of these studies found a strong correlation between sediment PAH concentration and stimulation of enzyme activity, which indicates that the PAHs are a main component of the adverse effects observed in the fish (Huggett et al. 1992). In another study, dramatic declines in PAH concentrations in sediment following the closure of a coking plant were followed by decreased incidences of liver cancer and liver lesions in brown bullhead catfish (Baumann and Harshbarger 1995). A study of aquatic insect larvae from a contaminated area of the Niagara River found that higher body burdens of PAHs were associated with menta ("teeth") deformities. The researchers determined that the deformities were not passed on to future generations (i.e., PAHs caused malformations but did not permanently alter genes or chromosomes) (Dickman et al. 1992).

PESTICIDES

Information on the adverse effects of the pesticides of concern in the Great Waters (chlordane, dieldrin, α-HCH, DDT/DDE, HCB, lindane, and toxaphene) is summarized in Tables II-9 and II-10. These data suggest that the pesticides of concern may produce a wide range of adverse effects in terrestrial and aquatic wildlife of the Great Waters. Recent literature on the ecological effects of pesticides was available mainly for DDT/DDE, HCB, and lindane; this information is summarized below.

DDT. Much of the recent literature on the ecological effects of DDT (and DDE) focuses on reproductive impairment in birds. Other studies demonstrate effects to the immune systems of mammals and impaired growth and survival of aquatic plants following DDT exposure.

- *Reproductive and Developmental Effects.* Recent studies on natural bird colonies in the Great Lakes indicate a decreased role of DDT/DDE in the reproductive success of these populations (Weseloh et al. 1994; Weseloh and Ewins 1994; Bowerman et al. 1995). Weseloh et al. (1994) determined that for herring gull colonies in Lake Superior, eggshells were only eight percent thinner than before the introduction of DDT. In this study, reproductive failure was attributed to causes other than toxic contamination (e.g., predation and shortage of food supply). Similarly, Weseloh and Ewins (1994) attributed increasing populations of double-crested cormorants in Lake Ontario to reduced levels of contaminants (especially DDT/DDE) and increased availability of forage fish. A study investigating the combined effects of DDE and food stress on reproduction in ringed turtle doves found that DDE interacts with food stress to depress breeding success by limiting levels of hormones necessary to develop and maintain active gonads, adequate courtship and brooding behavior, and functional crop glands (Keith and Mitchell 1993). A study of freshwater pond snails, which are an important component of invertebrate fauna in most eutrophic and mesotrophic lakes, found that DDT exposure reduced their reproductive output (Woin and Brönmark 1992).

- *Immune System Effects.* In a study of dolphins, decreased immune system response was associated with elevated levels of DDT and PCBs in their blood (Lahvis et al. 1995).

- *Other Noncancer Effects.* In one study, exposure to DDE caused mortality and had detrimental effects on the quality and quantity of growth in aquatic plants (Doust et al. 1994).

HCB. Recent studies on ecological effects from HCB exposure demonstrate reproductive and neurological effects in aquatic plants and animals. Impaired growth and survival of aquatic species and liver damage in fish and birds have also been shown.

- *Reproductive and Developmental Effects.* Reproductive effects from HCB exposure documented in recent literature include reduced reproduction in protozoa and fathead minnows and reduced fertility in waterflea crustaceans (Constable and Orr 1994; Government of Canada 1994).

- *Neurological/Behavioral Effects.* In freshwater snails, exposure to HCB inhibited body growth, altered metabolic activity, and stimulated egg production, which researchers attributed to neurotoxic effects (Baturo et al. 1995). Kestrels, a small falcon species, have shown ruffling of feathers and tremors following exposure to HCB (Government of Canada 1994).

- *Other Noncancer Effects.* In aquatic species, HCB was lethal to some marine invertebrates and significantly reduced the survival rates of freshwater caddisfly larvae and fathead minnows (Constable and Orr 1994; Government of Canada 1994; Schulz and Liess 1995). In another study, exposure to HCB caused mortality and had detrimental effects on the quality and quantity of growth in aquatic plants (Doust et al. 1994). One report documented the following other effects in aquatic species related to HCB exposure: reduced production of chlorophyll, dry matter, carbohydrate and nitrogen in some algae; decreased growth of algae and protozoa; digestive gland damage in crayfish; and liver necrosis in largemouth bass (Government of Canada

1994). In birds, HCB exposure induced porphyria, increased liver weight, and slightly damaged livers in Japanese quail and produced significant weight loss, increased liver weight, and decreased heart rate in kestrels (Government of Canada 1994).

Lindane. Recent studies show effects in aquatic species following exposure to lindane, including reproductive effects in aquatic invertebrates, immune effects in fish, and behavioral effects in aquatic invertebrates.

- *Reproductive and Developmental Effects.* One study documented reduced reproduction in waterflea crustaceans from lindane exposure (Ferrando et al. 1995).
- *Immune System Effects.* Laboratory experiments on rainbow trout found that antibody production was significantly suppressed from lindane exposure at doses comparable to those found in fish in polluted fresh waters, and as the dose increased the more the immune system was suppressed (Dunier and Siwicki 1994; Dunier et al. 1994).
- *Neurological/Behavioral Effects.* As a result of brief exposures to high concentrations of lindane, the mating behavior of freshwater crustaceans was disrupted (Malbouisson et al. 1994). In another study, the common mussel, *Mytilus edulis*, showed lower rates of feeding-type behavior after exposure to lindane-contaminated sediments (Hermsen et al. 1994). Also, an annelid worm exposed to lindane in seawater exhibited delays in settlement and metamorphosis; this species is otherwise known to be a colonizer of disturbed or polluted areas (Hill and Nelson 1992).
- *Other Noncancer Effects.* In two different studies, survival of a waterflea crustacean species was inhibited by lindane exposure (Tidou et al. 1992; Ferrando et al. 1995); in one of these studies, lindane also significantly repressed growth in the waterflea crustaceans (Ferrando et al. 1995). In studies comparing data for several species of fish, acute toxicity of lindane decreased with increasing total body fat in fish (Geyer et al. 1993; Geyer et al. 1994).

NITROGEN COMPOUNDS

The availability of biologically-usable nitrogen normally limits biological productivity in coastal waters, but an over-abundance of nitrogen is a concern in areas where nutrient enrichment problems, known as eutrophication, have developed. In addition to increasing productivity, nutrient enrichment generally alters the normal ratios of nitrogen to phosphorus and to other elements, such as silicon. This alteration may induce changes to phytoplankton community structure. Species that normally occur in low abundance may be favored, and in some cases, toxic and/or noxious algal blooms may result. On the New England coast, for example, the number of red and brown tides and shellfish problems from nuisance and toxic plankton blooms have increased over the past two decades. In coastal areas with poor or stratified circulation patterns (e.g., Chesapeake Bay, Long Island Sound), the "overproduced" algae tends to sink to the bottom and decay, using all (anoxia) or most (hypoxia) of the available oxygen in the process, killing or driving away organisms that require oxygen. In addition, the increase in suspended matter due to overproduction of algae decreases light infiltration, causing a loss of underwater seagrass and coral communities.

Atmospheric deposition of nitrogen compounds is recognized by all U.S. estuarine programs on the east coast as either a significant contributor to estuarine eutrophication or a mechanism of possible concern (ECARA 1996). A important consideration for controlling atmospheric deposition of nitrogen is that the region from which the atmospheric nitrogen pollution arises is much larger than the water surface that is potentially affected, and even much larger that the watershed that drains into the waterbody. The extent of "airsheds" is now starting to be recognized (e.g., see the discussion of the Chesapeake Bay airshed in Chapter IV).

II.E Other Effects

In addition to adverse effects on human and ecological health, atmospheric deposition of the pollutants of concern may contribute to other actual and potential adverse effects including environmental justice concerns (e.g., effects on subsistence fishers), commercial and recreational fishing losses, and other recreational losses.

Environmental Justice Concerns

As introduced in the First Report to Congress, an important environmental justice issue in the Great Waters is whether certain groups of people may have higher exposures to the pollutants of concern than the general population, and therefore greater risks for adverse health effects (see Table II-11 below).

Subpopulations with Higher Lifetime Cancer Risks

In developing the Great Lakes Water Quality Guidance, EPA *estimated lifetime cancer risks for* certain subpopulations in the Great Lakes due to consumption of contaminated fish. These lifetime cancer risks are compared to EPA's "acceptable" range of lifetime cancer risk for human health in Table II-1. As shown in the table, the risks in the subpopulations are well above the accepted risk range.

TABLE II-11
Lifetime Cancer Risks in Various Great Lakes Subpopulations Versus EPA's Appropriate Range of Risk to Human Health

Subpopulations	Lake Superior	Lake Michigan
Native Americans	18 in 10,000	370 in 10,000
Low income minority sport anglers	25 in 10,000	120 in 10,000
Other sport anglers	9.7 in 10,000	450 in 10,000
Acceptable Range of Lifetime Cancer Risk	0.01 in 10,000 to 1 in 10,000	

Source: U.S. EPA 1995a.

Some populations are at higher risk because they are more vulnerable to effects (e.g., nursing women and their children). Other populations may be more highly exposed because they consume larger quantities of Great Waters fish than the general population (e.g., for subsistence reasons); these populations include Native Americans, the urban poor, and sport anglers. Recent studies have investigated the relationship between certain populations and exposure through fish consumption:

- Members of the Mille Lacs Band of Chippewa Indians in Wisconsin catch 90 percent of the fish they eat (U.S. EPA 1992). Given this high rate of consumption, the potential for exposure to pollutants is much greater in these Native Americans than the general population.

- In a study of Michigan licensed sport anglers, researchers found a relationship between fish consumption levels and socioeconomic characteristics. A combination of minority status and relatively low annual income (less than $25,000) was correlated with higher levels of fish consumption (West et al. 1993).

Other studies have established a link between people with higher fish consumption rates and levels of contaminants in their blood:

- High levels of dioxins and furans were found in frequent consumers of fish living near the Baltic Sea in Sweden. Individuals with the highest percentage of fish in their diet, specifically fishermen and fishing industry workers, had blood levels that were approximately three times that of non-fish consumers (Svensson 1991).
- On the north shore of the Gulf of the St. Lawrence River, fishermen who consumed large amounts of seafood had elevated levels of PCBs in the blood. The blood levels among the highly exposed fisherman were 20 times higher than controls (Dewailly et al. 1994).

As indicated in Section II.A, fish consumption advisories are designed to take into account the fact that some populations are potentially at a greater risk than the general population. Recent studies indicate that some high-risk populations are changing their fish consumption and preparation habits in response to fish consumption advisories. A survey of 8,000 sport anglers in the U.S. Great Lakes states found that 36 percent of the respondents had changed their fish consumption behaviors, including modifying fish cleaning and preparation methods and eating less Great Lakes fish (Connelly and Knuth 1993). Another study found that pregnant women of the Mohawk nation had substantially reduced their fish consumption (Fitzgerald et al. 1993).

An on-going Canadian study established as a First Nations/Health Canada partnership is assessing the extent of exposure of Native people living in the Great Lakes basin to bioaccumulating pollutants and the associated risk to their health and well-being. The project, Effects on Aboriginals from the Great Lakes Environment (EAGLE), began in September 1990 and is expected to be completed in 1997. EAGLE is a community-based epidemiological project involving the approximately 100,000 First Nations people living in 63 aboriginal communities in the Great Lakes Basin. The project builds on earlier studies and is examining exposure in both adults and children, socioeconomic effects, and the impacts on traditional ways of life, culture, and values (Manno et al. 1995).

In addition to individuals that fish and hunt for subsistence reasons, hunting, fishing, and gathering also are an integral part of the cultural and spiritual practices in many U.S. subpopulations. Most of the limited information available on these subpopulations focuses on Native American communities. For example, for centuries, some Native American tribes in Wisconsin have built cultural traditions around spearing fish and sharing the catch. These traditions are negatively impacted when limiting fish consumption is advised and certain fishing locations are banned (U.S. EPA 1992).

Commercial and Recreational Fishing Losses

Bans and advisories on commercial fishing due to pollution can cause economic and social losses to owners and employees of commercial fishing enterprises. In addition, bans and advisories on recreational fishing have potentially far-reaching effects on the U.S. economy; expenditures on recreational fishing stimulate the economy, provide jobs in the industry, and generate state and federal taxes (Fedler and Nickum 1992). In 1991, more than 35.6 million Americans over 16 years of age spent a total of $24 billion on sport fishing, averaging 13 trips each and 14 days a year (U.S. FWS 1993). As shown in the sidebar, losses due to fish consumption advisories can be significant. Recreational fishing bans and advisories also can affect fishermen's health (e.g., from eating fish they catch despite fish consumption advisories) and quality of life (e.g., cannot fish due to fishing bans).

> The Arkansas Game and Fish Commission has estimated a loss of fishing expenditures due to mercury fish consumption advisories of over $5 million dollars from 1991 to 1992.
>
> Source: Armstrong 1994.

Other Recreational Losses

In addition to recreational fishing, recreational hunting (e.g, for food or fur) may be negatively impacted by exposure of wildlife to the pollutants of concern. These impacts include decreases in the populations of hunted species (e.g., stemming from adverse reproductive effects) and increased health risks to the hunters who consume their kill. Poor water quality (and/or public perception of poor water quality) may negatively affect recreational uses other than fishing and hunting, such as boating, swimming, and visiting parks. Decreased recreational use also may affect the related tourism economies; the annual outdoor recreation/wildlife industry in Great Lakes states generates approximately $12 billion (NWF 1993). Another non-consumptive recreational use that could be negatively impacted by the effects of exposure to the pollutants of concern is the decreased ability to observe and photograph wildlife.

II.F Exposure and Effects Summary

This chapter describes current information available on exposure and effects of pollutants of concern, and the potential impact these pollutants may have on the Great Waters. Recent studies continue to show that persistent pollutants are found in air, water, sediments, and biota, that air transport and deposition are significant contributors to this load (discussed in subsequent chapters in this report), and that a strong association is found between the exposure to Great Waters pollutants of concern (often mixtures) and deleterious effects.

Exposure to pollutants of concern by humans or wildlife can occur directly (e.g., through intake of drinking water, direct contact with water) or as a result of food web contamination. As summarized in Sections II.A and II.B, the exposure of humans and animals to Great Waters pollutants of concern is measured by various indicators such as reports of exceedances of water quality criteria and the issuance of fish consumption advisories. The water column of the Great Lakes in recent years has been found to contain levels of PCBs and dieldrin that exceed the most stringent water quality criteria for these Lakes (Table II-2). Also, PCBs are most commonly associated with fish advisories issued in the Great Waters, with dioxins having the next highest occurrence of fish advisories (see Tables II-4 and II-5 and Appendix B). Elevated levels of other pollutants, such as chlordane and mercury, in fish also have warranted fish advisories in many Great Waters. These water quality criteria exceedances and fish advisories suggest, as in the First Report to Congress, that toxic contamination continues to be present in the Great Waters.

Measurements of pollutant levels in biota, such as game fish, provide useful information about the extent of contamination in a waterbody, as well as the potential bioaccumulation in the food web. The Great Waters program relies on information collected from national monitoring programs designed to assess ecological quality of, and extent of contamination to, the major U.S. waterbodies. Most of these programs, however, do not determine the potential source(s) of the toxic contamination and the relative contribution of atmospheric deposition to the site. These large-scale programs, as well as research conducted at the specific waterbodies, provide useful data on pollutant levels in biota, sediment, and water. For example, in the Great Lakes, researchers from the United States and Canada have provided collected information on current trends of several pollutants of concern in the aquatic ecosystems and the extent of bioaccumulation and biomagnification of these pollutants.

In Sections II.C and II.D, potential human health and ecological effects of the Great Waters pollutants of concern are summarized (see Tables II-7, II-9, and II-10). The pollutants act on many target organs, including the liver, kidney, nervous system, reproductive organs, and immunological system. As in the First Report to Congress, the contribution of atmospheric deposition to potential human health and ecological effects associated with exposure to the pollutants cannot be quantified at this time. The information presented in this report on the effects associated with each of the pollutant groups, in general, describes effects that may occur in humans or wildlife with long-term (i.e., chronic) exposure to pollutants of concern, which may differ from effects caused by acute, high-level exposures (e.g., accidental spills). In addition, the information presented in this report covers only recent studies, and therefore, cannot be used alone to determine whether these effects exhibit trends or are widespread in the environment.

Overall, the recent information on the effects of the pollutants of concern to the Great Waters continue to support, and expand on, the findings from the First Report. Recent developments in human health effects research include an increased awareness of endocrine-disrupting effects of pollutants which act by interfering with hormone action in the body. There also has been continuing research on mercury compounds, particularly regarding the effect of methylmercury on neurological development in children. The recently-released dioxin reassessment document by EPA provides comprehensive data on the various effects of TCDD/TCDF and their potential mechanism of action.

As with human health effects, endocrine disruptors have been an emerging issue of interest regarding ecological effects of the Great Waters pollutants of concern. The potential mechanisms of action of these pollutants are a major focus for researchers, as well as linking observed effects to hormone disruption in wildlife. In addition, data and models are currently available for analyzing TCDD exposure and effects in aquatic and terrestrial wildlife.

Besides direct human and ecological effects, the pollutants of concern contribute to other adverse impacts such as the increased vulnerability to health effects in susceptible populations (Section II.E). For example, Native Americans and sport anglers (especially poor, minority anglers) have the potential for higher lifetime cancer risk because historically their diets consist of higher amounts of contaminated fish. To protect humans from the adverse effects of these pollutants, restrictions or limits are placed on fishing and recreational activities, fish consumption, and drinking water contaminant levels. However, these restrictions raise environmental justice concerns, because they may disproportionally affect the welfare or lifestyle of certain individuals. For example, fishing provides a free source of nutritious food for poor populations; if fishing and fish consumption are restricted (and these restrictions are followed), these populations have lost an important part of their diet.

CHAPTER III
ATMOSPHERIC TRANSPORT AND DEPOSITION PROCESSES

Under section 112(m) of the CAA, EPA is directed to, among other things, perform activities such as monitoring atmospheric deposition, investigating sources and deposition rates, and conducting research to improve monitoring methods. Accomplishing these tasks requires an understanding of the processes by which the pollutants of concern are transported and deposited to the Great Waters. Understanding these processes also is necessary for analyzing the relationships between source emissions, relative loadings, and the potential for adverse effects in humans and the environment. Because it is often difficult to establish these relationships clearly and quantitatively through available measurement data (e.g., it can be difficult to differentiate between the contribution of distant versus local sources to the loading of a pollutant to a particular waterbody), researchers frequently use mathematical models of atmospheric transport and deposition. These models are based on processes characterized with intensive data gathering and take into account variables such as emission rates, wind direction and speed, pollutant concentration, and removal rates of a pollutant from the atmosphere.

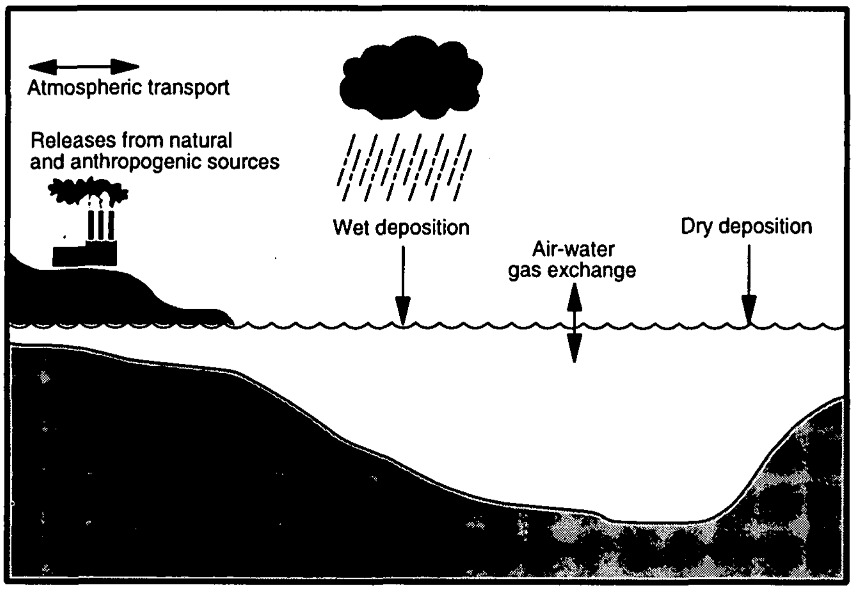
This chapter describes the basic processes of atmospheric transport and deposition and some of the recent measurement programs and mathematical models that are being used to estimate the rates of these processes and quantities of pollutants involved. It focuses on the achievements of the most recent four years and on efforts by EPA and NOAA plus other researchers, mostly in academia. Further discussion of the uses of some of these models and process understanding are included in Chapter IV.

III.A Understanding of Atmospheric Transport and Deposition

Long-range atmospheric transport and deposition of pollutants have been widely acknowledged to make a significant contribution to contaminant inputs to surface waters, including the Great Waters (e.g., Rappaport et al. 1985; Bidleman et al. 1988; Poster and Baker 1996). Atmospheric deposition refers to the removal of pollutants (following transport) from the air to soil, water, and other surfaces. Deposition may occur directly to the water surface and/or indirectly to the land surface in the watershed, with subsequent runoff from rainfall carrying contaminants to the waterbody. It is important to recognize that both local and distant air emission sources may contribute to the pollutant loading at a given location and time. Also, pollutants may be emitted from a combination of point, mobile, or area sources; or even from re-suspension in the air of previously deposited material.

There are three major processes of atmospheric deposition to natural waterbodies: wet deposition, dry deposition, and gas exchange across the air-water interface. A schematic of these processes is presented in Figure III-1. Monitoring and assessment of these processes provide important information on the atmospheric contribution of a given pollutant to a specific site, as well as extent of deposition of the pollutant. In addition, semi-volatile compounds (e.g., PCBs, PAHs) are transported in the air in both gaseous and particulate phase, and persistent compounds can cycle among the environmental media (e.g., air, water, sediment) for long periods of time. Measurements of these processes allow researchers to evaluate quantitatively the long-term distribution of a pollutant in an aquatic system. Hence monitoring at Great Waters study sites requires meteorological measurements very near to the site and if possible over the surface of the waterbody itself. The Great Waters program is

FIGURE III-1
Atmospheric Deposition Processes



one sponsor of research on atmospheric deposition; this research is needed so that future monitoring can become less research-specific and more routine. Monitoring in the future will be needed to evaluate trends in atmospheric loading of pollutants and to determine the effects resulting from efforts to reduce emissions.

Wet Deposition

Wet deposition refers to the incorporation of both gases and particles into cloud droplets and into "hydrometeors" (e.g., snow, sleet, and rain) where they are carried to the Earth's surface in precipitation. Pollutants may be removed from air by wet deposition through three main mechanisms: (1) minute pollutant-bearing particles can serve as cloud condensation nuclei and become naturally embedded into water droplets during cloud formation (e.g., Poster and Baker 1996); (2) particles may be incorporated into falling hydrometeors by collisions; or (3) gaseous pollutants may be dissolved into cloud droplets and falling rain. Cloud and precipitation droplets, therefore, may contain pollutants in both particulate and dissolved (gaseous) forms. The removal of gases by wet deposition is dependent on their solubilities in the elements of precipitation. Wet deposition is essentially an intermittent process, although if precipitation events are frequent or long-lasting, wet deposition may be the major pathway for deposition of air pollutants.

To get useful and quantitative measurements of wet deposition, well-prepared protocols and quality assurance approaches are designed and used, with careful handling of field equipment, samples, and subsequent chemical analyses. For example, wet deposition rates are determined using collectors that are designed to open only during precipitation events. Work must be designed and carried out so as to minimize measurement problems with the samples that can arise from chemical or biological activity, evaporation of gaseous pollutants, or possible contamination during storage and handling. The concentrations of most of the pollutants of concern in any one precipitation event are small, so rather advanced field and chemical-analytical methods are needed for quantitative work. The persistence and tendency to bioaccumulate of the pollutants of concern make them important over time, though measurements must be taken of short-term events. Thus, the kind of monitoring used in the Great Waters program cannot be considered routine; rather, it involves research into new and more accurate monitoring techniques and must be carefully designed to focus efforts on well-located sites, protocols, and methods.

Dry Deposition

Dry deposition refers to removal from the air of pollutants (bound to particles or in the gaseous form) to the land or water surface in the absence of precipitation. Dry deposition is essentially a continuous process and often represents a substantial removal of the pollutants from the atmosphere, and their deposition to surfaces. The pollutants reach the surface by the turbulent movements of the air or through gravitational sedimentation (i.e., downward settling of particles due to Earth's gravity force). Dry deposition rates are affected by factors such as wind conditions, relative humidity, temperature, surface characteristics, and range of sizes of the particles bearing the pollutants.

Difficulties in measuring dry deposition of a pollutant are due to the typically low concentrations of atmospheric pollutants found in remote areas of large lakes or watersheds, interferences from upward movements of pollutants, and varying micrometeorological conditions. Other complicating factors include the need to account for the nature of the various receiving surfaces (soil, vegetation, or water, and even factors such as water surface roughness caused by waves). Consequently, in an effort to clarify some of these issues, researches on measurement methodologies and modeling of dry deposition are being pursued under the Great Waters program (see sidebar).

Research on Dry Deposition

In 1994, a research program was established between the EPA, the University of Michigan, the Illinois Institute of Technology, Carnegie-Mellon University, and Oak Ridge National Laboratory to develop and test new techniques for the direct measurement of dry deposition of mercury and other trace elements to natural water surfaces. The data will be used to develop new mathematical models for predicting dry deposition onto water surfaces under a variety of atmospheric and surface conditions. Work has begun; measurement methodologies have been developed and tested, and modeling is underway. Field studies in 1996-1997 are planned to gather basic data, and to validate the models.

Gas Exchange Across the Air-Water Interface

In addition to wet and dry deposition of pollutants, gaseous pollutants may be directly exchanged between air and water (i.e., transferred in either direction across the air-water interface). The gaseous exchange of organic compounds at the air-water interface is an important phenomenon in the balance of pollutants occurring in air and water (Eisenreich and Hornbuckle 1996), and extensive North American waterbodies such as the Great Lakes, Chesapeake Bay, and the Gulf of Mexico, provide large surface areas for this exchange to occur.

Gas exchange is a two-way process involving both *gas absorption* or invasion (air to water) and *volatilization* or evasion (water to air) across an air-water interface. When the concentration of a volatile chemical (usually in gaseous form under every-day conditions) or semi-volatile chemical (e.g., PAHs, PCBs) in water exceeds the concentration in air, the chemical tends to approach equilibrium by moving from the water to the air through volatilization (i.e., going from a liquid to a gaseous state). When the concentration in air exceeds the concentration in water, gas absorption tends to produce net movement of the chemical from air to water. There is a natural tendency for pollutants to reach near-equilibrium between their concentrations in water and their concentrations in air. The direction and magnitude of gas transfer is a function of the chemical concentrations in air and water, wind speed, temperatures in air and water, characteristics of the water (e.g., chemistry, surface films), and the physical and chemical properties of the pollutant (e.g., molecular weight, vapor pressure, and solubility).

Gas absorption and volatilization occur simultaneously, even when near-equilibrium has been achieved. Taken together, volatilization and gas absorption contribute to the *net flux* (the difference between the amount of pollutant invading and the amount evading) or effective movement of a chemical across the air-water interface. Net flux may be expressed as the mass of gas moving across a unit of area over a unit of time (e.g., 8 mg per square meter of water surface per day). For Great Waters to achieve quantitative estimates of net deposition of pollutants of concern, many of which are semi-volatile, such physical and chemical processes must be quantified so they may be correctly used in models. Some earlier work on pollutant loading made simplifying assumptions of "one-way" flux or deposition without quantifying gas exchanges, but recent work shows the need for more complete representation of the natural processes for each pollutant.

Models of Gas Exchange at the Air-Water Interface. Two well-established models are used to describe gas exchange at the air-water interface. The Stagnant Two-film Model is used to estimate gas exchange at low wind speeds, which results in an essentially stagnant boundary layer just above the water. As higher wind speeds generate more turbulence in the boundary layers of both air and water, more rapid gas exchange of chemicals occurs. To characterize this stage, the Surface Renewal Model may be used. Under the conditions normally prevalent in the Great Lakes and coastal waters, researchers apply these two models and obtain reasonable results for use in Great Waters studies. A third model exists for highly turbulent conditions of wind and water surface over the open ocean, where gas exchange may be enhanced; but a third model has not been found necessary for Great Waters studies to date (Eisenreich and Hornbuckle 1996).

For nitrogen compounds (e.g., nitrates, ammonia, organic-nitrogen) estimates of overall air-water exchange rates have been developed for open ocean conditions. However, these rates may not apply to coastal waters and estuaries due to differences in meteorological conditions. In coastal areas, the primary obstacle to quantitative estimation of the exchange rate has been the lack of near-surface, over-water meteorological data for model input. To address this need, a network of monitoring buoys has been deployed in the Chesapeake Bay (Valigura 1995; Luke and Valigura 1996). Two objectives of this project are to: (1) develop and evaluate a model based on data from the buoys to estimate air-water exchange of heat, water, and momentum; and (2) use the model outputs to estimate air-water transfer rates of nitric acid, the main chemical form in which nitrogen oxide emissions are deposited. These model runs and results will be important in quantifying the link from emissions of nitrogen oxides (or "NO_x") to the impact of excessive nitrogen loading on Chesapeake Bay waters.

Environmental Cycling of Semi-Volatile Compounds

The exchange of gases between air, water, and soil is especially important for the pollutants of concern that are characterized by chemists as "semi-volatile" in nature. These include organic compounds, such as PAHs, PCBs, and pesticides such as hexachlorobenzene, α-HCH, and lindane. Even mercury and other metals can form semi-volatile compounds. In every-day terms these "semi-volatile" chemicals will evaporate into the air (though not as readily as compounds such as benzene or gasoline). Because these compounds possess very low vapor pressures and water solubilities, they are distributed between the gaseous and particulate phase in the atmosphere and between the dissolved (gaseous) and particulate phase in the water column (Eisenreich and Hornbuckle 1996). Thus, the pollutants of concern that are semi-volatile will have complex ways of being distributed among gas, liquid, and solid phases of matter, which makes tracking or modeling their movements in the environment very complex. Each pollutant has a particular "balance" of these chemical-physical attributes, so quantitative work requires that each pollutant be well understood. The pollutants of concern are also chemically persistent, maintaining chemical identity (not being broken down) even as they move among physical phases and, in many cases, into biological tissues.

Once released to the environment, persistent semi-volatile compounds may repeatedly cycle between the atmosphere, land, and waterbodies. This cycle can extend over long time periods, resulting in transport for long distances. Long-distance transport, with repeated deposition to land or waters and then re-evaporation (re-volatilization) to the atmosphere has been dubbed the "grasshopper effect." The extent and duration of cycling depends on various factors: volatility and persistence of the substance; molecular weight; concentrations and temperatures in air, soil, and water; and atmospheric circulation, pressure, and weather conditions. As the seasons change, the behaviors of atmospheric contaminants change relative to their location in physical media; so sampling work and modeling calculations must be adjusted, for each compound, to correctly estimate their presence and impacts. Warmer conditions, on seasonal and global scales generally favor net movement into the atmosphere. Often redeposition takes place in areas of colder atmospheric temperatures (Fellin et al. 1996).

III.B Atmospheric Transport and Deposition Models

The emission, transport, transformation, and deposition of pollutants to the Great Waters is a large and complex series of processes involving many different pollutants that have very different behaviors in air and water systems, over very large geographic areas. Therefore, numerous models, as well as input parameters for these models, have been developed or evaluated for estimating the atmospheric transport and deposition processes for the various pollutants associated with the Great Waters.

Atmospheric transport and deposition models are used to:

- Predict the direction and distances pollutants will travel in the environment;
- Provide calculated estimates which fill spatial and temporal gaps in monitoring networks, to provide a smoothed or coherent picture for analyses;

- Test hypotheses about characterizations of atmospheric transformations and removal;
- Assist in designing monitoring networks for efficiency and specific analyses, and in placing a limited number of monitors effectively;
- Develop source-receptor relationships; and
- Evaluate prospective control strategies.

This section discusses different types of models related to atmospheric transport and deposition processes. The mass balance models consider the mass (or weight) of a pollutant that is exchanged across interfaces between air, water, land, and sediments as inputs and outputs, to assess the relative loadings of a pollutant into a waterbody by different pathways. Source apportionment techniques provide useful information on the relative contribution of different sources to air pollutant levels, taking into account emission inventories and meteorological data. Dispersion modeling, a source apportionment technique, traced pollutants from sources to the air at various locations (waterbody, land), and quantified relative contributions from a particular point source or groups of sources. This model required detailed information on location and emission characteristics of various sources, which was often lacking. The source apportionment approach is quite simple in its use of meteorology, with no "cells" and thus no calculation of meteorological events within cells. It lacks methods to calculate transformations of chemicals within the atmosphere and lacks algorithms for process of flux and deposition. Its limitations have led to the development of other models such as the receptor and air quality simulation models, as well as hybrid models. These models are more frequently used and are discussed in this section.

Mass Balance Model. Estimates inputs and outputs of a pollutant to a waterbody (i.e., total amounts of a given pollutant that enter and exit a water body by each of the various pathways).

Source Apportionment Technique. Estimates the relative contribution of different sources to air pollutant levels at a specific receptor site (e.g., a particular air mass over Lake Michigan on a particular day).

Mass Balance Models

A mass balance model provides the essential framework for determining the relative contribution of pollutant loadings from various mechanisms of input (e.g., direct discharge, river input, atmospheric deposition) and output (e.g., sedimentation, volatilization, outflow) to and from a waterbody. As introduced in the First Great Waters Report to Congress, when reliable information is available for contributions from the various sources, the mass balance equation may be used to estimate the importance of atmospheric deposition in causing contamination of a waterbody.

Mass balance studies have given insights on atmospheric deposition issues relevant to the Great Waters. These studies have provided quantitative estimates indicating that: (1) atmospheric deposition in some cases is the main contributor of toxic chemical contamination and nitrogen enrichment to Great Waters, although uncertainties still exist; (2) the importance of atmospheric load for a specific pollutant in a given waterbody depends on characteristics of the waterbody, chemical property, and source location; and (3) chemicals may cycle between soil, air, water, and biota for many years.

The First Report presented mass balance case studies for some Great Waters pollutants of concern, such as PCBs in Lake Superior, mercury in lakes in Wisconsin, and nitrogen in several Atlantic states. Considerable research continues on the development and use of mass balance models for the pollutants of concern in the Great Waters. The Lake Michigan Mass Balance Study, an EPA-sponsored research project, is responsible for a comprehensive sampling effort in Lake Michigan, including measurements of pollutants of concern -- PCB congeners, trans-nonachlor (a chlordane metabolite), mercury, and atrazine -- in the atmosphere, tributaries, open lake water, sediments and food chain (fish tissue). Samples collected over a two-year period (1994-1995) for modeling will be used to improve understanding of key environmental processes that govern cycling and bioavailability of contaminants within the Lake Michigan ecosystem. The approach modifies the classic mass balance within a closed system to consider inputs from transport, including long range transport. A data summary is expected in late 1996, with some modeling runs in 1997. A more detailed discussion of this model effort is described in Section IV.A on the Great Lakes.

Receptor Models

The receptor model, a source apportionment technique, traces pollutants in the air at various locations (such as over a waterbody) back to particular source types in order to estimate the contribution to pollutant levels from a group of sources with similar emissions. This type of model bypasses the need for meteorological data or extensive emission inventories, which are used in other types of models (see the discussion below of air quality simulation models). Receptor models assume that chemicals of concern are affected in the same way by all of the processes involved in pollutant transport and dispersion. This assumption is particularly questionable when clouds are present, when precipitation occurs, or when extensive chemical transformations of a pollutant are known to occur. The model is also limited by the lack of adequate "source profile" data that allow air pollution to be linked to a particular source type. Source profiles refer to "signatures" or "fingerprints" of emissions from a type or category of sources; these profiles are determined through samples taken close to the actual emissions (i.e., near the "smokestacks") followed by analyses comparing the chemical signature to those of other categories of sources.

Receptor Model for Great Waters

A project on atmospheric deposition in the Great Waters was started in 1993 by EPA and scientists from the Universities of Minnesota, Michigan, Maryland, Delaware, and the Illinois Institute of Technology. The objectives of this 3-year research program are to determine: (1) the wet and dry depositional fluxes of critical urban contaminants to northern Chesapeake Bay off Baltimore and southern Lake Michigan off Chicago; (2) the contributions of urban source categories to measured atmospheric concentrations and deposition; and (3) the air-water exchange of contaminants and their partitioning into aquatic phases. The contaminants being studied include mercury, trace metals, PCBs, and PAHs. Work continues, though slowed by funding constraints in 1996. Techniques involve using all three modeling approaches -- CMB, PCA, and trajectory analyses.

Some receptor model types include:

- *Chemical Mass Balance (CMB)*: The CMB model assumes that emission characteristics (i.e., chemical and element composition, physical size, morphology) of various source types are sufficiently different from one another that their contributions to a receptor may be identified by measuring the characteristics in samples collected at the site. The observed concentration pattern of an ambient sample is equated to a linear combination of the source patterns, each weighted by an unknown source strength term. The primary application of the CMB model has been on urban areas such as Chicago, or Baltimore. CMB models assume composition of all contributing sources are known, and when this is not the case the uses of the model are limited.

- *Principal Component Analysis (PCA)*: This model incorporates quantitative monitoring at the receptor site (e.g., the shore of a large waterbody) of measurements such as wind speed, wind direction, and relative humidity, along with concentrations of the various pollutants. The objective of PCA is to use mathematical analyses to find a minimum number of factors, or source categories, that explain most of the variance in a system. The number of statistically significant factors is usually found to be six or less. PCA is limited because it lacks fine resolution of contribution from various distant sources.

- *Trajectory clustering*: A back trajectory is computed from upper air wind data to represent the spatial probability of an air "parcel" reaching a particular receptor site at a particular time. Grouping trajectories and concurrent pollutant measurements into common spatial clusters is a method used to classify different meteorological situations associated with pollutant transport from different sources and source regions. This approach is being used to assess mercury deposition in the Great Lakes basin (see sidebar).

Trajectory Clustering Technique in the Great Lakes

To determine the sources of mercury deposition to the Great Lakes Basin, a regional network of 10 monitoring sites was established in 1993 by EPA and the University of Michigan to measure atmospheric mercury over a two-year period. The sampling will continue into 1997. Data will be analyzed, using an improved trajectory clustering technique, to determine the sources and source areas most responsible for mercury deposition to the Great Lakes.

Air Quality Simulation Models

Air quality simulation models (AQSMs) are frequently used to characterize the emission, transport, and deposition of hazardous air pollutants over large geographic areas. These models incorporate fairly extensive source emission inventories and meteorological data bases (e.g., wind fields, temperature, mixing height), and apply the collected data to simulated processes such as dispersion, transformation, and deposition. The models are run to generate estimates of pollutant concentrations and deposition rates over a spatial and temporal pattern. The AQSMs are based on two approaches. One approach is based on a model in which characteristics or properties of air are assigned to fixed points in space at a given time. The second approach is based on a two- or three-dimensional grid system for the geographical pattern of interest, and all the fundamental processes (e.g., emissions, chemical and physical transformations, deposition) of discrete pollutant parcels are considered to occur within the individual grids or boxes.

The mathematical relationships between emissions and concentration (or deposition) are typically nonlinear, due to the influences of the atmospheric transport, chemical and physical transformation, and deposition processes. Therefore, one cannot extrapolate, based on measurements alone, the quantitative relationship between changes in emissions and changes in atmospheric concentrations (or deposition). AQSMs attempt to account for the nonlinear physical and chemical processes influencing atmospheric concentrations and deposition. AQSMs also are useful tools in providing the analytical framework required to predict the environmental impacts of proposed emission control programs, and consequently, performing both scientific and regulatory assessments.

An air quality simulation model that has been applied to the Great Waters is the Regional Lagrangian Model of Air Pollution (RELMAP). To estimate atmospheric mercury concentration and wet and dry deposition patterns, the RELMAP has been used to simulate emission, transport and diffusion, chemical transformation, and deposition of mercury compounds (gaseous and particulate forms) (see Table III-1). The goal of this modeling was to determine the extent of mercury emissions to air in the United States over an entire year, the deposition to U.S. soil and waterbodies, and the source category contribution to total amount of mercury emitted and deposited within the United States (Bullock et al. 1996). The simulated results were found to correspond well with the majority of limited annual deposition and concentration data around the Great Lakes and Florida.

The Regional Acid Deposition Model (RADM), first developed under the National Acid Precipitation Assessment Program (NAPAP) to address policy and technical issues associated with acidic deposition, has been applied to the Chesapeake Bay watershed to study nitrogen deposition (Dennis 1996). This air quality simulation model has three steps of analysis: (1) assess the range of influence of nitrogen oxide emissions with respect to nitrogen deposition; (2) estimate or define the airshed affecting the Chesapeake Bay watershed; and (3) delineate the contribution to nitrogen deposition by major nitrogen oxide emitting sectors. A range of complex issues to consider in modeling acidic deposition include emissions of chemicals, meteorological processes (clouds), physical and chemical transformations of emitted pollutants, and meteorological factors and properties of Earth's surface affecting deposition. The information gathered from this modeling effort provides useful information for the Great Waters program regarding nitrogen deposition in the Chesapeake Bay watershed, as well as the sources responsible in atmospheric contribution to the Chesapeake Bay region (see Table III-1). Results from the RADM are presented in Section IV.C on Chesapeake Bay.

AQSMs are limited by the quality of their process-based algorithms (e.g., transformation, deposition) and their input data (e.g., emissions, meteorology). Typically, the quantity and quality of available input data is the greatest limiting factor in the application of AQSMs. This is especially true for atmospheric pollutants that have relatively poorly developed emission inventories, or for which re-emission and environmental cycling are significant. Efforts sponsored by the Great Waters program are underway to improve the quality of emission estimates for the hazardous air pollutants (HAPs), which historically have been inventoried only in a few places and for short time intervals. The eight states that border the Great Lakes have worked together, with EPA, to develop an approved protocol for a coordinated emission inventory of 57 HAPs, including the Great Waters pollutants of concern other than pesticides. The inventories for point sources and area sources should be completed for most of the states by the end of 1996, with mobile source emissions to be pursued in 1997. The data are stored in a regional data base system (RAPIDS) developed for this project. Public access to the data is expected in 1997. EPA's national air office has developed other data bases that assemble emissions data on HAPs from several studies.

Hybrid Models

In addition to the above types of models, hybrid models are similar to receptor models, but also incorporate meteorological data, and work from both the source "end" and the receptor "end" of the air movement situation. For example, dry deposition of trace metals and semi-volatile compounds to Lake Michigan is being estimated by a hybrid receptor-deposition modeling approach (see Table III-1), as part of the Lake Michigan Urban Air Toxics Study (LMUATS) in 1991 (Keeler 1994; Pirrone and Keeler 1996).

Comparing Models Used in Great Waters Studies

Several numerical atmospheric transport and deposition models or modeling strategies are currently being developed and used for understanding deposition of pollutants to the Great Waters. To discuss these models and others and their applications to Great Waters with the general scientific community, a session was set up at a national scientific meeting. At the 15th annual meeting of the Society of Environmental Toxicology and Chemistry (SETAC) in 1994, a session on "Atmospheric Deposition of Nutrients & Contaminants" was held, co-sponsored by EPA Great Waters program. Following that meeting, the presenters and other scientists prepared written chapters for a SETAC Special Publication: *Atmospheric Deposition of Contaminants to The Great Lakes and Coastal Waters*, J.Baker, editor (Baker 1996). That SETAC special publication (being published by the society) provides an in-depth technical reference that supplements this Report. Table III-1 summarizes some of the specific models of interest to Great Waters, with a very brief description of the model, its application to Great Waters studies, and how the model compares with actual monitoring data; further information is presented in Baker (1996). For each model, it is important to note that either assumptions have been made about relevant physical or chemical processes (Bullock et al. 1996; Ching et al. 1996; Pirrone and Keeler 1996; Seigneur and Constantinou 1996) or an established model is being used in a novel approach, such as the use of the RADM to determine nitrogen deposition in the Chesapeake Bay watershed (Dennis 1996). Models have many roles in EPA's atmospheric programs, and are widely used to tie together emissions data, meteorology, receptor sites (people, or lakes, exposed to pollutants), and monitoring of the ambient air (in cities, or over lakes for Great Waters.) New approaches to modeling have been needed to deal with the particular complexities of the questions in Great Waters studies.

A brief discussion of Table III-1 will explain some of the contributions made by modelers, as well as some of the limits to each approach. The first modeling effort listed in the table is the regional-scale source attribution of nitrogen deposition to the Chesapeake Bay, by Dennis (1995). RADM, which has been well tested for source-receptor modeling of sulfur compounds, but was not tested for nitrogen compounds. Since atmospheric nitrogen oxide (NO_x) is known to be involved in reversible reactions in the nitrogen photochemical cycle, the source attribution technique developed and tested for the simpler sulfur cycle may not be valid. To resolve the problem, a special technique was used: to define the deposition range for NO_x from a particular source sub-region, a simple fractional reduction of emissions from the sub-region was prepared, followed by application of the RADM for the full suite of 30 aggregation cases to obtain a field of annual deposition. This technique was used to determine the fraction of nitrogen deposition to the Chesapeake Bay watershed that was attributable to each of nine sub-regions in and around the watershed.

Two of the modeling efforts described in the table are focused on the issue of atmospheric mercury simulation. The numerical modeling of atmospheric mercury by

TABLE III-1
Summary of Several Atmospheric Transport and Deposition Models

Model	Description of Model	Great Waters Related Application	Goodness of Fit[a]	References
Regional Acid Deposition Model (RADM)	Developed under the NAPAP to predict regional changes that may occur as a result of nitrogen and sulfur deposition. The geographic area covered by the model is the eastern U.S. and Canada.	Determines the fraction of nitrogen deposition to the Chesapeake Bay watershed attributable to each of the sub-regions in the area.	Not given; however, emission estimates are considered to be conservative.	Dennis (1996)
Regional Lagrangian Model of Air Pollution (RELMAP)	Simulates concentrations of wet and dry deposition patterns of gaseous pollutants and particulate matter (both fine and coarse), and can generate source-receptor matrices for user-defined regions.	Modeled deposition of metals including cadmium and lead to Lake Superior; modeled the emission, transport, and fate of airborne mercury in the U.S., including the Great Lakes and Florida.	Wet deposition results from RELMAP for atmospheric mercury agree with actual measurements within a factor of 2.	Bullock et al. (1996); Eder et al. (1986)
Atmospheric Mercury Deposition Model	Incorporates ranges of deposition velocities and scavenging coefficients from literature with estimates of turbulent mass transfer at the surface layer (i.e., dry deposition) and hourly precipitation rates (i.e., wet deposition).	Estimates wet and dry deposition of mercury to the Great Waters.	Theoretical only and has not been compared with actual data.	Seigneur and Constantinou (1996)
Regional Particulate Model	Based on RADM and further computes the chemical composition and size distribution of the secondary sulfur and nitrogen species, to identify airborne particles that may serve as sites for condensation or volatilization.	Predict wet and dry deposition of airborne semi-volatile organic toxic compounds to the Great Lakes on a regional scale.	Theoretical only and has not been compared with actual data.	Ching et al. (1996); Binkowski and Shankar (1993)
Hybrid receptor-deposition model	Uses backward trajectory calculations and estimates dry deposition and gas exchange flux. Parameters incorporated into the model include transport distance, meteorological conditions, particle size distribution, and water surface roughness.	Estimates deposition of trace metals and semi-volatile organic compounds to Lake Michigan for the Lake Michigan Urban Air Toxics Study.	Experimental model; variation in the model depended on the nature of the chemical species and was ± 3-fold that of values in literature.	Pirrone and Keeler (1996)

TABLE III-1 (continued)
Summary of Several Atmospheric Transport and Deposition Models

Model	Description of Model	Great Waters Related Application	Goodness of Fit[a]	References
Hybrid single-particle Lagrangian integrated trajectories (HY-SPLIT) model[b]	This model is a hybrid between Eulerian and Lagrangian approaches. An initial single emitted particle is divided into multiple particles to represent the more complex flow field as gridded 3-dimensional wind data are introduced. Air concentrations are calculated on a fixed 3-dimensional grid.	Although this model has not yet been applied in Great Waters research, it may be a good way to evaluate alternative control strategies when the pollutant of interest does not undergo extensive transformation.	Not given; however, the spatial and temporal density of input wind data is probably one of the most important factors.	Cohen et al. (1995); Draxler (1988)

[a] "Goodness of fit" refers to how well the deposition estimates from the models correspond to actual measured deposition data.

[b] Not included in Baker et al. (1996).

Bullock et al. (1995) uses RELMAP (Eder et al. 1986) with relatively simple mercury parameterizations. An entire year of transport and deposition was simulated over a large computational domain (e.g., the continental United States) to obtain annually integrated results. The RELMAP-simulated annual results agree quite well with the majority of the limited annual deposition and concentration data available around the Great Lakes and in Florida, but the model cannot be well tested over the entire model domain without annual observations in a large number of additional locations. The RELMAP simulates chemical transformations only within individual Lagrangian pollutant "puffs." Therefore, the chemical interaction of emissions from widely separated locations is not considered. The other mercury model described (Seigneur and Constantinou 1995) suggests the importance of some processes not modeled by Bullock et al. (1995). Since this model is not used in a numerical simulation of observed conditions, however, the validity of the assumptions and parameterizations have not been tested. The complex chemical transformation processes outlined would not be well suited to a Lagrangian modeling framework, as they allow for the interaction of mercury emissions from widely separated sources.

An innovative approach for the modeling of semi-volatile organic compounds and their deposition to Lake Michigan is proposed by Pirrone and Keeler (1995). A hybrid receptor-deposition modeling approach has been developed to estimate the dry deposition flux of various HAPs to Lake Michigan. The approach uses backward trajectory calculations and parameterizations for dry deposition and volatile exchange with water surfaces to obtain an iterative correction estimate of the transport distance, horizontal dispersion and hydrodynamic parameters responsible for particle deposition fluxes and air-water vapor exchange rates. This technique appears to be very experimental and uses unconventional methods. However, it demonstrates a novel approach to estimating surface flux quantities that currently cannot be directly measured with confidence.

This generic survey of atmospheric transport and deposition models, although brief, should be sufficient to demonstrate that a variety of these models is now available. The models differ widely in the level with which they treat the important physical and chemical processes involved, in the applications for which they were designed, and in the input data and computational capacity they require. No single model is adequate for all types of air quality applications encountered in the Great Waters program. In selecting an appropriate model, a potential user should consider the questions to be answered, or hypotheses tested, by the model; the type, quantity, and quality of data available as input to the model; the available resources (e.g., computer capability, time); and the experience of the modeler with appropriate geographic scale and multimedia aspects of Great Waters concerns.

III.C Summary of Atmospheric Transport and Deposition Processes

Given the number of pollutants and waterbodies at issue for the Great Waters program, it is impractical to provide all the information needs through actual deposition and flux measurements. Thus, mathematical models of atmospheric transport and deposition models are used to provide estimates for spatial and temporal gaps in actual monitoring data bases, to test hypotheses about characterizations of atmospheric transformations and removal, and to predict the effectiveness of prospective control strategies. These mathematical models are also used to better understand specific processes (e.g., gas exchange at the air-water interface), to calculate the inputs and outputs of pollutants in a waterbody, and to link air pollutant sources with receptor bodies (e.g., the Great Waters). The models often must account for physical and chemical transformations during transport, as well as the depositional processes. As such, to develop the models, it is necessary to understand the processes by which the pollutants of concern are transferred across the air-water interface (i.e., wet deposition, dry deposition, and gas exchange processes), and create model parameters from this understanding. This chapter has briefly described what is currently known about these processes and research that is currently underway.

The brief summary of atmospheric transport and deposition models presented in this chapter demonstrates that a variety of these models is now available. The models differ widely in the level with which they treat the important physical and chemical processes involved, in the applications for which they were designed, and in the input data and computational capacity they require. No single model is adequate for all types of air quality applications encountered in the Great Waters program. In selecting an appropriate model, a potential user should consider the questions to be answered, or hypotheses tested, by the model; the type, quantity, and quality of data available as input to the model; the available resources (e.g., computer capability, time); and the experience of the modeler with appropriate geographic scale and multimedia aspects of Great Waters concerns.

CHAPTER IV
MAJOR WATERBODIES OF THE GREAT WATERS:
AN OVERVIEW OF PROGRAMS AND EFFORTS ADDRESSING ATMOSPHERIC DEPOSITION

Section 112(m) of the Clean Air Act specifically designates the Great Lakes, Lake Champlain, Chesapeake Bay, and other U.S. coastal waters as Great Waters. Researchers have found that the Great Waters have been impacted by metals, pesticides, toxic chemicals, and nutrients that deposit in the waters through different pathways, including atmospheric deposition.

This chapter is organized by sections on each of these major waterbodies of the Great Waters. For each section, information currently known about contamination occurring at the respective site is presented, including problems or issues that challenge each waterbody, followed by discussion of current strategies or efforts to respond to these concerns. The chapter is divided accordingly:

- Section IV.A presents information available on atmospheric deposition of persistent toxic pollutants from the Great Lakes and the many programs to characterize and reduce loadings;
- Section IV.B describes Lake Champlain and current research to assess atmospheric mercury deposition in the basin;
- Section IV.C discusses the impacts of nitrogen and toxic pollutants on Chesapeake Bay, and the modeling strategies to address these concerns;
- Section IV.D provides an overview of U.S. estuary programs and some major efforts to characterize loadings of nitrogen and toxic pollutants; and
- Section IV.E summarizes current information on addressing atmospheric deposition of pollutants of concern in the Great Waters.

Although this chapter describes data and programs specific to the subject waterbody, much of the information will be relevant to other waters as well. For example, those interested in smaller estuaries will gain insight from information presented in the Chesapeake Bay section.

The Great Lakes and Lake Champlain represent two important freshwater systems in the United States. Lakes are sensitive to pollution inputs because they lack any dominant, unidirectional flow, and as a result, there is a slow turnover of water and a resulting retention of pollutants.

The Great Lakes contain one-fifth of the world's supply of fresh surface water. These lakes have played a vital role in the history and development of the United States and Canada. They are stressed by a wide range of pollution sources associated with the large urban centers located on their shores. Because the Great Lakes system is a relatively closed water system (very large volume, with relatively small water inflows and outflows), many of the pollutants that reach the Great Lakes remain in the system for extended periods of time. For example, Lake Superior replaces all the water in the lake every 173 years, Lake Eric every 2.7 years.

Lake Champlain is located in the northeastern United States, shared by the States of New York and Vermont and the Province of Quebec. Although much smaller in surface area than that of the Great Lakes, Lake Champlain is still one of the largest freshwater lakes in the United States and its natural resources are important to the local economy. The Lake Champlain basin, or watershed, is much larger relative to its water surface area than the Great Lakes, and so watershed throughput is much more of an issue for Lake Champlain. Toxic pollutants are an issue of wide public concern in the Lake Champlain Basin, due in large part to fish consumption advisories for PCBs and mercury for both New York and Vermont, and the potential impact on drinking water and the Lake's many other uses.

"Coastal waters," for the purposes of CAA section 112(m), are defined as those estuaries designated for the National Estuary Program (pursuant to section 320(a)(2)(A) of the federal Water Pollution Control Act) or designated for the National Estuarine Research Reserves (pursuant to section 315 of the Coastal Zone Management Act). Chesapeake Bay is identified by name in section 112(m).

Estuaries occur where rivers empty into the ocean, mixing together fresh water and salt water, and creating an ecosystem distinct from, and more productive than, either fresh or salt water systems. Estuarine waters include bays, sounds, marshes, swamps, inlets, and sloughs. These environments are characterized by varying degrees of salinity, high turbidity levels, and complex water movement affected by ocean tides, river currents, and wind. Estuaries are critical coastal habitats that serve as spawning grounds, nurseries, shelters, and food sources for many different species of shellfish, fish, birds, and other wildlife. The leading environmental problems in estuarine systems at present are eutrophication,[13] contamination by toxic chemicals and pathogens (disease-causing organisms), over-harvesting, and loss of habitat.

In 1975, Chesapeake Bay became the nation's first estuary to be targeted for protection and restoration. Over the past decade, other coastal programs, such as the National Estuary Program, the National Estuarine Research Reserve System, and the Gulf of Mexico Program, have been established to protect and restore water quality and living resources in U.S. estuaries and coastal waters. Chesapeake Bay was also the first estuary where atmospheric sources of nutrients and toxic pollutants were recognized as significant inputs to the waterbody. Recently, research in other U.S. coastal waters has begun to evaluate the loadings of nutrients and toxic pollutants to their watersheds from atmospheric sources. The Great Waters program has focused primarily on Chesapeake Bay for estuarine issues, and has found that information gathered from this waterbody is directly applicable to some of the other east coast estuaries, with adjustments made for a respective waterbody's characteristics.

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[13] As discussed in Section II.D and in this chapter, eutrophication is over-enrichment of waters that is characterized by algae blooms, turbid waters, and low or no dissolved oxygen conditions.

IV.A The Great Lakes

The Great Lakes, comprised of Lakes Superior, Michigan, Huron, Erie, and Ontario, are an important part of the physical and cultural heritage of North America (see Figure IV-1). The Great Lakes ecosystem, the interacting components of air, land, water, and living organisms, including humans, is one of the largest surface systems of freshwater on earth. This ecosystem contains 18 percent of the world's freshwater supply and 95 percent of the surface freshwater within the United States. Only the polar ice caps and Lake Baikal in Siberia contain more freshwater. By virtue of their size, the Great Lakes affect the climate of the surrounding region. Areas of Michigan, Ontario, and New York generally have warmer, though snowier, winters than other parts of North America at similar latitudes because, as a result of little continual current, the lakes retain a large amount of heat. In spring and early summer, the lakes are slow to warm, thereby keeping the nearby land areas cool.

The Great Lakes sustain a rich diversity of fish, birds, and other wildlife. Native fishes important for commercial and recreational harvest include lake trout, lake whitefish, and walleye. Non-native species such as smelt, white perch, brown trout, rainbow trout, and several Pacific salmon species also contribute substantially to the total annual fish harvest. Approximately three million waterfowl follow the Atlantic and Mississippi flyways through the Great Lakes basin each year. Native animals include deer, fox, moose, wolves, beaver, mink and muskrat. In addition, the Great Lakes ecosystem supports more than 100 globally endangered or rare species (Nature Conservancy 1994).

Economic Highlights of the Great Lakes

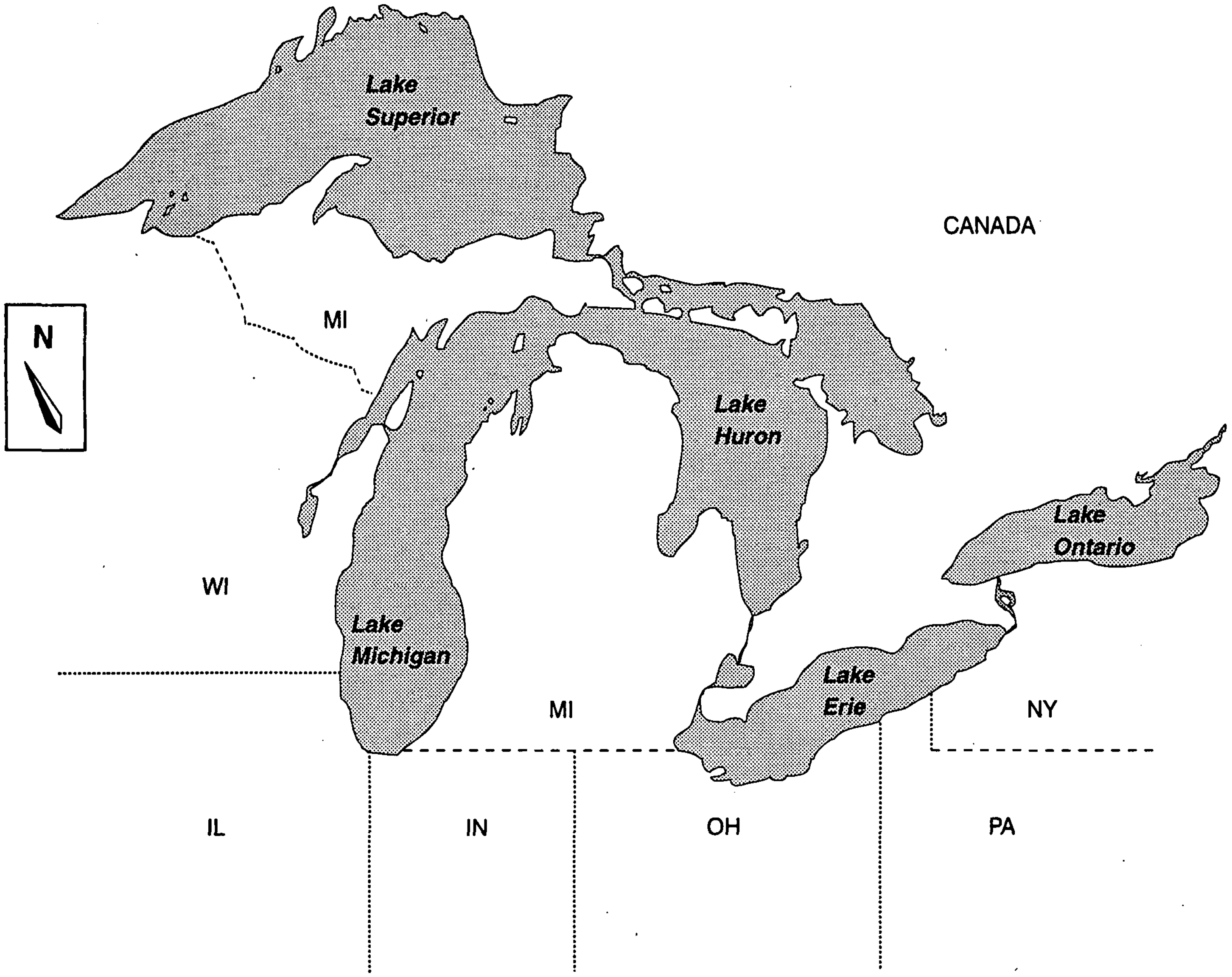
- Approximately 11% of total employment and 15% of manufacturing employment of combined U.S. and Canadian workers are located in the Great Lakes basin.
- Trade between Canada and the eight Great Lakes States in 1992 was valued at $106 billion (56.2% of the U.S.-Canada total).
- Estimated 900,000 to 1 million United States and Canadian boats operate each year resulting in a direct spending impact on the regional economy of more than $2 billion.
- About 2.55 million U.S. anglers fished the Great Lakes; total trip-related and equipment expenditures of $1.33 billion in 1991.

Source: Allardice and Thorp (1995)

The Great Lakes region is home to more than 40 million people, including 10 percent of the U.S. population and 50 percent of the Canadian population. Over 23 million of these people depend on the Great Lakes for drinking water. Industries use the water to make products, to cool manufacturing processes or power generation equipment, and to ship raw materials and finished products. Residents and visitors alike enjoy an abundance of recreational activities, including boating, fishing, sightseeing, camping, and hiking.

The concentration of human activities in the Great Lakes basin (e.g., manufacturing, transportation, agriculture, fishing) imposes stresses on the ecosystem and has prompted significant concerns for the health and well-being of the human residents. Many of the major stressors and resultant effects were documented in the First Great Waters Report to Congress. The current report builds on this information and presents some of the potential problems that may impact the Great Lakes basin from the perspective of the CAA, and major programs that are underway to address those problems.

FIGURE IV-1
Great Lakes Basin



Physical Features of the Great Lakes						
	Superior	**Michigan**	**Huron**	**Erie**	**Ontario**	**Totals**
Volume (cu. miles)	2,900	1,180	850	116	393	5,439
Maximum Depth (feet)	1,332	925	750	210	800	NA
Area (sq. miles)	31,700	22,300	23,000	9,910	7,340	94,250
Land Drainage Area (sq. miles)	49,300	45,600	51,700	30,140	24,720	201,460
Retention Time (years)	191	99	22	2.6	6	NA

NA = not applicable

The remainder of Section IV.A presents:

- Current knowledge and recent measurements of atmospheric levels and deposition of toxic pollutants in the Great Lakes;
- Information on major activities/programs currently in progress to assess atmospheric deposition of air contaminants to the Great Lakes;
- Efforts supported by the United States, as well as Canada, to reduce and mitigate atmospheric emissions in the Great Lakes basin; and
- Brief discussion on current information gaps, and future research needs to improve understanding of atmospheric deposition of pollutants into the Great Lakes.

Common Terminology for Air Deposition of Pollutants

FLUX

Transport of a chemical across an interface (e.g., between air and water) for a given area and time, accounting for both inputs and outputs. Net flux is equal all positive loadings minus all negative loadings.

INPUTS (positive loading)

Wet Deposition: Gases and particles carried in precipitation (rain, snow, sleet) and deposited on land and water surface.

Dry Deposition: Airborne particles falling by gravity or impacting through turbulence.

Gas Absorption: Gaseous form of pollutant crossing air-water interface into the water (portrayed as a positive number in a flux calculation).

Waterborne Discharge: Pollutant discharged directly to water (e.g., by industrial discharge, urban storm-runoff).

Tributary loading: Pollutants entering waterbody through connecting channels, streams, and rivers.

OUTPUTS (negative loading)

Volatilization or Gas Evasion: Gaseous form of pollutant crossing air-water interface into the air (portrayed as a negative number in a flux calculation).

Sedimentation: Settling of particles by gravity to bottom sediments.

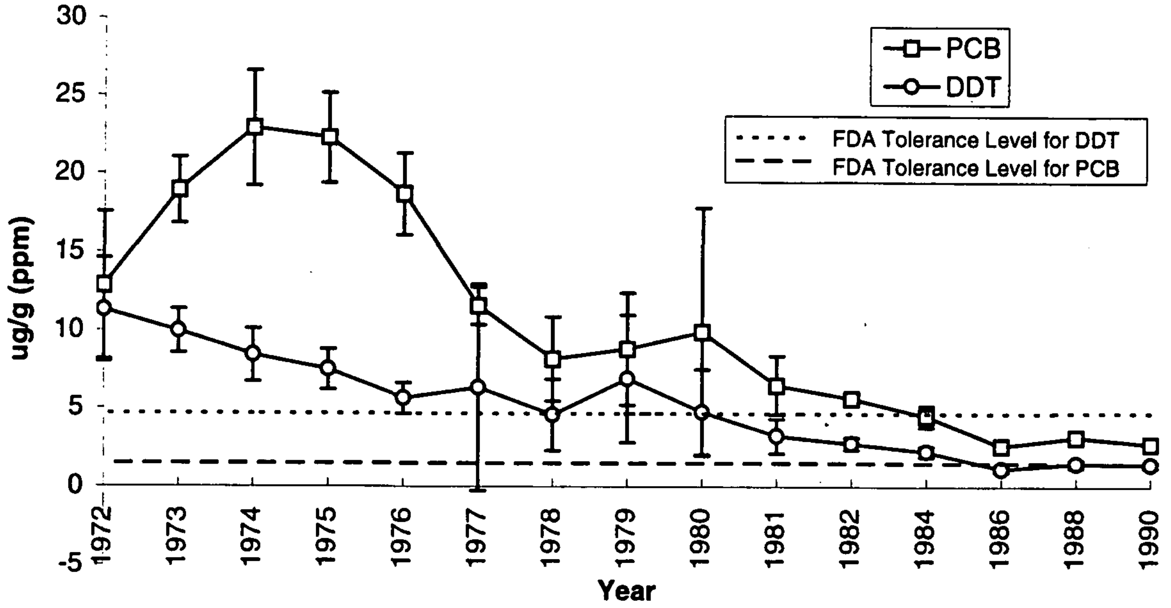
Outflow: Pollutants flowing with water out to rivers or to the ocean.

Atmospheric Deposition of Great Lakes Contaminants

Hundreds of man-made chemicals have been identified in the Great Lakes ecosystem. Unacceptable levels of certain bioaccumulative pollutants, such as mercury, DDT, and PCBs, remain in certain fish and wildlife species, and advisories have been issued by many Great Lakes states for these pollutants of concern (specific fish advisories are listed in Appendix B). For example, although concentrations of PCBs and DDT in Lake Michigan lake trout are currently about one-tenth of those of 20 years ago (Figure IV-2), the concentrations are still at levels that warrant issuance of public health advisories regarding the consumption of these fish. Advisories especially apply to vulnerable subpopulations, such as children, women who are pregnant or anticipate bearing children, and frequent fish consumers.

The pollutants of concern have been associated with health problems in certain fish and wildlife species, although with the decline of some pollutant levels, many species seem to be recovering. For example, the number of double-crested cormorants living on the Great Lakes has increased more than 20-fold during the past 15 years. Prior to this, numbers of these fish-eating birds declined during the 1970s due to reproductive failure from DDE-induced egg shell thinning. Health problems persist for fish and wildlife in certain locations,

FIGURE IV-2
PCB and DDT in Lake Trout from Lake Michigan



Source: DeVault et al. 1995.

particularly in waters with highly-contaminated bottom sediments, and for predators high in the food web, such as lake trout, mink, and bald eagles.

During the 1980s, studies in the Great Lakes showed that atmospheric deposition may be the major route of introduction of a number of pollutants to the Great Lakes. For example, atmospheric transport of toxaphene was implicated when the insecticide was found in fish in Lake Siskiwit, located on an island in Lake Superior Because the elevation of Lake Siskiwit is above that of Lake Superior, it does not receive any groundwater from Lake Superior and thus pollutant input. This pesticide was used on cotton crops in the southeastern United States, prior to its cancellation in 1982 (McVeety and Hites 1988; Swackhamer et al. 1988).

As a result of this and other findings, the United States and Canada established a joint monitoring network called the Integrated Atmospheric Deposition Network (IADN). The IADN is designed to assess the magnitude and trends of atmospheric deposition of target chemicals to the Great Lakes, and to determine emission sources wherever possible. The program responds to the Great Lakes Water Quality Agreement (GLWQA), specifically to the needs of Annex 15 which addresses issues concerning airborne contaminants in the Great Lakes basin. A more detailed discussion of the network rationale, design, and results is presented in the following section (Program Actions to Characterize Atmospheric Contamination in Great Lakes).

The first consensus data report for IADN (Eisenreich and Strachan 1992) revised and improved the very broad estimates of atmospheric deposition of toxic contaminants that were previously compiled (Strachan and Eisenreich 1988). More recent data were incorporated into deposition estimates for 1994 and compared to the earlier results (Hoff et al. 1996), shown in Table IV-1.

Atmospheric loadings of pollutants can occur as wet deposition, dry deposition, or through air-water gas exchange (i.e., absorption and volatilization) (these terms are defined in Chapter III). Atmospheric loadings of pollutants are calculated using atmospheric concentration data gathered by IADN and a combination of parameters that include lake surface area.

The remainder of this subsection presents atmospheric concentration and deposition data collected primarily from IADN on pollutants of concern, as well as information on current trends of pollutant deposition. The pollutants of concern to the Great Lakes include PAHs, PCBs, pesticides (e.g., DDE, DDT, lindane, toxaphene), and trace metals (e.g., lead, mercury). PCBs, toxaphene, and mercury are discussed in more detail below. These pollutants have high levels of contamination in the Great Lakes, resulting in the subsequent issuance of fish consumption advisories for these pollutants. Potential sources of the contamination and uncertainties in the data are also discussed.

TABLE IV-1
Atmospheric Loading Estimates[a] for Selected Pollutants (kg/year) in the Great Lakes

Pollutant of Concern	Superior	Michigan	Huron	Erie	Ontario
PCBs[b] (Wet and Dry)					
1988	550	400	400	180	140
1992	160	110	110	53	42
1994	85	69	180	37	64
PCBs[b] (Net Gas Transfer)[c]					
1988	-1900	-5140	-2560	-1100	-708
1994	-1700	-2700	--	-420	-440
DDT (Wet and Dry)					
1988	90	64	65	33	26
1992	34	25	25	12	10
1994	17	32	37	46	16
DDT (Net Gas Transfer)					
1988	-681	-480	-495	-213	-162
1994	30	67	--	34	13
Benzo(a)pyrene (Wet and Dry)					
1988	69	180	180	81	62
1992	120	84	84	39	31
1994	200	250	--	240	120

[a] Waterborne discharges are not included.
[b] Data presented for PCB congeners 18, 44, 52, and 101 (each with 3-5 chlorines in chemical structure).
[c] The convention is to assign a negative number to loss of pollutant from lake (i.e., volatilization). Thus, the resulting number expresses the mass of a pollutant going into or coming out of the lake per year (i.e., a positive net gas transfer indicates a net input of the pollutant to the lake and a negative net loading indicates a net loss or output from the lake).
-- Not determined or reported.

Source: Eisenreich and Strachan (1992), Hoff et al. (1996), and Strachan and Eisenreich (1988).

PAHs

Polyaromatic hydrocarbons (PAHs), a subset of POMs, are semi-volatile compounds produced in combustion processes and are widely distributed in the environment. As indicated in the First Report, about 80 to 90 percent of the input of one common PAH, benzo(a)pyrene, to Lakes Superior, Michigan, and Huron is attributed to atmospheric deposition.

PAHs are detected both in the gaseous and particulate phases, but some of the most toxic PAHs are largely in the particulate phase in the atmosphere. Thus for the most toxic PAHs, dry deposition is the main route of transport to the lakes (Hoff and Brice 1994). For benzo(a)pyrene, wet deposition seems to be the major source of atmospheric loadings to Lake Michigan in all seasons of the year. The net movement of the gaseous phase benzo(a)pyrene is largely to the lakes (see Table IV-2). Comparing recent wet and dry deposition values with historic data, the loading of benzo(a)pyrene to the lakes appears to have increased (Table IV-1). However, this finding may be attributed to an underestimation of benzo(a)pyrene in the 1992 studies (Hoff et al. 1996).

A recent study found that total wet and dry deposition for benzo(a)pyrene was 50 times higher at an urban site (Chicago) than at remote IADN sites in Lakes Michigan and Superior (Sweet and Harlin 1996). The investigators concluded that large areas of Lake Erie, Lake Ontario, and southern Lake Michigan have elevated PAH deposition rates due to emissions from urban areas. The relative abundance of individual PAHs is very similar at urban and nonurban sites, suggesting that little chemical degradation occurs during transport from urban source areas to rural and remote sites several hundred kilometers away.

PCBs

PCBs are a class of highly toxic, persistent, and bioaccumulative chemical compounds. PCBs were produced from 1927 to 1977 for the purpose of insulating and cooling electrical equipment. In the late 1970s, Monsanto Company, sole manufacturer of PCBs in the United States, voluntarily stopped production of PCBs. Estimates suggest that 282 million pounds of pure PCBs -- 20 percent of PCBs produced -- were still in service at the end of 1988.

PCBs manufactured before production was banned are still found in the Great Lakes. They have been detected in older commercial and industrial equipment (e.g., transformers, capacitors). There are no phaseout deadlines that require removal of this equipment to avoid breakage and release, although this equipment is tightly regulated under Toxic Substances Control Act (TSCA). As a result of past use and disposal practices, PCBs may reside in sediments in the water and in other areas, such as hazardous waste sites. As the contaminated sediment is disturbed, the PCBs may be re-released and resuspended in the water, allowing for continued bioaccumulation in Great Lakes fish. Current remediation programs are in place to address PCB-contaminated waste sites. Other continuing PCB sources include unregulated sources that potentially contain PCBs and releases or uses that are unknown, as well as releases by those PCB owners who are not aware of the presence of PCBs or of the special management requirements for PCB-containing equipment.

PCBs in Great Lakes fish have long been linked to developmental and growth problems in infants born to women who regularly consumed PCB-contaminated fish in the late 1970s. PCBs are one of the most tightly regulated and controlled group of pollutants in the United States under TSCA. Despite existing controls, however, fish consumption advisories are still issued for PCBs in all five of the Great Lakes (see Appendix B). For example, although PCB levels have declined in Lake Michigan water, there has been a constant or increasing level of PCBs in some Lake Michigan fish, possibly due to resuspension from sediment (see Section II.B).

TABLE IV-2
1994 Average Atmospheric Loadings[a] of Selected Pollutants to the Great Lakes (kg/year)

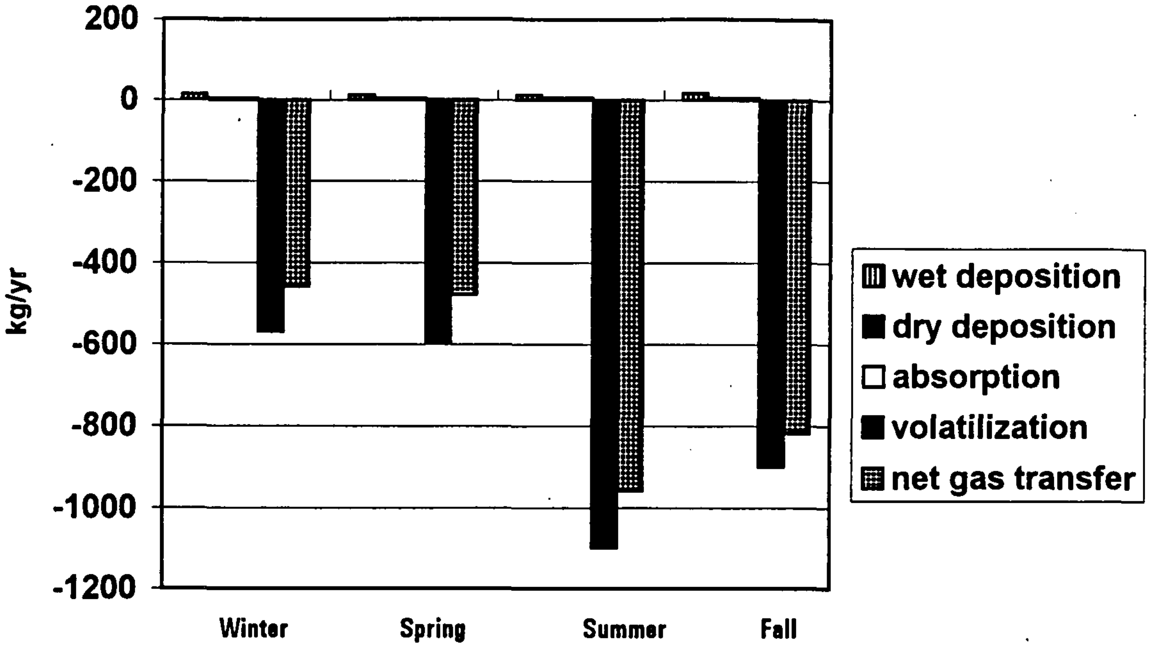
Atmospheric Process	Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario
PCB					
Wet deposition	58	52	180	21	38
Dry deposition	27	16	---	16	5.7
Net gas transfer[b]	-1700	-2700	---	-420	-440
Absorption	320	390	---	340	130
Volatilization	-2000	-3100	---	-760	-560
Dieldrin					
Wet deposition	21	58	10	28	11
Dry deposition	7.4	8	---	5.6	1.7
Net gas transfer	-780	---	---	-610	-320
Absorption	120	200	---	67	43
Volatilization	-910	---	---	-680	-370
DDE					
Wet deposition	2.6	3.8	10	4.6	4.5
Dry deposition	0.4	0.5	---	0.5	0.2
Net gas transfer	---	---	---	---	-170
Absorption	10	26	---	14	12
Volatilization	---	---	---	---	-180
Lindane					
Wet deposition	78	62	170	84	53
Dry deposition	1.1	1.5	---	1.2	0
Net gas transfer	370	-140	---	100	-41
Absorption	1300	1000	---	570	200
Volatilization	-930	-1200	---	-470	-240
Benzo(a)pyrene					
Wet deposition	140	170	---	180	56
Dry deposition	58	77	---	63	60
Net gas transfer	87	---	---	---	---
Absorption	100	92	---	51	7.5
Volatilization	-17	---	---	---	---

[a] Based on data collected from Integrated Atmospheric Deposition Network (IADN) and summarized in Eisenreich and Strachan (1992), Hoff et al. (1996), and Strachan and Eisenreich (1988).

[b] Net gas transfer is the sum of gas absorption and volatilization. Water concentration data are taken from past literature and compared with the more recent air measurements, which may lead to some potential error in gas transfer estimates (Hoff et al. 1996). Values for net gas transfer are rounded off and thus estimates may not add up in the table.

--- Not determined or reported

FIGURE IV-3
Seasonal Atmospheric Loadings of PCBs in Lake Michigan (1994)



Source: Hoff et al. 1996.

Volatilization is the dominant removal mechanism of PCBs from the water to the atmosphere (Table IV-2). Figure IV-3 shows how large volatilization is compared to wet and dry deposition in Lake Michigan for all seasons. In 1994, volatilization of PCBs from the Great Lakes was estimated to be as high as 3,100 kg/year for Lake Michigan and 2,000 kg/year for Lake Superior. In contrast, estimates of wet deposition of PCBs in 1994 were less than those for volatilization and were nearly the same for estimates for Lakes Superior and Michigan (52 and 58 kg/year, respectively), with Lake Erie showing a lower rate (21 kg/year) and Lake Huron showing the highest rate (180 kg/year) (Table IV-2). Dry deposition rates of PCBs were similar in Lakes Superior, Michigan, and Erie (16 to 27 kg/year), with Lake Ontario showing a lower rate (5.7 kg/year). As presented in Table IV-1, from 1988 to 1994, wet and dry deposition of PCBs to each of the Great Lakes have decreased. Therefore, the net loss to the atmosphere would suggest that PCB concentrations in water are declining. Tables IV-1 and IV-2 represent data only for the atmospheric movement of pollutants and therefore, do not indicate the waterborne inputs to each lake (such as particles in the water, industrial water discharges, and especially urban storm-runoff which goes directly into the lakes). To date, no studies on seasonal variation in PCB concentrations in water have been published (Hornbuckle et al. 1995).

PESTICIDES

Volatilization of the pesticides dieldrin and DDE (a metabolite of DDT) for many of the Great Lakes was a significant process during 1994. Table IV-2 shows the net atmospheric loading to be negative, indicating that movement of these two pesticides between air and water is mostly volatilization. In contrast, gaseous lindane generally seemed to be in equilibrium at the lakes, with deposition inputs roughly equaling outputs. Inclusion of waterborne inputs may show a net input of pesticide to all lakes. Fluctuations in gas equilibrium conditions may be influenced by the water concentration data, differences in temperature, and/or errors in the Henry's Law constant used in calculating gas movement. For example, DDT net gas transfer estimates are hindered by difficulties in obtaining precise water concentration data because DDT levels in the lakes are close to the analytical detection limit. From Table IV-1, DDT wet and dry deposition loadings declined between 1988 and 1992, but rose slightly for all lakes except Lake Superior in 1994 (Hillery et al. 1996).

For many pesticides, gas transfer is strongly dependent on seasons, with net outputs in the summer and net inputs in the winter (Achman et al. 1993; Dolan et al. 1993; McConnell et al. 1992). For example, loadings of lindane to Lake Michigan in 1994 showed that the net gas transfer is into the lake in the winter and spring and out of the lake in the summer and fall. Wet and dry deposition of lindane appeared to be fairly uniform across all lakes and were less important than gas transfer (see Table IV-2).

Toxaphene in Lake Trout

Since 1991, the State of Michigan has issued a consumption advisory for Siskiwit lake trout from Lake Superior based on exceedance of the FDA's 5.0 ppm action-level for toxaphene. In 1995, the Canadian Province of Ontario issued fish consumption advisories for several different species in Lake Superior and upper Lake Huron, triggered by their toxaphene levels and a lowering of Health Canada's action level for toxaphene to 0.2 ppm.

Toxaphene in the Great Lakes Basin. Toxaphene, a semi-volatile insecticide containing a mixture of chlorinated bornanes (class of aromatic hydrocarbons), has been recognized as one of the contaminants with the highest concentrations in Great Lakes fish (Ribick et al. 1982; Schmitt et al. 1981, 1985, 1990). Because of its volatility and persistence, toxaphene is still widely distributed through the atmosphere, even though it is no longer used in the United States (Rapaport and Eisenreich 1986). Toxaphene was the second most abundant contaminant in coho salmon taken from four of the Great Lakes in 1980 (Clark et al. 1984) and the most abundant contaminant in lake trout and whitefish from Siskiwit Lake on Isle Royale, Lake Superior (Swackhamer and Hites 1988) (see sidebar).

Its discovery on pristine Isle Royale, exposed only to atmospheric contamination, seemed indicative of long-range transport via the atmosphere since the pesticide toxaphene had been used primarily in the southern United States. This hypothesis has been supported by studies that found toxaphene concentrations in Canadian air masses that had originated in the southern United States. However, there is also evidence to suggest that some of the toxaphene found in Lake Superior and northern Lake Michigan may have local origins. A study that analyzed fish from rivers in the southeastern United States, the Great Lakes, and Isle Royale, collected during 1982, found differences in toxaphene levels in fish between sites, leading to the conclusion that potential local influence may be important, rather than long-distance atmospheric transport from the southeastern United States to the Great Lakes (Petty et al. 1987). A recent study reported that differences in toxaphene levels in fish between sites in neighboring Arctic lakes was attributed to differences in food chains, where longer than normal food chains produced elevated toxaphene levels in upper trophic level fish (Kidd et al. 1995).

EPA recently supported monitoring of toxaphene in Great Lakes fish and sediment which has revealed two trends. First, there is a statistically significant decline in the concentration of toxaphene in fish from most waters, as might be expected following reduced use and later cancellation of the pesticide. Second, there is no discernible decline in toxaphene levels in Lake Superior lake trout; toxaphene levels are higher than levels of other measured contaminants in fish from anywhere in the Great Lakes (refer to Section II.B).

At this time, there are several possible explanations for the relatively elevated levels of toxaphene observed in Lake Superior and northern Lake Michigan. First, the continued use of toxaphene and subsequent atmospheric transport to the Great Lakes basin by other countries may increase levels. Another possibility is the previous local use of the pesticide. Toxaphene was once used to kill undesirable fish communities (Lockhart et al. 1992; Muirhead-Thompson 1971; Stern et al. 1993). This practice occurred in parts of Canada and the northern United States for fish restocking on small glacial lakes; it was applied to at least 80 lakes during the 1950s and 1960s in Wisconsin (Hughes 1968). There is also greater persistence of toxaphene in colder, less productive waters such as Lake Superior. Lake trout analyzed from Lake Superior may be slower to reflect a decrease in contaminant levels in their food web because of their greater age. However, age may not be a likely explanation for higher toxaphene levels in Lake Superior, since older fish should metabolize toxaphene when they are initially exposed and then dilute it via subsequent growth. Finally, toxaphene may be or have been formed during the production of paper. There are 74 pulp and paper mills that directly discharge to all the Great Lakes (IJC 1995). The paper industry is most concentrated near Lake Superior and upper Lake Michigan (Green Bay), providing a general circumstantial association with the highest concentrations of toxaphene in Great Lakes trout and sediment. The U.S. pulp and paper industry has taken many steps during recent years to reduce dramatically the toxicity of its discharges. A similar improvement is believed to be underway in Canada.

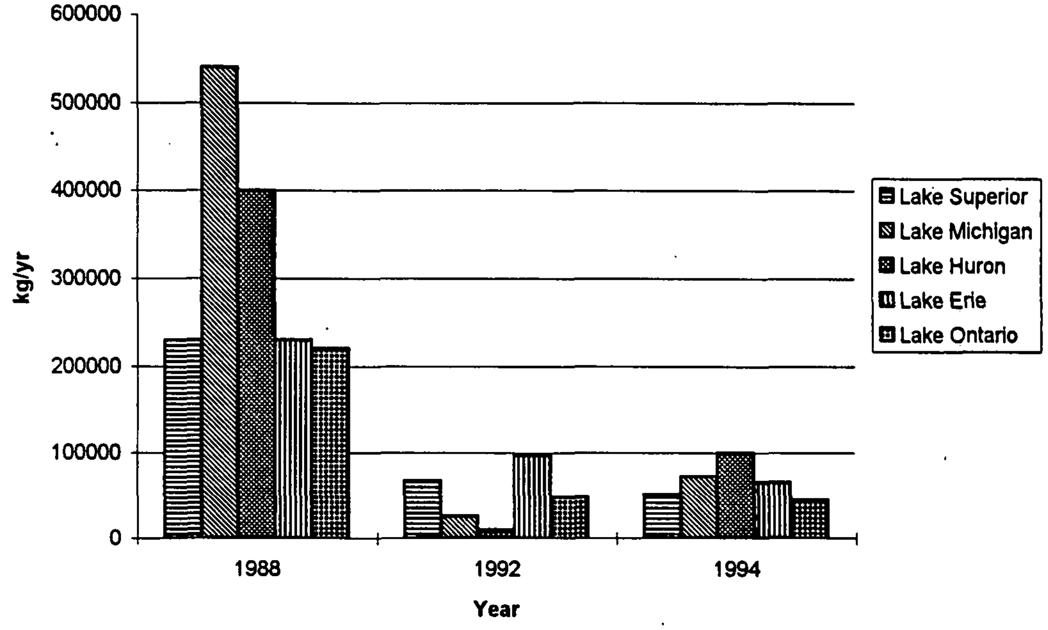
Uncertainties Related to Toxaphene in the Great Lakes

- ***Collected data:*** Evidence of continuing presence of toxaphene in the upper Great Lakes is based on data collected in the early 1990s (1992 fish tissue data, sediment cores through 1990). It is possible that past local sources have by now taken steps to prevent generation of such contamination. Difficulties also exist in showing a clear distinction between long-range inputs and local ones, and establishing definitive relationships between specific sources and receptor sites.
- ***Toxicity:*** Measurement of toxaphene is difficult because of the complexity of the compound. Certain components in the mixture of toxaphene are more toxic than others. Until analytical advances allow toxicity on a compound-by-compound basis to be more fully established, there will be some uncertainty about the magnitude of risk posed by environmental levels. In addition, such ubiquitous contaminants as PCBs, DDE, and other organochlorine pesticides can interfere with measurement of toxaphene.
- ***Representativeness of sediment cores:*** Toxaphene contamination trends should be analyzed under sediment cores from northern Lakes Michigan and Superior and the Apostle Islands. These cores are considered representative of contaminant loadings because they have revealed historical trends for PCBs and DDT that tend to fit with past use patterns.

TRACE METALS

A number of trace metals are of concern in the Great Lakes, though new data are limited. Data from 1994 suggest that wet deposition is the dominant transport mechanism for trace metals to the Great Lakes (Hoff and Brice 1994), but data are not shown. The most consistent trend in the deposition of trace metals was the reduction in lead in 1994 compared with 1988 values for all the lakes (Figure IV-4). This finding is not surprising given the phaseout of leaded gasoline in the United States beginning in the 1970s and accelerating in the mid-1980s. The gaseous phase of lead is assumed now to be negligible. Arsenic deposition also has decreased. The reason for this finding is not as clear but it has been hypothesized that process changes by Noranda, a major emitter of arsenic in Canada through mining, smeltering, and refining of metal products, may have led to the decline.

FIGURE IV-4
Atmospheric Loadings of Lead to the Great Lakes (1988-1994)



Source: Hoff et al. 1996.

Mercury in the Great Lakes Basin. Currently, six of the eight Great Lakes states (Michigan, Ohio, Wisconsin, New York, Pennsylvania, and Minnesota) have issued advisories restricting consumption of fish from some state waters due to mercury contamination. Mercury contamination, or high mercury levels in fish tissue, is also the most frequent basis for fish advisories issued by the Province of Ontario. Many of their advisories are applicable to areas of the Great Lakes.

During the early 1970s, mercury was found in fish from Lake Huron, Lake St. Clair, western Lake Erie, eastern Lake Ontario, and the St. Lawrence River at levels that led the United States and Canada to close commercial fisheries. Subsequently, mercury levels fell in these waters, because of modification or closure of certain chloralkali facilities and pulp and paper mills whose wastewater discharges contained large quantities of mercury. In 1970, mercury levels in Lake St. Clair walleye were 2 parts per million (ppm); by the mid-1980s, levels in these walleye had subsided to 0.5 ppm (Environment Canada et al. 1991). There are other signs of reduced mercury levels in the Great Lakes through dated sediment cores and populations of smelt in the lakes (refer to Section II.B).

For most surface waters, especially those not as industrialized as the Great Lakes, atmospheric deposition is the principal source of mercury contamination (U.S. EPA 1994a). Atmospheric levels of mercury in the northern hemisphere have increased three-fold during

For most surface waters, especially those not as industrialized as the Great Lakes, atmospheric deposition is the principal source of mercury contamination (U.S. EPA 1994a). Atmospheric levels of mercury in the northern hemisphere have increased three-fold during the past 150 years. Yet, there is some heartening evidence that mercury deposition may have decreased during the last several decades in industrialized areas of Europe and America (EPRI 1994). In the upper midwestern United States, a peat bog core from northeastern Minnesota indicates that mercury deposited during the 1980s was one-third less than average deposition during the preceding three decades (Benoit et al. 1994). This improvement could be attributable to declining mercury use in the United States, which should be accompanied by declining anthropogenic releases and atmospheric concentrations. Domestic mercury consumption fell by two-thirds between 1980 and 1992 (Lawrence 1994). However, broad conclusions about mercury deposition trends are uncertain, since the number of peat cores that have been analyzed are few and may reflect changes in local sources.

Since large discharges of mercury were terminated following implementation of the Clean Water Act (CWA), the atmosphere is the dominant pathway by which mercury reaches the Great Lakes. Currently, the best estimate of atmospheric deposition to the five Great Lakes is 6804 pounds per year (15,000 pounds per year) (Eisenreich and Strachan 1992). Virtually all loadings of mercury to Lakes Superior and Michigan are via the atmosphere, while the atmospheric contribution of mercury to Lake Ontario is relatively modest because it receives mercury from the upper lakes (Sitarz et al. 1993). Even some of the mercury borne to the Great Lakes via its tributaries includes contamination previously deposited from the atmosphere to their watersheds.

Unlike other trace metals, mercury exists in the air predominantly in the gaseous phase due to its volatility. Wet and dry deposition of mercury to Lake Superior were about five times higher than volatilization to the atmosphere. Net annual atmospheric loadings of mercury to Lake Superior were calculated to be 635 kg/year (Hoff et al. 1996), which was represented by:

- 560 kg/year as wet deposition;
- 250 kg/year as dry deposition;
- 65 kg/year as absorption; and
- -240 kg/year as volatilization.

In an earlier study, gaseous phase mercury represented 1.57 ng/m^3, particulate phase mercury, 0.02 ng/m^3, and precipitation mercury, 10.2 ng/L, at a northern Wisconsin site near Lake Michigan (Fitzgerald et al. 1991). Comparison of these values with other sites are presented in Table IV-5.

In Michigan, atmospheric concentrations and wet deposition of mercury have been observed to vary geographically. Northern Michigan received only one-half the wet deposition of mercury deposited to southern portions of the state. Wet deposition varied by season, with mercury concentrations two times greater during spring and summer than during winter. Higher levels of particulate mercury were observed in large urban areas. Modeling indicated that the dominant sources of mercury were located to the west, southwest, south, and southeast (Keeler and Hoyer 1995).

Program Actions to Characterize Atmospheric Contamination in the Great Lakes

Research has occurred in the past few years to increase understanding of the effects, fate, and transport of toxic substances in the Great Lakes ecosystem. These efforts are designed to provide information to further characterize, as well as reduce, atmospheric contamination in the Great Lakes region. Some of the programs to assess the extent of atmospheric contamination in the Great Lakes basin are described below. At this time, many of these projects are collecting and/or compiling data, and results are not yet available for evaluation. This information should be in the next Great Waters Report to Congress. Also, several notable programs/activities have been introduced in recent years to begin to reduce loadings and to mitigate existing contamination and are discussed in the following section (Toxics Reduction Efforts in the Great Lakes).

LAKE MICHIGAN ENHANCED MONITORING PROGRAM

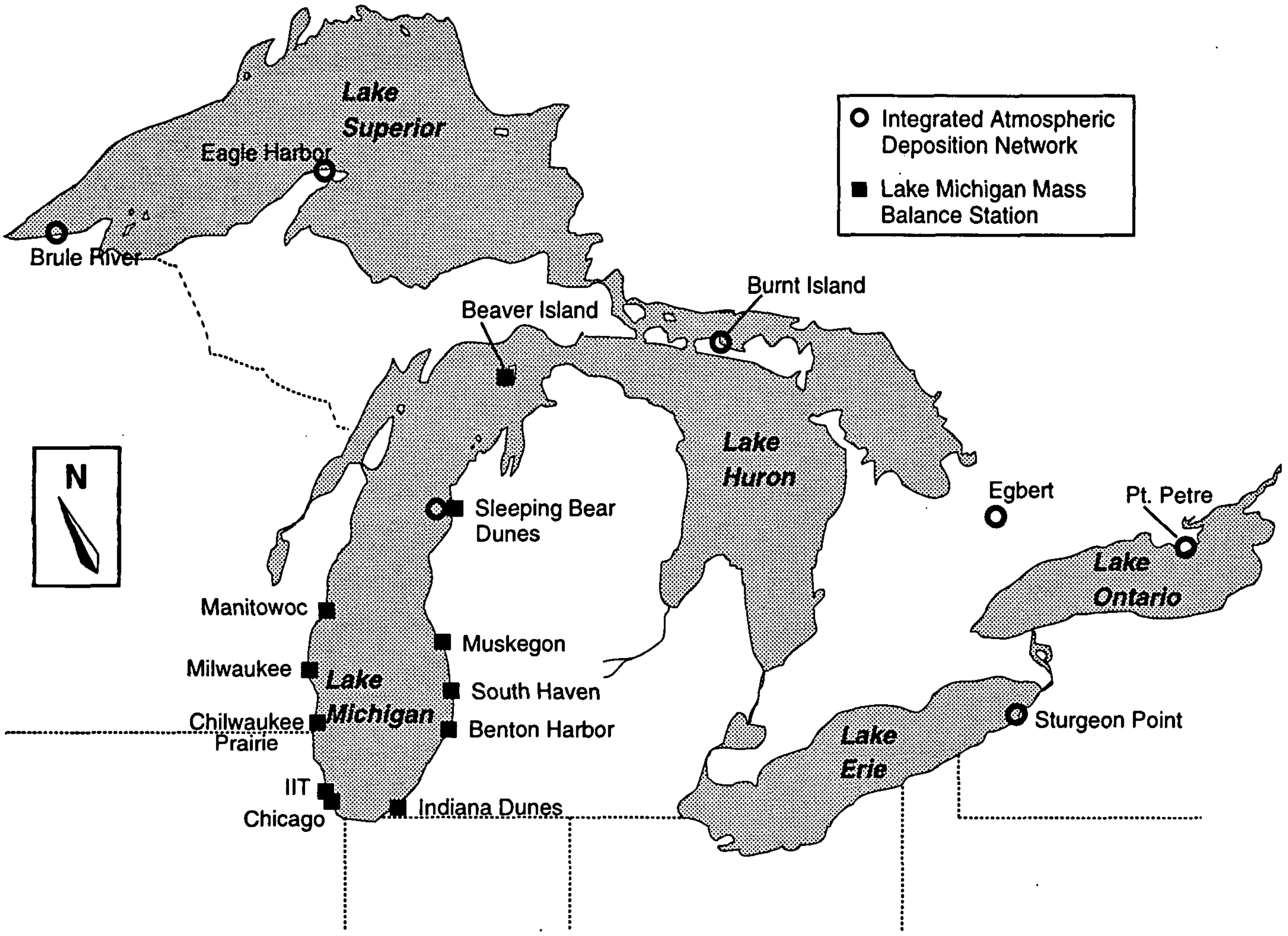
The Enhanced Monitoring Program for Lake Michigan is being implemented to support a number of activities that address reductions in the release of toxic substances, particularly persistent, bioaccumulative substances, to the Great Lakes system. The Program is a key element of the Lakewide Management Plan (LaMP) for Lake Michigan (see section on LaMPs).

The water quality criteria and values provided in the GLWQ guidance, once adopted by the Great Lakes states, would apply to the entire Great Lakes system, regardless of the source of pollutants to those waters. In this manner, the proposed water quality criteria and the measured values provide the basis for integrating actions carried out under the range of environmental programs available to federal, state, and tribal agencies to protect and restore the Great Lakes ecosystem. The mass balance approach will facilitate this integration by evaluating multi-media load reduction actions required to ensure that Lake Michigan water quality meets the Great Lakes water quality criteria (GLWQC).

The primary goal of the Lake Michigan Enhanced Monitoring Program is to develop a sound, scientific base of information to guide future toxic load reduction efforts at federal, state, tribal, and local levels. In particular, the following specific objectives are identified:

- Evaluate relative loading rates of critical pollutants by media (atmospheric deposition, contaminated sediments, tributaries) and to establish a baseline loading estimate to gauge future progress;
- Develop the predictive ability to determine the environmental benefits of specific load reduction scenarios for toxic substances and the time required to realize those benefits, including evaluation of benefits of existing environmental statutes and regulations as required under section 112(m) of the CAA, and section 303 of the CWA; and
- Improve our understanding of the key environmental processes that govern the cycling and bioavailability of contaminants within relatively closed ecosystems.

FIGURE IV-5
Atmospheric Monitoring Sites in the Great Lakes



Lake Michigan Mass Balance Study. One of the ways to address the objectives of the Lake Michigan Enhanced Monitoring Program, as well as to assist EPA in meeting the requirements of section 112(m) of the CAA, is through a mass balance study. EPA believes that the CAA requirements will be advanced through a coordinated effort to quantify and characterize the loadings, transport, and fate of selected pollutants in a defined ecosystem, through monitoring and modeling, and then by applying these measuring and estimating techniques to other ecosystems. Therefore, EPA initiated the Lake Michigan Mass Balance Study, a comprehensive sampling effort that includes measurements of contaminants in the atmosphere, tributaries, lakewater, sediments and food chain, to support model components. The modeling effort is central to the study; data are collected to support the model, which integrates the measurements and our knowledge of the Great Lakes system. The sampling sites for this mass balance study is shown in Figure IV-5.

The Lake Michigan Mass Balance model is constructed for a limited group of pollutants (PCBs, trans-nonachlor [the most bioaccumulative form of chlordane], and total mercury) present in Lake Michigan at concentrations that pose a risk to aquatic and terrestrial organisms (including humans) within the ecosystem, or that may accumulate to problematic concentrations in the future and that can serve as examples for other chemicals. In addition, atrazine (under consideration for addition as a Great Waters pollutant of concern), a commonly used herbicide in the Great Lakes basin and elsewhere in the United States, was also modeled. This herbicide has been reported at elevated concentrations in Lake Erie tributaries, in the open waters of the Great Lakes, and in the atmosphere over the lakes. The inclusion of this chemical will provide a model for the more reactive, biodegradable compounds in current use. The model will be less comprehensive than that for PCBs and trans-nonachlor, since atrazine does not appreciably bioaccumulate and it will not be analyzed in the food chain.

Application of Modeling Tools from Green Bay Mass Balance Study

In a pilot mass balance study by EPA and the Wisconsin Department of Natural Resources, water-insoluble organic compounds were monitored in Green Bay, Wisconsin, from 1988 to 1992. The analytical and modeling tools used in the study may be applied to the Great Lakes, Lake Champlain, and coastal estuaries. The **Lake Michigan Mass Balance study** is the first full-scale application of this methodology for toxic pollutants and will serve as the basis of any future mass balance efforts for persistent, bioaccumulative chemicals. Data collected from this model are anticipated at the end of 1996.

The chemicals chosen cover a wide range of chemical and physical properties and are representative of other classes of compounds that could pose water quality problems. This approach will allow modeling of many other chemicals with limited data. Resource limitations, quality assurance requirements, and analytical and data handling limitations preclude intensive monitoring and model calibration for more than the above described target chemicals. While mass balance modeling will focus on the above chemicals, the determination of loadings and concentrations for additional contaminants and compounds useful for source apportionment and deposition modeling will be undertaken as part of the Lake Michigan Enhanced Monitoring Program. The development of calibrated models will allow the CAA requirements for other HAPs to be met with limited monitoring data and will allow future resources to be directed to other areas, such as emissions inventories and dispersion modeling.

The Lake Michigan Mass Balance Study includes an atmospheric monitoring component to address research issues concerning urban and atmospheric deposition and exchange processes. The data will be used to calculate atmospheric loads to the ecosystem. Within this air transport component, special studies are being performed to determine whether emissions of HAPs from the urban coastal regions (Chicago, IL, and Gary, IN) contribute to atmospheric deposition to the adjacent waterbodies. The objectives of these special studies are to (1) measure wet and dry deposition fluxes of urban contaminants, (2) determine contributions of urban source categories to measured concentrations and deposition rates, and (3) assess the extent of air-water exchange of contaminants.

Previous studies indicated that urban emissions have a large impact on atmospheric concentrations of air toxics and on atmospheric deposition to the Great Lakes. Dry depositional flux of PCBs from Chicago was shown previously to be three orders of magnitude higher than that of non-urban areas (Holsen et al. 1991). In addition, the Lake Michigan Urban Air Toxics Study demonstrated that concentrations of several pollutants were significantly higher in Chicago urban areas than at less urbanized sites (Keeler 1994). An intensive study was recently conducted around Chicago to assess the impact of the urban area on atmospheric deposition and exchange with Lake Michigan, with three land-based monitoring sites around Chicago and one over-water site on a research vessel approximately five miles off the shore of Chicago. The three sampling periods which occurred between 1994 and 1995 were designed to provide information to track atmospheric plumes over and across the lake. Wet deposition, dry deposition, and lake water were analyzed for semi-volatile compounds (such as PCBs and PAHs) and trace metals (such as arsenic, mercury, and lead). All samples were taken on the same day to provide information on air-water exchange of contaminants. Results from this study are expected in autumn 1996. The information gathered from this study is expected to contribute useful information on urban impact on Lake Michigan, as well as to address process-oriented research issues and provide data in support of source apportionment and trajectory modeling.

INTEGRATED ATMOSPHERIC DEPOSITION NETWORK (IADN)

As mentioned earlier in this chapter, IADN is a long-term, binational program between the United States and Canada to assess the magnitude and trends of atmospheric deposition of target chemicals to the Great Lakes and to determine emission sources wherever possible. The program addresses the mandate of the Great Waters program and the needs of Annex 15 of the GLWQA between Canada and the United States. It is designed to (1) provide the necessary standardized methods, monitoring data, and loadings estimates to assess the relative importance of atmospheric deposition compared to other inputs, (2) determine temporal trends and geographic variations in deposition, and (3) ultimately provide information on sources of these atmospheric pollutants. It is a combination of a surveillance/monitoring network and a research program. Its goals are source attribution, process identification, and assessment of atmospheric impacts on environmental systems. At this time, annual and seasonal averages have been completed for four years of IADN operation. Data for selected pollutants were presented earlier in this section. Additional details may be found in Eisenreich and Strachan (1992), Gatz et al. (1994), and Hoff et al. (1996).

Compounds Measured in IADN

- ***Highest priority group:*** PCBs, lindane, PAHs, and lead. These pollutants are chosen for the first phase of IADN (1990-1992) to demonstrate the feasibility and accuracy of sampling and analytical methods.
- ***Second priority group:*** Chlorinated pesticides, HCB, DDT/DDE/DDD, trans-nonachlor, methoxychlor, mirex, dieldrin, aldrin, and trace metals, such as arsenic, selenium, cadmium, and mercury. Except for mercury, the sampling and analysis methods for most of these species had been implemented by 1993. Mercury monitoring was added at each IADN site by 1995.
- ***Third priority group:*** Compounds such as toxaphene, dioxins/furans, and agrochemicals which have an important atmospheric component but require additional methods development to accurately measure their concentrations in atmospheric deposition samples.

Target compounds were chosen for IADN based on their potential to bioaccumulate, their tendency to be transported atmospherically, and the availability and efficiency of detection methods (see sidebar). The benefit of IADN is the ability to monitor long-term atmospheric concentration changes of such compounds as PCBs and other chemicals of concern. In the past, such regional-scale atmospheric data have been sparse.

Urban Influence on Atmospheric Deposition of Contaminants

The primary focus of IADN is to determine regionally representative atmospheric deposition loadings of toxic chemicals to the Great Lakes. Thus, monitoring stations were strategically positioned to minimize the influence of local sources and to monitor the atmospheric environment over the lakes as much as possible. This approach does not directly enable the determination of the role of urban air pollution. Recent research suggests that deposition of contaminated large particles carried by winds passing over urban areas can result in substantial inputs of toxic chemicals to the Great Lakes (Holsen et al. 1991).

The influence of pollution from the Chicago-northwest Indiana area on water quality in southern Lake Michigan was studied by Sweet and Basu (1994). The Sleeping Bear Dunes site (in the State of Michigan) is located one kilometer from the northeastern shore of Lake Michigan and 50 kilometers from the nearest urban area or major source and, thus, is considered a "remote" site. The first urban site is located 1.5 kilometers from the shore on the campus of the Illinois Institute of Technology, which is near major expressways and surrounded by commercial and residential areas. The second urban site is located at the Indiana Dunes National Lakeshore in the vicinity of large steel mills. Prevailing southwest winds carry the urban emissions out over the southern portion of Lake Michigan (Sievering 1976). Particulate concentrations were measured for target compounds (PCBs, pesticides, and trace metals). Gas concentrations of PCBs and pesticides were determined, and rain was analyzed only for PCBs.

Results indicated that, for PCBs, DDT (and its metabolites), dieldrin, chlordane, and several trace metals (manganese, zinc, chromium, and lead), the measured values were 10 to 40 times higher in urban areas than at the remote site. For other pesticides (α-HCH, lindane, HCB) and trace metals (arsenic and selenium), concentrations were nearly the same at all three sites, indicating these pollutants were well mixed in the air throughout the region (i.e., few local sources).

Though 90 to 99 percent of the PCBs were found in the gas phase, the most toxic PCB congeners were enriched in the particulate phase. Dry deposition may be an important transport mechanism for PCBs to the lakes. Urban particulate matter also carried high concentrations of trace metals and pesticides, causing dry deposition of these materials to southern Lake Michigan. Dry deposition of large particles may be especially significant for Lake Michigan because 200 kilometers of the southwest shoreline are heavily developed. Prevailing southwest winds carry emissions over the lake where they travel for 100 to 150 kilometers before reaching land again, allowing a significant portion of the deposition to enter the lake. Finally, the concentration of PCBs in precipitation is roughly the same in urban and rural sites. The relatively low levels found in urban precipitation may be due to the fact that many contaminants are collected in clouds or by rain upwind of polluted areas.

Clearly the influence of urban areas on atmospheric deposition to the Great Lakes is substantial, especially in heavily developed areas, such as the southwestern shores of Lake Michigan.

The network originally consisted of a research station on each of the Great Lakes, two stations in Canada and three in the United States, which were considered to be representative of regional deposition patterns: Eagle Harbor, MI, Sleeping Bear Dunes, MI, Burnt Island in Lake Huron, Sturgeon Point, NY, and Pt. Petre, Ontario (see Figure IV-5). Two sites were later added; one is at Brule River near Lake Superior and the other is at Egbert, Ontario. The IADN implementation design allows for periodic re-evaluation of the existing sites to determine whether other sites are needed. For example, it is now thought that an urban site should be added to the network based on data collected from recent studies (see sidebar).

At each IADN site, concentrations of target chemicals are measured in rain and snow (wet deposition), airborne particles (dry deposition), and airborne organic vapors. In addition, precipitation rate, temperature, relative humidity, wind speed and direction, and solar radiation are measured at each site. IADN results for selected pollutants are presented in Tables IV-1 and IV-2.

At this time, after five years of operation, many of the sampling and analysis issues of IADN have been resolved. For example, comparability of sampling and analytical procedures between jurisdictions was achieved through extensive laboratory intercomparison studies. However, the uncertainty in the analytical measurement of some compounds is still above the uncertainty threshold acceptable most policy makers. Toxic chemicals at extremely low concentrations in air, such as PCBs and some agricultural chemicals, have the highest uncertainties (over 40 percent), and seasonal and annual fluctuations in air concentrations of the chemicals may vary by up to 90 percent (Hoff et al. 1996).

Uncertainty in the deposition estimates are due to various elements: (1) general approximations for estimating deposition; (2) climatic and meteorological variations; (3) differences in the instrumentation and the scope and objectives of the various jurisdictions and agencies involved; and (4) estimation of factors used to calculate loadings (e.g., magnitude of Henry's Law constants, rates of contaminant transfer between the air and water). Despite these limitations, the reported estimates are the best that are currently available. Also, data on the concentration of contaminants in the water column for all the Great Lakes have improved recently with more samples being collected and analyzed.

GREAT LAKES EMISSIONS INVENTORIES

A significant step towards assessing the need to reduce atmospheric loads of hazardous air pollutants to the Great Lakes is to determine, categorize, and estimate the magnitude of the pollutant sources. By creating an emissions inventory data base, it is possible to identify the sources and source categories that contribute most to the total emissions in a given geographic area, as well as to model emissions transport and deposition. An air emissions inventory is typically a mathematical estimate of pollution through the use of an emission factor (i.e., a number that represents emissions per unit burned, produced, or processed). These emission factors are derived from actual measurements of the emissions from representative sources and are derived specifically for one type of process or process equipment. Emission factors can be used to estimate both the amount and type of pollutants being emitted from an air pollution source based upon the quantities of material processed.

The 1986 Great Lakes Governor's Toxic Substances Control Agreement specified provisions to address atmospheric deposition, including a commitment "to cooperate in quantifying the loadings of toxic substances originating from all sources, with the purpose of developing the most environmentally and economically sound control programs." In response to the governors' direction, the air regulatory agencies in the eight Great Lakes states and the province of Ontario began to work cooperatively in 1987 to investigate "the development of a computerized air toxics data base for the purpose of obtaining a better understanding of the nature and sources of toxic air emissions and their migration, dispersion, and resulting impact upon the Great Lakes basin." Under the auspices and management of the Great Lakes Commission (represented by the eight Great Lakes states) and with major funding from EPA's Great Waters program, the Great Lakes states began developing a *regional* air toxics emissions inventory. This first regional inventory is scheduled for completion in 1996 and is expected to compile 1990 to 1993 emissions data for 49 toxic air pollutants from point and area sources. Emissions data on toxic air pollutants from mobile sources will be developed in 1997. These 49 pollutants include mercury and

Components of Great Lakes Emissions Inventory

To date, the Great Lakes States have developed and tested two fundamental components of the inventory effort:

1. **The Air Toxics Emissions Inventory Protocol for the Great Lakes** that guides each jurisdiction's efforts to identify key sources and estimate emissions of the 49 pollutants.
2. **RAPIDS**, a data base software and data management system; the Great Lakes States may adopt RAPIDS (or some variant of it) for their State system, and may also submit their data for incorporation with the regional RAPIDS data base at EPA.

several persistent, bioaccumulative chemicals identified in the GLWQ Guidance. The continued partnership of the region's air regulatory agencies, now in its eighth year, and the high level of regional cooperation and coordination exemplifies the commitment to decreasing toxic deposition into the Great Lakes ecosystem. Yet the inventory must be accompanied by an ongoing commitment to further quantify, assess, and report on the effects of voluntary and regulatory reductions of air toxic emissions.

The key to the state's coordinated efforts is *The Air Toxics Emissions Inventory Protocol for the Great Lakes States*, developed in June 1994 as part of this project (see sidebar). The Protocol will be followed by each participating state ensuring that consistent, agreed-upon best methodologies are used among all states when compiling a quality-assured inventory. This Protocol is an evolving document and will be updated or revised as needed and agreed upon by all the Great Lakes states.

The second fundamental component in developing a toxic air emissions inventory is the Regional Air Pollutant Inventory Development System (RAPIDS), a multi-state pollutant emissions estimation and storage software system. RAPIDS is a state-of-the-art, networked, relational data management and emission estimation system, bridging each state's individual inventory and computer system to the regional RAPIDS repository of inventory data. This regional repository is expected to be located at EPA's Great Lakes National Program Office and will be accessible electronically to any interested party. RAPIDS' strength is considered to be its versatility. States can modify or build upon it to serve their particular needs. It can be used to estimate both toxic and criteria pollutant emissions from a single device within a facility or a complex grouping of devices and controls, or even across geographic areas, ranging in size from one facility to the entire Great Lakes region. It is designed to run on a personal computer and applies a flexible data model that can be easily expanded in the future to support multi-media, permitting, monitoring, reporting, and compliance activities in the states. Emission factors are uploaded from EPA's Factor Information Retrieval System (FIRE) which contains quality-rated emission factors for both criteria and hazardous air pollutants.

Using RAPIDS, the Great Lakes states' air regulatory agencies are building a comprehensive, updatable statewide and regional air toxics inventory for point, area, and (in the future) mobile sources for the 49 air pollutants. Each of the eight Great Lakes states will be responsible for compiling, uploading, and validating their state emissions inventory data. It is anticipated that the inventories will be updated on a one- or two-year basis.

Four states (Illinois, Indiana, Wisconsin, and Michigan) completed a pilot study of major urban areas along the southwest shore of Lake Michigan in December 1995 using RAPIDS and the Protocol. The states created an inventory of small point and area source categories in the 12 shoreline counties encompassing Chicago (Illinois), Gary (Indiana), and Milwaukee (Wisconsin) that contribute the most to the total emissions of the 49 pollutants of concern. These area sources include gasoline stations, foundries, asphalt and cement plants, and hospitals, among others. The project was the first rigorous test of the regional Protocol and the RAPIDS software.

The level of emissions resolution and the source categories contained in RAPIDS were planned to meet the modeling needs of Great Lakes air quality researchers. This inventory will be available for dispersion and deposition models to characterize source, source category, and geographic culpabilities, and for mass balance models to characterize media contributions.

Toxics Reduction Efforts in the Great Lakes

In recent years, several programs/activities have been developed approaches to reduce loadings and to mitigate existing contamination. These programs are described below. They may provide information to further characterize and reduce atmospheric contamination in the Great Lakes region. At this time, many of the programs are not implemented, but results should be available for the next Great Waters Report to Congress.

VIRTUAL ELIMINATION

The Great Lakes Water Quality Agreement of 1978 between the United States and Canada called for the "virtual elimination" of persistent toxic substances, especially those which bioaccumulate, from the Great Lakes basin. In keeping with the obligations of the Great Lakes Water Quality Agreement, two major efforts have occurred: (1) a pilot project sponsored by EPA to develop the framework to achieve virtual elimination of two pollutants, mercury and PCBs; and (2) development of the Virtual Elimination Binational Strategy (draft released in August 1996) between the United States and Canada to set goals to reduce the use and release of selected pollutants. Many of the recommendations from the pilot project were incorporated into the Binational Strategy.

Virtual Elimination Pilot Project. Since 1990, both countries have initiated separate efforts for virtual elimination. EPA began the Virtual Elimination Pilot Project in 1993, which was designed to answer the following question: "What options exist for improving the current regulatory and non-regulatory framework to encourage continuing reductions towards zero in the use, generation, and release of selected toxic substances?" The aim of the project was to identify barriers to achieving virtual elimination and to develop strategies to overcome these barriers.

The first iteration or "pilot" portion of the U.S. project focused on the reduction opportunities of two substances, mercury and PCBs. EPA held a meeting with stakeholders in the Great Lakes region in 1993 to share information on mercury and PCBs, and to offer participants the opportunity to make recommendations on ways to reduce the use and release of each pollutant. Based on the results of this meeting, a draft report was developed by EPA to identify options to reduce mercury (GLNPO 1995). A draft options paper for the virtual elimination of PCBs is currently being prepared by EPA. It is expected that this project will continue with the analyses of classes of substances rather than the use of a chemical-by-chemical approach.

Mercury presents an unusual challenge to society because of its semi-volatility, persistence, complex environmental chemistry, and tendency to bioaccumulate in fish. The draft mercury options paper (GLNPO 1995) proposed regulatory and voluntary measures to prevent or reduce atmospheric mercury contamination, and introduced the concept of the mercury "life cycle." A comprehensive approach to virtually eliminating mercury releases was proposed:

- Increase public awareness of mercury problems and mercury-containing items;
- Influence supply of mercury to minimize primary production and manage federal holdings;
- Minimize use of mercury through pollution prevention and alternative technologies;
- Reduce uncontrolled releases by encouraging recycling and regulating releases; and
- Manage disposal of mercury-containing items and mercuric wastes.

Virtual Elimination Binational Strategy. Based on many of the recommendations from the Virtual Elimination Pilot Program, a draft of the *Canada-United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes Basin,* also known as the Binational Strategy, was released in August 1996 for public comment (strategy is expected to be finalized in October 1996). This Binational Strategy was developed jointly by EPA and Environment Canada and responds to the recommendations of the International Joint Commission (IJC). Both Canada and United States have domestic virtual elimination strategies but a coordinated strategy is necessary for the greatest reduction in toxic substances throughout the Basin. Both nations encourage and support voluntary programs by industries to reduce generation, use, and release of toxic contaminants to the Great Lakes.

The Binational Strategy provides the framework to achieve quantifiable goals in a specified timeframe (1996 to 2005). Flexibility is provided in the Strategy to allow for the revision of targets, timeframes, and the list of substances. All actions and activities, both regulatory and nonregulatory, will be considered to help speed reductions. The Strategy also recommends a four-step process (see sidebar).

A Four-Step Process Towards Virtual Elimination

1. Gather information on generation, uses, and sources of pollutant;
2. Analyze current regulatory and non-regulatory programs and initiatives that manage or control the pollutant;
3. Develop cost-effective options and provide recommendations for increasing the pace and level of reductions; and
4. Recommend and implement options.

Both the United States and Canada have set milestones (or challenges) to achieve reductions through implementation of voluntary efforts and regulatory actions. One of these challenges is the commitment of these countries to work together to assess atmospheric inputs of persistent toxic substances, with the goal of evaluating and reporting impacts of long-range transport of these

Regulatory and Voluntary Options that may Prevent or Reduce Atmospheric Mercury Contamination

- ***Pollution Prevention.*** Mercury use in the United States has substantially declined during the past two decades. This trend parallels that of western Europe. Ebbing use implies that less mercury will be included in waste streams reaching incinerators and landfills, or released during production processes. The decline in use is attributable to both government bans and technological advances that create environmentally kinder products. Mercury was banned in pesticides (1972-1976 and 1993) and in paints (1990). The number of U.S. chloralkali facilities using a mercury cell process has declined from 25 during the early 1970s to 14 in recent years. Minnesota, Wisconsin, and New York restricted mercury content in batteries, and consequently, national use of mercury in batteries fell from 448 tons during 1988 to 10 tons during 1993. Mercury content in fluorescent bulbs has also declined.

- ***Recycling.*** A number of users of mercury are taking steps to boost recycling. Various municipal wastewater treatment plants are working with dental associations to encourage recovery of mercury in dentist offices so that mercury amalgam does not enter their treatment systems. Some manufacturers who rely on mercury in instruments are starting or considering take-back programs; EPA and states are working with several such firms to ensure that liability concerns do not preclude voluntary efforts. A number of hospitals are diverting mercury-containing wastes from incineration. Several states require that fluorescent bulbs be recycled (e.g., Florida, Minnesota), because such bulbs are commonly broken in or en route to landfills, allowing mercury vapors to escape to the atmosphere; while the quantity of mercury contained in a single bulb is minuscule, the number of discarded bulbs is great.

- ***Management.*** Pursuant to implementation of CAA, EPA has proposed performance standards for municipal and medical waste incinerators. A revised municipal waste combustor rule was promulgated in December 1995 (Federal Register, 60 FR 65387, December 19, 1995). There are about 4,000 medical and 180 municipal incinerators across the nation. These sectors are estimated to generate fully one-half of current national emissions of mercury, primarily through combustion of discarded products that contain mercury. When fully implemented by 2002 through state plans, these proposed standards hold promise of reducing mercury emissions from these two classes of incinerators by 95 and 80 percent, respectively. Performance standards are scheduled for other sectors that also emit mercury, including the Portland Cement industry; commercial and industrial boilers; primary lead smelters; the chloralkali sector; primary copper smelters; sewage sludge incinerators; and lime manufacturing.

- ***Global Production and Release of Mercury.*** During recent years, the U.S. government has held a significant stockpile of mercury on the world commodity market. To dispose of its holding, the Department of Defense has periodically auctioned mercury. From 1988 through 1993, sales totaled four million pounds. Sales were suspended in 1994, pending consideration of their environmental impacts. One positive environmental impact of sales is that they may forestall virgin production of mercury; the last mercury mines in the United States closed several years ago. Both western Europe and the United States have become substantial net exporters as their mercury consumption has fallen, whereas world use may be growing (Lawrence 1994). Because of diminished use of mercury, the United States is nearly meeting its entire need for mercury through recycling. Much mercury sold by the federal government has been exported. Foreign use can potentially have two undesirable consequences. It can add to mercury contamination of ocean fish resources. Also, foreign use and release, due to less stringent controls, can contribute to global atmospheric contamination which can travel for long distances, and directly contaminate United States surface waters. Suspension of government sales has positioned the United States to seek needed international cooperation in minimizing mercury releases on a global scale.

substances from worldwide sources by 1998. If long-range sources remain at this time, efforts within the existing international framework will be made to reduce releases of such pollutants. Specific coordinated efforts between EPA and Environment Canada to meet this challenge include:

- Coordinate efforts to identify sources in order to better define and coordinate emission control programs;

- Maintain atmospheric deposition monitoring stations;

- Continue research on atmospheric science of toxic pollutants to refine and improve existing source, receptor, and deposition models, as well as improve integration of existing air toxic monitoring networks and data management systems to track deposition of contaminants within the Great Lakes; and

- Conduct impact assessment of long-range transport of persistent toxic pollutants from worldwide sources by 1998.

Environment Canada will also demonstrate alternative processes to lessen emissions from five predominant sources by 1998, and complete inventories of ten selected air pollution sources to support environmental impact assessment of air toxics by 2000.

In addition, the Strategy includes several reduction goals or challenges for specific pollutants (Table IV-3). For the United States, the baseline from which these reductions will be measured is the most recent and appropriate inventories (e.g., mercury will be based on estimated emissions during the early 1990s). Canada plans to use their 1988 emissions inventory.

Another challenge is to increase remediation of priority sites with contaminated bottom sediments in the Great Lakes by 2005. The Binational Strategy is intended to fill in the gaps that exist where ongoing programs or emerging initiatives do not address toxic releases, to provide a context of basinwide goals for localized actions, and to provide "out of basin" support to programs such as LaMPs.

LAKEWIDE MANAGEMENT PLANS (LAMPS)

In Article VI, Annex 2 of the GLWQA, the U.S. and Canadian governments agreed to develop and implement LaMPs for each of the five Great Lakes. The purpose of the LaMPs is to document an approach to reducing input of critical pollutants to the Great Lakes and restoring and maintaining Great Lakes integrity. LaMPs are management tools designed to (1) integrate federal, state, provincial, and local programs to reduce loadings of toxic substances from both point and nonpoint sources; (2) assess whether these programs will ensure attainment of water quality standards and designated beneficial uses; and (3) recommend any media-specific program actions or enhancements to reduce toxic loadings in waters currently not attaining water quality standards and/or designated beneficial uses. Unlike the other four Lakes, Lake Michigan lies entirely within the boundaries of this country and therefore, the Lake Michigan LaMP has been developed solely by U.S. federal and state agencies with input from a public forum. The development of this program, as well as the deadlines established for the completion of the program, is mandated under section 118 of the CWA. In addition, as noted in Chapter I, section 112(m) of the CAA requires that EPA, in cooperation with NOAA, monitor the Great Lakes, investigate atmospheric deposition rates and pollutant sources, improve monitoring methods, and determine the relative contribution of atmospheric pollutants to the total pollution loadings to the Great Lakes and other Great Waters.

TABLE IV-3
Binational Pollutant Reduction Goals under the Draft Virtual Elimination Strategy[a]

Pollutants of Concern	United States Challenge	Canadian Challenge
Aldrin/Dieldrin, Chlordane, DDT, Toxaphene	Confirmation by 1997 that these pesticides are no longer used, generated, or released from sources that enter Great Lakes Basin. If long-range sources (outside North America) are confirmed, use existing international framework to reduce or phase out releases.	
Alkyl lead	Confirmation by 1997 that no longer used in automotive gasoline; reduce or replace in aviation fuel by 2005.	90 percent reduction in use, generation, or release by 2000.
PCBs	90 percent reduction nationally of high level PCBs (>500 ppm) in electrical equipment by 2005.	90 percent reduction nationally of high level PCBs (>50 ppm) that were once, or are currently, in service and accelerate destruction of stored high level PCB wastes by 2000.
Mercury	50 percent reduction nationally in deliberate use and in release from human activity sources by 2005.	90 percent reduction nationally in deliberate use from human activity sources by 2000.
Dioxins/Furans	75 percent reduction nationally in total releases from human activity sources by 2005.	90 percent reduction nationally in releases from human activity sources by 2000.
Benzo(a)pyrene, HCB	Reduction nationally in releases from human activity sources by 2005.	90 percent reduction nationally in releases from human activity sources by 2000.

[a] Based on the draft Binational Virtual Elimination Strategy (see August 19, 1996, Federal Register Notice).

A LaMP is a dynamic, action-oriented process encompassing a number of components. These include an evaluation of beneficial use impairments and pollutants contributing to those impairments; a summary of sources and loads of these critical pollutants; identification of ongoing prevention, control, and remediation actions, as well as additional efforts needed to reduce pollutant loads and to restore beneficial uses; and monitoring activities to evaluate the effectiveness of program actions. This approach for developing and implementing LaMPs is an evolutionary and iterative process for identifying and reducing critical pollutants. Public participation and cooperation with states and local governments is a key component to the LaMP development process.

LaMPs are in various stages of development for each of the Great Lakes states (see sidebar). Not all of the Lakes have LaMPs published in the Federal Register; however, commitments have been made by key stakeholders in the respective basins to pursue toxics reduction. Actions are being taken to achieve this goal. Each LaMP addresses a different list of critical pollutants, but some common ones are mercury, PCBs, hexachlorobenzene, dioxins, furans, chlordane, DDT and metabolites, and dieldrin.

Several activities have been initiated through LaMPs:

- In the Lake Michigan basin, agricultural "clean sweeps" to properly collect and dispose of unused pesticides have been conducted in Indiana, Michigan, and Wisconsin. Also, a variety of pollution prevention and technical assistance projects have taken place in Milwaukee, Chicago, and western Michigan.

- In most lake basins, tributary and atmospheric deposition monitoring is occurring. The Lake Michigan LaMP is utilizing the information generated from the Lake Michigan Mass Balance Study (described earlier in the section) to identify and reduce loadings.

- The Lake Superior LaMP was initiated as a component of the binational efforts to restore and protect Lake Superior. One of the goals of the LaMP is to achieve zero discharge and emission of persistent toxic pollutants through its Zero Discharge Demonstration Project. Another effort of the Lake Superior LaMP is an extensive pollution prevention outreach and education program developed for mercury. Among the activities are battery collection, energy efficiency, and product takeback programs. For example, Honeywell, Inc., the largest manufacturer of mercury thermostats used in regulating heating in the home, has instituted a thermostat takeback program in which the company recycles the mercury.

Current Status of LaMPs in the Great Lakes

Superior	Binational Program to Restore and Protect the Lake Superior Basin announced (1991) Proposed Stage 1 LaMP published in Federal Register (1994)
Michigan	LaMP published in Federal Register (1994)
Huron	LaMP not established
Erie	LaMP Management Committee formed (1994)
Ontario	Lake Ontario Toxics Management Plan (1989) LaMP Workplan signed (1993)

The LaMPs often provide the needed coordination and oversight for many such projects being implemented all over the Great Lakes Basin.

THE GREAT LAKES WATER QUALITY (GLWQ) GUIDANCE

Another notable toxics reduction effort was the recent GLWQ Guidance. It stems from the Great Lakes Water Quality Initiative, which began when the states of the Great Lakes Region recognized the unique feature of the Great Lakes ecosystem to accumulate persistent pollutants. The guidance is not only designed to address existing problems, but also to prevent emerging and potential problems posed by additional chemicals in the future, which may damage the overall health of the Great Lakes system. The guidance includes criteria for the protection of human life, wildlife, and aquatic life, taking into account the ability of many pollutants to biomagnify. Antidegradation requirements assure that current water quality will not be diminished. The guidance also outlines procedures to ensure consistent implementation and appropriate flexibility for long-term protection of the Great Lakes.

The GLWQ Guidance promotes the use of pollutant minimization plans to stop pollution before it enters the environment. Reducing pollution at its source is the most effective way of protecting public health and the environment, and is often more economical than cleaning up after a pollutant is released.

The water quality criteria in the GLWQ Guidance apply to all of the waters in the Great Lakes system, regardless of the source of pollution. Pollutants enter the Great Lakes from the air, stirred-up bottom sediments, urban and agricultural runoff, hazardous waste and Superfund sites, spills, and industrial and municipal wastewater discharges. Although the implementation procedures of the guidance apply mostly to industrial and municipal water discharges, a state may find it is more effective (or cost-effective) to improve water quality by reducing air emissions or cleaning up contaminated sediments, and that may choose not to impose additional requirements on wastewater dischargers.

ADDITIONAL EFFORTS TO ADDRESS TOXIC CONTAMINATION ISSUES

In addition to activities described above, many other pollutant-specific efforts provide significant information on atmospheric contamination in the Great Lakes. For example, EPA and the state of Wisconsin are currently analyzing the chemical form of mercury emitted from a coal-fired utility, a copper smelter, and a municipal incinerator. EPA has also promulgated standards for municipal waste combustors and proposed standards for medical waste incinerators which will, when implemented by 2002, provide about a 70 ton reduction in mercury emissions, or 35 percent of current total U.S. emissions, based on 1990 emissions estimates. Implementation of other Maximum Available Control Technology (MACT) standards offer the probability of further mercury emission reductions in the future.

For toxaphene, many issues remain about its sources and continuing presence in the Great Lakes. EPA held a research workshop on March 27-29, 1996, with scientists to assess these concerns (U.S. EPA 1996c). Based on the meeting, the following steps were identified to maintain progress toward resolving several questions concerning the presence and sources of toxaphene in the Great Lakes:

- Convene a meeting of international experts to review environmental data on fate and transport of polychlorinated bornanes;
- Inform public health authorities in states bordering Lakes Superior and Michigan about toxaphene levels in lake trout;
- Inform fish and game agencies in the same states and verify that toxaphene is no longer used;
- Analyze lake trout from northern Lake Michigan to identify recent levels;
- Analyze archived trout collected during the 1980s from Lake Superior and northern Lake Michigan to provide a more comprehensive picture of the contaminant trend in the lakes;
- Analyze sediment, air, and plant samples near several pulp and paper mills; and
- Monitor toxaphene levels in the atmosphere along the southern border of the United States to determine if foreign use of toxaphene may contribute to contamination in the United States.

Results from these efforts will provide a more firm technical basis upon which to explore the need for and extent of appropriate management actions.

Another major binational effort to broadly address Great Lakes issues was the 1994 State of the Lakes Ecosystem Conference (SOLEC) organized by the governments of the United States and Canada. Based largely on information gathered for during the conference, a report (and five background papers) was released in 1995 on the current condition of the Great Lakes (Environment Canada and U.S. EPA 1995). The report addresses the entire Great Lakes system in terms of ecological and human health, and the stressors which affect it. The six areas of discussion are human health; aquatic community health; aquatic habitat; toxic contaminants; nutrients; and the economy. Although the report does not describe or evaluate pollution control or natural resource management programs, it does focus on environmental conditions within the Great Lakes system. The SOLEC report indicates that there is considerable improvement in all the Great Lakes compared to 30 years ago, although serious losses in habitat of native plants and animals continues. Nutrient and toxic contaminant concentrations appear to be decreasing, although bioaccumulative pollutants still cause problems. The report presents a "mixed picture" of the current conditions of the Great Lakes and challenges managers and decision makers throughout the basin to obtain adequate information, deal with subtle effects of long-term exposure to low levels of toxic contaminants, protect biodiversity, restore habitat for native plants and animals, connect decisions with ecosystem results, and attain sustainability. The Great Waters program will benefit from many aspects of SOLEC, such as the information gathered on the impact of air pollutants to human health and ecological effects.

State of the Lakes Ecosystem Conference (SOLEC)

As part of the continuing response by the governments of the United States and Canada to the binational Great Lakes Water Agreement, SOLEC was initiated and held in October 1994. This conference is viewed as part of a process of sharing information needed to make well informed decisions that affect the ecosystem of the Great Lakes. It is attended by managers and other decision makers from the private sector, government and non-government environmental organizations. The next binational meeting of SOLEC will be held in Windsor, Ontario, on October 30-November 1, 1996. The nearshore areas, both aquatic and terrestrial, will be the focus of this second conference because they are most ecologically diverse and productive areas within each of the Lakes and are the parts of the system most affected by human activity.

Addressing Data Gaps/Future Needs

Considerable process has been made in the recent past in characterizing and reducing toxic pollution in the Great Lakes. The programs presented above and summarized in Table IV-4 provide an overview of some notable and recent activities by the United States, as well as Canada, to respond to concerns related to atmospheric pollution in the Great Lakes. Some of these measures include identifying emission sources; characterizing contamination from pollutants; developing and implementing voluntary and regulatory measures; and developing guidance for evaluating levels of risk of pollutant contamination. These programs have usually involved coordination among various federal, state, and/or local agencies. Though each program is designed to address specific goals, many of these programs coordinate their efforts to ensure that results are not duplicated. For example, the Binational Virtual Elimination Strategy was developed to achieve virtual elimination of persistent toxic pollutants in the Great Lakes, but it also supports and builds upon ongoing processes in the LaMPs, such as the Zero Discharge Demonstration Project through the Lake Superior LaMP. Furthermore, the Great Lakes Emissions Inventories will eventually provide

TABLE IV-4
Summary of Major Programs to Address Atmospheric Contamination in the Great Lakes

Data Collection/Research Projects	
Lake Michigan Enhanced Monitoring Program/Lake Michigan Mass Balance Study	Scientific base for future load reduction effort at all government levels. Mass balance study addresses CAA section 112(m) through coordinated effort to quantify and understand loadings, transport, and fate of selected HAPs. Also to provide a validated method to estimate loading for other waterbodies.
Integrated Atmospheric Deposition Network (IADN)	Binational monitoring network and research program to determine magnitude and trends of atmospheric deposition for the region.
Great Lakes Emissions Inventories	Inventory of sources and source category emissions in Great Lakes region, with a multi-state data base (RAPIDS).
Toxics Reduction Efforts	
Virtual Elimination Pilot Program/ Binational Strategy	Regulatory and non-regulatory efforts to encourage reduction in use and release of bioaccumulative pollutants in Great Lakes.
Lakewide Management Plan (LaMP)	Management tool to document approach to decrease pollutant input to each of the Great Lakes.
Great Lakes Water Quality (GLWQ) Guidance	Promotion of pollution minimization plans to stop pollution before it reaches the environment and consistent standards to protect human health, wildlife, and aquatic life.

information for determining whether the reduction goals set in the Binational Strategy have been met.

The Great Lakes programs described in this chapter also complement the Great Waters program in assessing and identifying the extent of atmospheric contamination of HAPs to the Great Lakes. Further coordinated progress is needed to improve knowledge and understanding of pollutant contamination, as well as to increase public awareness. In addition to the continuation of the current programs/activities, some high priority efforts for the Great Lakes basin are presented below.

- Improvement in research and monitoring techniques to reduce uncertainties in loading calculations (such as those for the IADN) and therefore, result in better estimates of atmospheric levels and deposition.

- After Lake Michigan Mass Balance data have been analyzed, application of results and modeling tools from the study to the development of a general mass balance model for other HAPs.

- Increase in efforts to identify local and long-range sources of Great Lakes pollutants through various source apportionment modeling and emissions inventories, such as in the RAPIDS data base.

- Continue to develop and implement strategies and recommendations to reduce use, generation, and release of pollutants affecting the Great Lakes, particularly through binational efforts such as the reduction challenges proposed under the Binational Virtual Elimination Strategy.

Once characterization and reduction of atmospheric deposition of toxic pollutants to the Great Lakes has improved, the results of these efforts, as well as the tools used, may be applied to other waterbodies, such as Lake Champlain.

IV.B Lake Champlain

Located around the northernmost borders of the states of New York and Vermont and the southern border of the province of Quebec, Lake Champlain is one of the largest freshwater lakes in the United States, with 435 square miles of surface water, over 70 islands, and 587 miles of shoreline (see Figure IV-6). It flows north from Whitehall, New York, almost 120 miles across the U.S.-Canadian border to its outlet at the Richelieu River in Quebec, where it joins the St. Lawrence River. Lake Champlain is unique because of its narrow width (12 miles at its widest point), great depth (over 400 feet in some parts), and large size of the watershed relative to the lake surface.

The Lake Champlain Basin, composed of the entire watershed or drainage area, spans from the Adirondack Mountains in the west to the Green Mountains in the east, and from the Taconic Mountains in the southeast to the St. Lawrence Valley in the north (Figure IV-6). The total area of the Basin is 8,234 square miles, of which 56 percent is in Vermont, 37 percent is in New York, and 7 percent is in Quebec. The Basin is characterized by an 18:1 ratio of watershed to lake surface area, indicating that the lake represents only about 5 percent of the total basin area. Approximately 89 percent of the Basin is categorized as forest and agricultural land.

By the end of 1994, approximately 645,000 people resided in the Lake Champlain Basin, with the population increasing each year by about 1.2 percent (LCBP 1994). Most of the region is classified as rural, with only Burlington, Vermont, recognized as a metropolitan area (1990 population of 52,000). The Lake Champlain Basin has traditionally had a rural resource-based economy, including agriculture, renewable natural resources (e.g., timber, fish, maple syrup), and non-renewable natural resources (e.g., iron ore, marble, gravel). In recent years, the economy has diversified into other activities, but is still dependent on the natural resources (see sidebar).

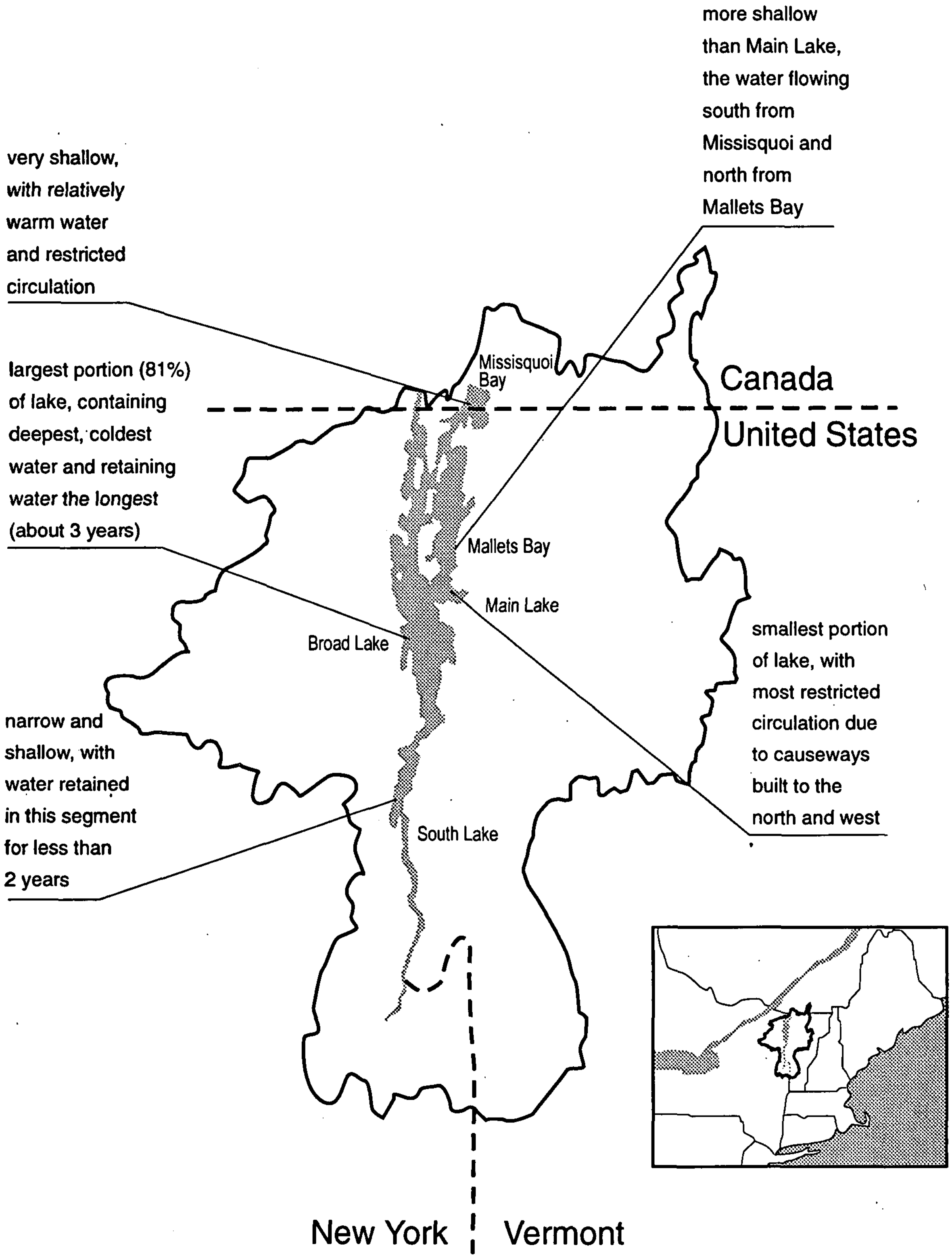
Economic Highlights of Lake Champlain Basin

- Twenty-five percent of the workforce is employed in natural resource-related activities (e.g., agriculture, mining, forestry).
- Resources of the Lake are a major reason why many Basin residents reside in this region.
- Recreational activities generate large revenues to the local area: $81 million per year from the fishing industry (in 1991), $50 million per year from bird and wildlife viewing (in 1990).
- Tourism represents a significant economic factor for the region, generating $2.2 billion in 1990, of which 40 percent were Lake related (e.g., marina, white-water rafting).

Sources: LCBP 1994; LCBP 1996.

Lake Champlain, unlike many other lakes which tend to be more evenly mixed, is made up of five distinct areas or lake segments, each with different physical and chemical characteristics (Figure IV-6). In these lake "divisions," pollutants may accumulate in shallow areas or bays where flushing and water circulation are reduced, and may be deposited at the mouths of rivers where runoff carrying sediment and other pollutants from the watershed is discharged. Human activities also concentrate in many of these areas of the lake, increasing the potential for contamination and exposure.

FIGURE IV-6
Lake Champlain Basin



very shallow, with relatively warm water and restricted circulation

more shallow than Main Lake, the water flowing south from Missisquoi and north from Mallets Bay

largest portion (81%) of lake, containing deepest, coldest water and retaining water the longest (about 3 years)

smallest portion of lake, with most restricted circulation due to causeways built to the north and west

narrow and shallow, with water retained in this segment for less than 2 years

An understanding of the hydrodynamics of Lake Champlain is essential to predicting how and where pollutants are transported, and where they will end up in the lake. A simplified three-dimensional hydrodynamic transport model, being developed with funding by the Lake Champlain Basin Program (LCBP), will be used as a management tool to determine potential effects of pollutant inputs and other changes to the waterbody (LCBP 1994).

Characterizing Toxic Contaminants in Lake Champlain

Levels of toxic contamination in Lake Champlain are low compared to the Great Lakes; however, concerns for protecting the public health still exist. Fish consumption advisories for two Great Waters pollutants (mercury and PCBs) are currently in effect in both New York and Vermont for fish from Lake Champlain (see Appendix B) (LCBP 1994). Testing of lake bottom sediment at the urbanized sites along the Lake show high levels of pollutants, indicating potential risks to aquatic life (see Section II.B). Because of these findings, as well as exceedances of water quality standards set by EPA, NOAA, and the province of Ontario, LCBP gives highest priority to these two pollutants.

The contribution of air deposition as a source of loadings for pollutants, such as metals and organic compounds, is of concern for Lake Champlain and the Basin, and has been the subject of recent studies. Although emissions of toxic pollutants within the Basin are considered low because of the few industries and utilities in the local area, high levels of pollutants may reach the Lake from more distant sources (LCBP 1994, 1996). Data have been limited regarding atmospheric sources, or the movement of pollutants from the atmosphere to the Lake directly or through the watershed.

Atmospheric Sources of Toxic Contamination in Lake Champlain

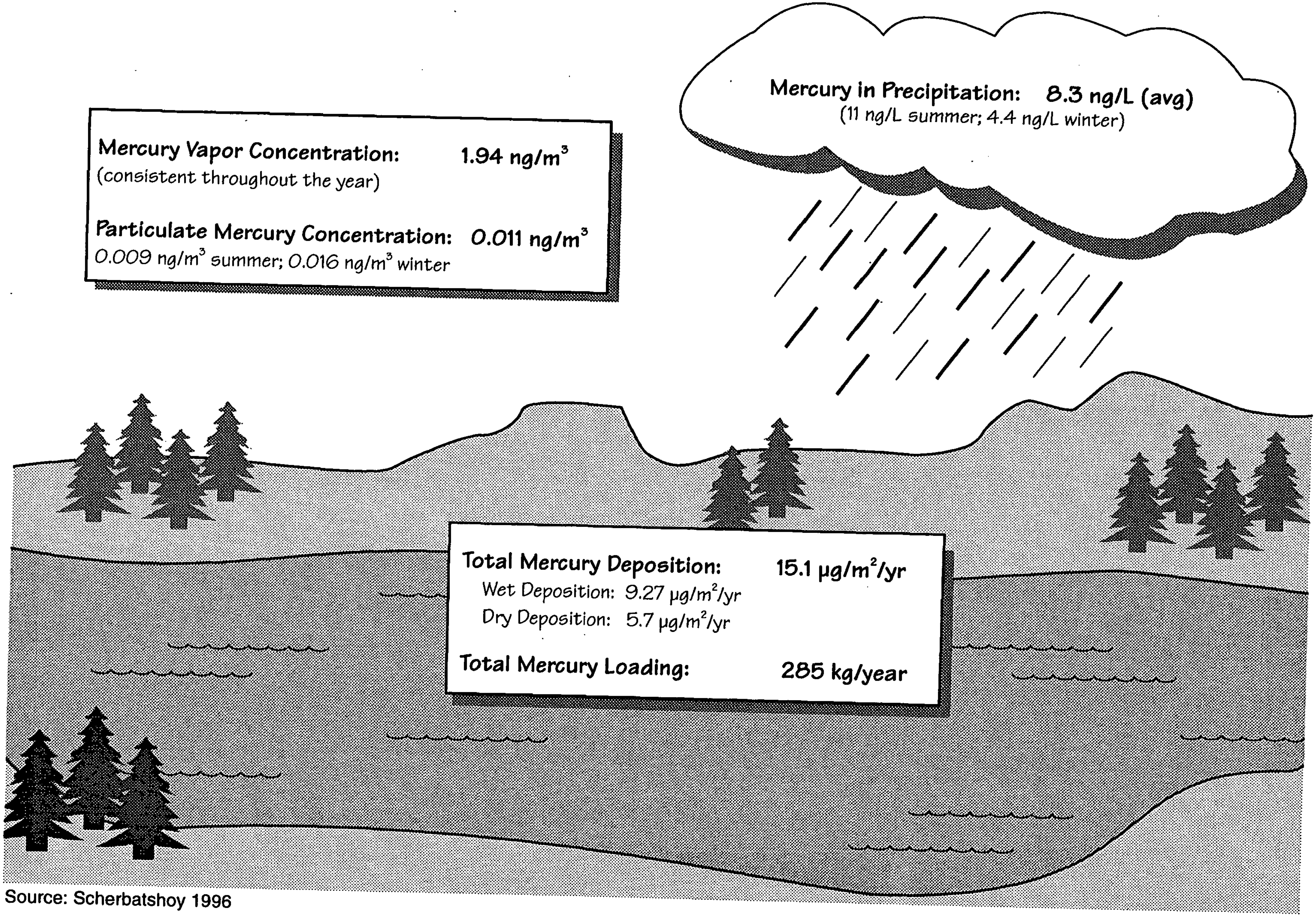
Local	Mobile emissions Residential energy consumption (e.g., wood burning) Waste incinerators
Regional/ Long-distance	Smelters (Quebec) Utilities (midwestern United States)

ATMOSPHERIC DEPOSITION OF MERCURY IN THE LAKE CHAMPLAIN BASIN

Mercury burden in Lake Champlain is evidenced by fish consumption advisories. Since direct discharges of mercury are quite limited, this has led to concerns that the atmosphere may be the major route of mercury to the lake. Recent studies supported by NOAA and EPA's Great Waters program have investigated the deposition of atmospheric mercury in the Lake Champlain Basin (Burke et al. 1995; Scherbatskoy et al. 1996). The air monitoring data collected from these efforts should provide an initial framework for a more comprehensive analysis of mercury cycling (i.e., deposition, transport, transformation, and accumulation) in the region, and beyond.

Atmospheric mercury concentration and deposition in Lake Champlain was investigated at a location just east of the Lake in Underhill, Vermont, between 1992 and 1994 (Scherbatskoy et al. 1996). The atmospheric concentration of mercury was measured as gaseous and particulate phases in the ambient air and in precipitation (snow, rain) (see sidebar). Findings on the mercury levels in Lake Champlain are presented below and in Figure IV-7:

FIGURE IV-7
Atmospheric Mercury in Lake Champlain Basin



Source: Scherbatshoy 1996

- *Atmospheric gaseous mercury concentration.* Average gaseous concentration for 1993 was 1.94 ng mercury/m^3 (comparable to other sites near the Great Lakes) staying relatively constant throughout the year (Table IV-5). Concentrations are typically two orders of magnitude (100x) higher than particulate mercury,

- *Average particulate phase mercury concentration.* Unlike the gaseous phase, particulate phase mercury in the air exhibited seasonal variability, with levels higher in winter than in summer (Figure IV-7). The source of the increased particulate mercury concentration in the winter has not been identified, but preliminary meteorological analysis suggests that a more regional influence is important in the transport of particulate phase mercury in the winter, due to higher average wind speeds and colder temperatures during this season. It also is suggested that colder temperatures in the winter may favor condensation of gas onto particles, increasing mercury particulate concentration in the atmosphere (Scherbatskoy et al. 1996). Furthermore, daily mercury concentrations did not fluctuate significantly, as would be expected for strong local sources.

Principal Phases of Atmospheric Mercury

***Gaseous (vapor) Phase Mercury*:** consists primarily of elemental mercury (Hg^o; predominant form in the atmosphere), although divalent mercury (Hg^{+2}) may also be present; Hg^{+2} is deposited more rapidly than elemental mercury.

***Particulate Phase Mercury*:** represented by fine particles that are not readily deposited from the atmosphere; bound Hg^{+2} is the predominant form and exists as the nucleus in dust particles.

***Mercury in precipitation*:** consists of mercury either as dissolved gas or bound to fine particles; primarily particulate Hg^{+2} which has been taken up by rain droplets.

TABLE IV-5
Comparison of Mean Total Atmospheric Mercury Concentrations[a]
(Gaseous and Particulate Phases and in Precipitation)

Location	Gaseous Phase (ng/m^3)	Particulate Phase (ng/m^3)	Precipitation (ng/L) (volume-weighted)
Lake Champlain	1.94±0.5	0.011±0.007	8.3±5.2
Northern Michigan[b]	NA	0.011	7.9±7.4
Southern Michigan[b]	NA	0.022	10.2±9.8
Northern Wisconsin[c]	1.57±0.4	0.02±0.02	10.5±4.8
Northern Wisconsin[d]	1.8±0.4	0.01±0.02	6.7±5.6

NA = Not Available
[a] Data are means for varying years and periods; different sampling methods for the studies.
[b] Hoyer et al. 1995; Keeler et al. 1995
[c] Fitzgerald et al. 1991
[d] Lamborg et al. 1995

Adapted from Scherbatskoy et al. (1996).

- *Average mercury concentration in precipitation.* Levels appeared to be typically higher in the summer than the winter. Partial data from 1994 suggest that the pattern observed in 1993 is typical for this area. Based on meteorological data, precipitation events with the highest mercury levels during the year were associated with regional transport from the south or west; transport from the west occurred only during the summer months (Burke et al. 1995). These mercury levels are low; studies have reported mercury concentration in precipitation generally less than 100 ng/L in areas not directly influenced by emission sources (U.S. EPA 1996a). The type of mercury in precipitation was not quantitatively determined (Scherbatskoy 1996); however, other studies of precipitation found mercury to be in the inorganic (Hg^{+2}) form, probably as mercuric chloride (Burke et al. 1995), with only 2-10 percent in the organic (methyl) form (Bloom and Watras 1989; Driscoll et al. 1994).

- *Yearly deposition of mercury.* Deposition to the entire Lake Champlain Basin, not just to surface water, was estimated from precipitation (wet deposition) and dry deposition data. Total annual mercury deposition was not very different from that observed at other U.S. locations (Table IV-6). Dry deposition was calculated by using the same monthly proportion of gas deposition to gas concentration reported in Lindberg et al. (1992). Deposition of particulate phase mercury was not a significant source of atmospheric mercury loadings when compared to deposition of mercury in precipitation and from the gas phase. The atmospheric mercury particulate concentration in the area (0.011 ng/m^3) was much lower than the gas concentration (1.9 ng/m^3); therefore, the estimate of mercury dry deposition was based solely on gaseous mercury (which may be an underestimate of deposition to some extent). As shown in Figure IV-7, the annual dry deposition is 5.7 µg mercury/m^2 (with levels higher in the summer than the winter). Mercury deposition in precipitation for 1993 was 9.27 µg/m^2, with deposition higher in the summer (1 µg/m^2/month) compared to winter (0.2 µg/m^2/month). The pattern for mercury deposition in precipitation is probably a result of higher mercury concentrations and higher amounts of rain during

TABLE IV-6
Comparison of Annual Mercury Deposition Estimates[a]

Location	Estimated Wet Deposition (µg/m^2/year)	Estimated Dry Deposition (µg/m^2/year)	Total Mercury Deposition (µg/m^2/year)
Lake Champlain basin[b]	9.27	5.7	15.1
Michigan[c]	5.5-13.0	NA	NA
Little Rock Lake, WI[d]	4.2 from rain 2.3 from snow	NA	24.5
Minnesota and Northern Wisconsin[e]	NA	NA	12.5

NA = not available

[a] Methods and assumptions for estimating these values varied with studies.
[b] Scherbatskoy et al. 1995
[c] Hoyer et al. 1995
[d] Fitzgerald et al. 1991
[e] Engstrom et al. 1994; Swain et al. 1992

the summer; however, insufficient data are available to determine the source of the increased mercury concentration in the summer.

Atmospheric mercury can also enter the lake from snowmelt, which adds higher concentrations of mercury to the rivers emptying into Lake Champlain. The total mercury concentration at three river sampling sites increased two- to six-fold during the peak snowmelt event as compared to the mean levels over the course of the year (Scherbatskoy et al. 1996).

Because the Lake Champlain basin has such a large ratio of watershed to lake surface area, it is important to understand the deposition and cycling of atmospheric pollutants by the largest categories of land use, forests and agriculture, prior to their entry into the lake in runoff. Unlike the Great Lakes watershed, which has relatively more lake surface area, Lake Champlain's water surface area makes up only 5 percent of the basin area. Therefore, it is important to understand mercury cycling through the terrestrial, atmospheric, and aquatic systems of the Lake Champlain Basin, as ongoing studies are beginning to address.

SOME RESEARCH EFFORTS RELEVANT TO ATMOSPHERIC DEPOSITION IN LAKE CHAMPLAIN

Other air deposition studies have been performed on the area surrounding Lake Champlain basin. These broad-scale studies have measured levels for several pollutants of concern to the Great Waters.

- A 1990/1991 air monitoring study measured the concentration of toxic metals at four sites in the Lake Champlain Basin (Whiteface Mountain, NY; Willisboro Bay, NY; Burlington, VT; and Underhill, VT) (LCBP 1994). Preliminary data revealed elevated levels of zinc in the air surrounding Burlington, possibly due to refuse incineration, tire wear, and industries. There were also periodic increases in arsenic levels at these and other sites across the Northeast. The source of these arsenic concentrations in the air is believed to be a smelter in Quebec (LCBP 1994). Mercury, lead, and cadmium compounds were also measured in this study; however, there has been no indication that the concentrations for these Great Waters pollutants of concern were elevated at the monitoring sites.

- The State of New York recently began a program for reducing toxics through a multi-media approach. The Multimedia Program for Pollution Prevention, when fully implemented, will integrate environmental protection programs across all "media," such as air, water, and land, to correct the problem with single media programs.

- The State of Vermont conducts a regulatory program for the sources of about 288 hazardous air contaminants (carcinogens, chronic systemic toxicants, and short-term irritants). Hazard Ambient Air Standards are established for each of these contaminants, with requirements imposed on new and existing sources (excludes fossil fuel combustion) emitting any of these contaminants in excess of a pre-determined "action level" for each pollutant.

- New York and Vermont established a Permit Exchange Agreement in accordance with the 1988 Memorandum of Understanding, in which both states are informed of permitted projects in the basin. It allows the affected public to participate in the comment and review process for the permits. Potential toxics sources subject to this agreement include air pollution sources within 50 miles of each state border that annually emit 50 tons of volatile organic compounds, sulfur dioxides, nitrogen oxides, carbon monoxide, or particulate matter, or 5 tons of lead, and/or are subject to Title V of the CAA.

Addressing Toxic Contamination Reduction in Lake Champlain

Over the past five years, the Lake Champlain Basin Program (LCBP) (see sidebar) has been the institutional framework for coordinating the development of a comprehensive pollution prevention, control and restoration plan for the future of Lake Champlain. The release of the draft plan in 1996 (LCBP 1996) was followed by public meetings that were held to allow interested parties to comment on the plan. Based on these meetings, modifications may occur to the plan before it is finalized. It is anticipated that the existing LCBP Steering Committee will be responsible for overseeing and implementing the objectives of the plan.

Lake Champlain Basin Program

The Lake Champlain Special Designation Act, sponsored by senators from Vermont and New York, was signed in 1990 and states that Lake Champlain is a resource of national significance. The intent of the Act is to create a comprehensive plan for protecting the future of Lake Champlain and its watershed. The coordination of the activities stated in the Act is the responsibility of the Lake Champlain Basin Program (LCBP), which is jointly administered by the U.S. EPA, the States of Vermont and New York, and the New England Interstate Water Pollution Control Commission. EPA was given $10 million in funds for 5 years to develop a comprehensive pollution prevention, control, and restoration plan for Lake Champlain; the draft plan was released in 1996. Other cooperating agencies include the U.S. Fish and Wildlife Service, U.S. Department of Agriculture, U.S. Geological Survey, NOAA, and National Park Service. Formal involvement of Quebec is through the Lake Champlain Steering Committee.

One major issue addressed in the plan was the prevention of pollution from toxic substances in order to protect public health and the Lake Champlain ecosystem. The implementation of the plan will require coordination along all levels of government, organizations, and individuals. The plan identified the following major technical and policy issues involved in determining the most appropriate and cost-effective actions to reduce toxic contamination of Lake Champlain.

- *Define scope of toxics reduction effort.* Research is needed to define the extent of problems related to toxic pollution in tributaries and in the air. Efforts should be made to improve source identification, with attention given to reducing both nonpoint and point sources through all media and remediating current sources of contamination throughout the Basin.

- *Focus efforts on Lake Champlain pollutants of concern and sites of concern.* LCBP established the List of Toxic Substances of Concern, identifying mercury and PCBs as Group 1 chemicals, meriting highest priority for management action (see sidebar on next page). These pollutants are found in the sediment, water, and biota at levels above appropriate standards or guidelines in Lake Champlain. Because of limited resources to study and monitor toxic substances, assessments should focus on specific sites where contamination is known.

- *Identify sources and quantify loads of toxic substances.* LCBP has begun to determine sources of toxic substances within Lake Champlain. Few "active" sources have been identified. Two major information gaps in this area include the extent of contamination from outside the basin, and the role of historical sources (e.g., discharged lead batteries released into the waterbody) and contaminated sediment.

- *Adopt strategy to prevent pollution.* Pollution prevention techniques (e.g., source reduction) represent the ideal approach for achieving reductions at the source of the problem, and eventually reducing pollutants in the lake. Vermont and New York have initiated programs to accomplish this goal; however, these programs are not yet integrated into the existing pollution control programs. In addition, greater public awareness is needed to adopt this approach.

Toxic Substances of Concern in Lake Champlain by Priority Group

Group 1: PCBs, mercury

Group 2: Arsenic, cadmium, chromium, dioxins/furans, lead, nickel, PAHs, silver, zinc

Group 3: Ammonia, persistent chlorinated pesticides, phthalates, chlorinated phenols, chlorine, copper

Group 4: Other contaminants known to be used or known to occur in the Basin (e.g., volatile organic compounds such as benzene, pesticides such as atrazine, strong acids and bases)

Source: LCBP 1996.

- *Establish firm and defensible toxic reduction goals.* The current chemical-by-chemical approach to managing pollution in Lake Champlain cannot account for the impact of cumulative or combined effects, and does not protect against unregulated (and potentially more toxic) chemicals entering the lake; therefore, the reduction strategy should be expanded to address toxic substances that do not yet exceed human health standards or cause measurable impacts within the basin.

In addition, as part of these efforts, relevant information gathered from other programs, such as those initiated in the Great Lakes, should be applied to Lake Champlain. As discussed in Section IV.A, the Great Lakes Water Quality Agreement Parties adopted a long-term goal for virtual elimination of sources of specific pollutants. This effort demonstrates the importance of binational cooperation to address concerns related to reducing toxic emissions in the Great Lakes, as well as in Lake Champlain.

IV.C Chesapeake Bay

The Chesapeake Bay, largest of the 130 estuaries in the United States, was the first in the nation to be targeted for restoration as an integrated watershed, airshed, and ecosystem. The 64,000 square mile drainage basin, shown in Figure IV-8, covers parts of six states (Delaware, Maryland, New York, Pennsylvania, Virginia, and West Virginia) and the District of Columbia, and includes more than 150 rivers and streams.

Stretching from Havre de Grace, Maryland, to Norfolk, Virginia, the Chesapeake Bay is 195 miles long, and ranges from 3 to 35 miles wide. The Bay has 56,000 miles of shoreline (more than the entire West Coast) and a surface area of over 4,600 square miles. Generally shaped like a shallow tray, the Bay's average depth, including all tidal tributaries, is only 20 feet, with a few deep troughs running along much of its length that average 60 to 70 feet, and reaching 174 feet at the deepest point. To visualize the relatively large watershed in contrast to the small Bay volume, imagine that the Bay's watershed is reduced to the size of this page; the relative size of the Chesapeake Bay would be a section 7.2 inches by 0.9 inches in the lower right hand corner, and the average depth of the Bay would be represented by one sixtieth the thickness of the paper.

Economic Highlights of Chesapeake Bay

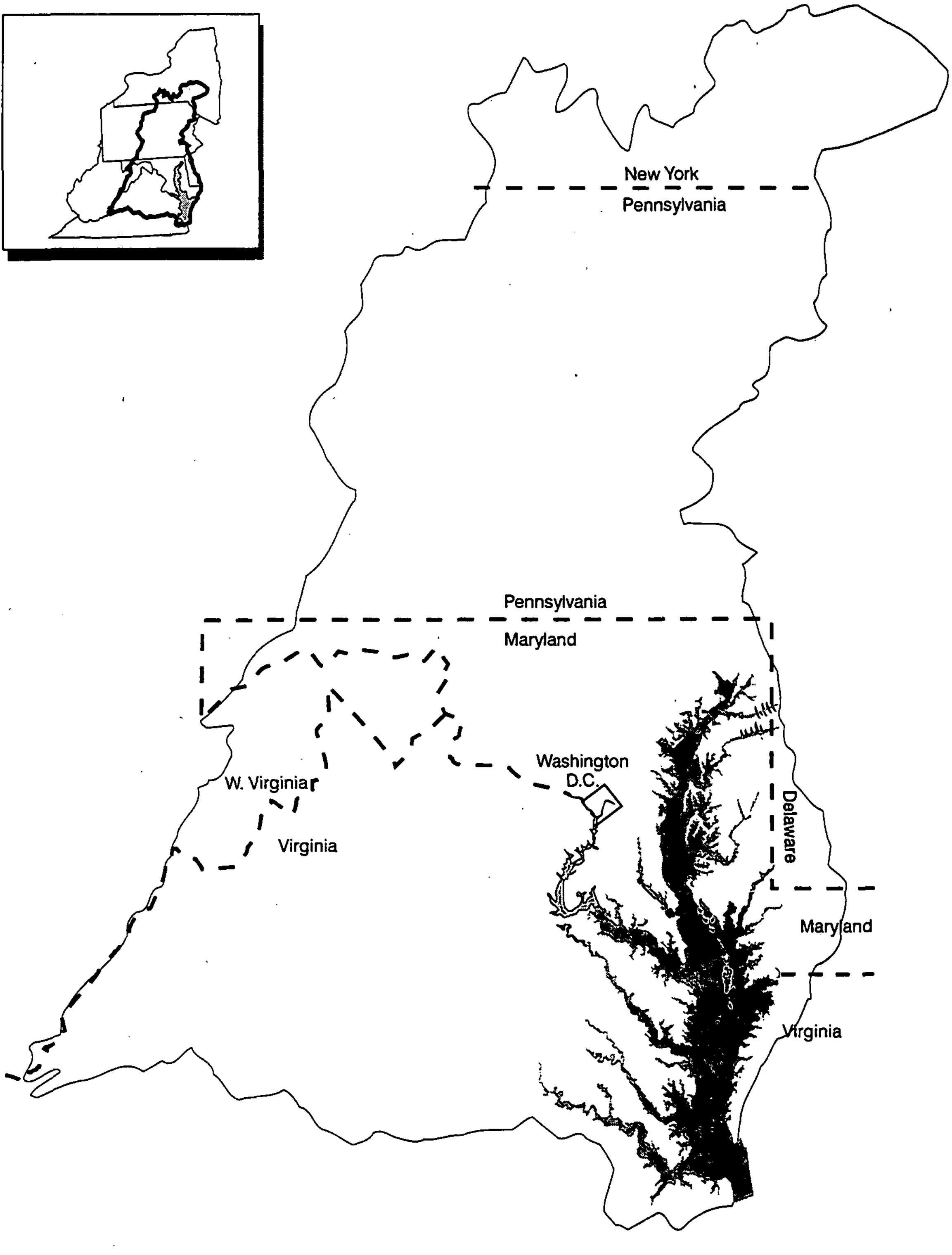
- In 1992, the dockside value of commercial shellfish and finfish harvests from Chesapeake Bay was close to $80 million.
- In 1993, more than 175,000 pleasure craft (e.g., sail boats) were registered in the Bay.
- Close to 1 million anglers in Maryland and Virginia made an estimated 600,000 fishing trips in 1991. Recreational fishing in these states is estimated at more than $1 billion annually.
- The Chesapeake is a key commercial waterway, and home to two of the nation's five major North Atlantic ports (Port of Baltimore, MD, and Hampton Roads Complex, VA). More than 90 million tons of cargo were shipped via the Bay in 1992.

Supporting 295 species of finfish, 45 species of shellfish, and 27,000 plant species, the Chesapeake Bay is a national treasure and an ecological gem. The Chesapeake Bay is also home to 29 species of waterfowl and is a major resting ground along the Atlantic Migratory Bird Flyway. Every year, one million waterfowl winter in the Bay's basin. In all, the Chesapeake is a commercial and recreational resource for more than 14 million Bay basin residents.

The remainder of Section IV.C presents:

- Background on the Chesapeake Bay Program;
- Information on atmospheric nitrogen deposition to the Chesapeake Bay, including discussion of the Bay program's nutrient reduction strategy, nitrogen loadings, modeling efforts, implications of nitrogen emission reductions, and addressing areas of uncertainty; and
- Information on atmospheric deposition of other pollutants of concern, including discussion of the Bay program's toxics strategy, pollutant concentrations and loadings, and addressing areas of uncertainty.

FIGURE IV-8
Chesapeake Bay Watershed



Chesapeake Bay Program

Now in its thirteenth year, the Chesapeake Bay Program (CBP) is a unique, regional partnership that has directed and coordinated Chesapeake Bay restoration since the signing of the historic 1983 Chesapeake Bay Agreement. Partners in the Chesapeake Bay Program include the State of Maryland, the Commonwealths of Virginia and Pennsylvania, the District of Columbia, the Chesapeake Bay Commission (representing the state legislatures), and EPA on behalf of the federal government.

In 1983, EPA identified an excess of the nutrients phosphorus and nitrogen, and the resulting accelerated eutrophication, as the primary reason for the decline in water quality in Chesapeake Bay (U.S. EPA 1983). Excess nutrients stimulate "blooms" of phytoplankton algae, which then sink to the bottom of the Chesapeake. In the bottom waters, decay of the phytoplankton consumes oxygen, which expands the area of anoxic bottom waters (i.e., dead waters largely devoid of oxygen and unable to support life). Blooms of algae also reduce light to underwater grasses, reducing an important habitat for juvenile fish and crabs.

The state of the Chesapeake restoration and protection effort was described in the latest *State of the Chesapeake Bay* report (CBP 1995b):

> "If the health of the Bay could be likened to that of a hospital patient, the doctor would report that the patient's vital signs, such as living resources, habitat, and water quality, are stabilized and the patient is out of intensive care. Some vital signs, such as striped bass and Bay grasses have improved dramatically, while a few, such as oysters, are in decline. Other vital signs are mixed but stable. Nutrients are being reduced, with phosphorus levels down considerably more than nitrogen levels and dissolved oxygen remains steady. Overall, the patient still suffers stress from an expanding population and changing land use, but it is on the road to recovery. Taken as a whole, the concentrated restoration and management effort begun ten years ago has produced tangible results--a state of the Bay that is better today than when we started..."

Using the watershed as the central focus, the Chesapeake Bay Agreement of 1983 recognized the historical decline of the Bay's living resources and recommended a cooperative approach among the federal and state governments within the watershed to address problems defined by the 1978-1983 Chesapeake Bay Research Program. The one-page agreement committed the signatories to work together to "fully address the extent, complexity, and sources of pollutants entering the Bay." The watershed approach of the state-federal partnership was chosen as the most practical method for implementing restoration efforts on both a local and regional scale.

Building on an expanded understanding of the Bay system and increasing experience with on-the-ground implementation within the cooperative basinwide partnership, a new Chesapeake Bay Agreement was signed in 1987 that set forth a comprehensive array of goals, objectives, and commitments to address living resources, water quality, growth, public information, and governance (Chesapeake Executive Council 1987). The centerpiece of the agreement was a commitment to achieve a 40 percent reduction of nitrogen and phosphorus entering Chesapeake Bay by the year 2000. This measurable goal added a specific direction to ongoing monitoring, modeling, and nutrient reduction implementation programs. Through the 1987 Bay Agreement, the signatories also committed to "quantify the impacts and identify the sources of atmospheric inputs on the Bay system." This seemingly minor commitment at the time set the stage for a decade-long path to formally address atmospheric deposition as an integral component of basinwide pollution reduction strategies and implementation actions.

Atmospheric Nitrogen Deposition to Chesapeake Bay

Although the Chesapeake Bay Agreement and the Nutrient Reduction Strategy (discussed below) focus on two main nutrients, nitrogen and phosphorus, this chapter focuses on nitrogen because, unlike phosphorus, atmospheric deposition is a significant pathway of concern for nitrogen loadings.

NUTRIENT REDUCTION STRATEGY

The Chesapeake Bay Agreement commits the signatories to reduce the "controllable" nutrient loads by 40 percent by the year 2000. Controllable loads are defined as the baseline year loads minus the loads delivered to the Bay under an all-forested watershed (i.e., a watershed providing only natural, uncontrollable sources of nitrogen) (Linker et al. 1996). In other words, controllable loads are defined as everything over and above the total phosphorus or total nitrogen loads that would have come from an entirely forested watershed in the States of Pennsylvania, Maryland, and Virginia, given existing rates of atmospheric deposition. In this definition, point source loads are considered entirely controllable; nitrogen emissions leading to atmospheric deposition were considered uncontrollable. Nonpoint sources may be controllable or uncontrollable.

Sources of Nitrogen Entering the Bay

Sources of the 170.8 million kilograms of nitrogen delivered to the Bay include:

- Point sources (23% or 39.3 million kg), such as sewage treatment plants;
- Nonpoint sources (68% or 116.1 million kg), such as runoff from agriculture and urban areas; and
- Atmospheric deposition directly to tidal waters (9% or 15.4 million kg).

A portion of the nonpoint source load from the watershed (26% of 116.1 million kg) can be attributed to atmospheric deposition on land with subsequent wash-off and transport to the Bay.

To measure the goal of reducing controllable nutrient loads by 40 percent, the Chesapeake Bay Program established a 1985 baseline of nutrient loads. The 1985 baseline load was defined using 1985 point source loads and a 1984-1987 average load for nonpoint sources. The Chesapeake Bay Program chose the average load of the 1984-1987 period as the base to be representative of nonpoint source loads for all tributaries, because river flow and associated nonpoint source loads may vary depending on rainfall. Table IV-7 presents the 1985 base load and 40 percent reduction target for the major tributary basins of the Bay (Figure IV-9 presents the locations of the tributary basins). After the year 2000, the tributary nutrient reduction targets (i.e., the 1985 base load minus the 40 percent reduction target) become nutrient caps that are not to be exceeded, as shown in Table IV-7.

In 1992, the basinwide reduction goal was reevaluated and allocated among the ten major tributary watershed basins. The state jurisdictions then developed comprehensive tributary nutrient reduction strategies through a process that directly involved the public within the individual watersheds. As part of the 1992 amendments to the Chesapeake Bay Agreement, the signatories committed "to incorporate in 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

Sources and Emissions of Nitrogen Compounds

Reactive nitrogen compounds, primarily oxides of nitrogen, are emitted to the atmosphere through both natural and anthropogenic pathways, overwhelmingly (95 percent) as nitric oxide (NO). Natural NO sources include emissions from soils and generation by lightning; dominant anthropogenic sources include emissions from automobiles, power plants, and biomass burning. On a global basis, anthropogenic and natural sources of oxidized nitrogen are approximately equal in strength. The dominant source of the reactive nitrogen oxides present in air over North America is high-temperature combustion (e.g., power plants, automobiles). Nitric oxide generated by combustion reacts quickly in the lower atmosphere, forming nitrogen dioxide (NO_2). The NO_2 is rapidly converted back to NO by ultraviolet light (photochemistry), then NO reacts again, resulting in a cycle driven by volatile organic compounds. From this photochemical cycle, ozone (O_3) is produced. The cycle is broken when NO_2 terminates into stable products, principally nitric acid vapor (HNO_3), and the NO gets used up. The ozone issue is therefore intimately related to the NO_x (defined as NO + NO_2) question. NO_2 slowly deposits to the underlying surface (too slowly to break the cycle), but nitric acid vapor (HNO_3) is easily and quickly deposited. HNO_3 is the source of most of the reactive nitrogen deposited to the earth's surface.

Reduced nitrogen compounds include ammonia (NH_3) and ammonium (NH_4^+). NH_3 is emitted into the atmosphere through both natural and anthropogenic pathways. Natural sources of NH_3 include microbial decomposition of organic nitrogen compounds in soils and ocean waters and volatilization from animal and human wastes. Anthropogenic sources include the manufacture and release of commercial and organic fertilizers during and after application and fossil fuel combustion. Human activities such as manure management and biomass burning exacerbate emissions from otherwise natural processes. The uncertainties apparent in these estimates are related to the intrinsically local nature of ammonia emissions, which make regional estimates highly difficult to construct. Ammonia is a highly reactive compound and has a short residence time in the atmosphere. It is primarily emitted at ground level and quickly deposits to the area near its source unless it reacts with other gaseous chemicals (e.g., SO_2, HNO_3) and is converted to ammonium (NH_4^+) aerosol (Langland 1992; Asman 1994). Ammonium can be transferred regionally as ammonium salts (e.g., ammonium nitrate NO_3NH_4, ammonium sulfate $(NH_4)_2SO_4$) and these salts are the primary contributor to ammonium concentrations measured in precipitation.

Organic nitrogen is not well understood. To date, studies of atmospheric nitrogen deposition have almost exclusively addressed dissolved inorganic nitrogen (reactive and reduced nitrogen). Because of the paucity of reliable measurements, the historical variability in analytical techniques and results, and the current lack of suitable and uniform analytical measurement techniques, only limited work has been reported on the deposition of organic nitrogen. In fact, only wet deposition of dissolved organic nitrogen (DON) has been addressed. This dearth of information is becoming widely recognized by the scientific community and is receiving increased attention. Recent reports (Milne and Zika 1993; Cornell et al. 1995; Scudlark et al. 1995) suggest that organic nitrogen is a significant fraction of the total nitrogen measured in precipitation. Various estimates for the relative flux of organic versus total nitrogen via wet deposition range from less than 10 percent to greater than 60 percent. These recent data suggest that the contribution of the unresolved organic fraction may significantly augment the atmospheric deposition of nitrogen to coastal waters. However, in addition to the lack of dry deposition data, there remain many conceptual questions related to source identification and the bioavailability of atmospheric organic nitrogen.

Current analytical techniques are unable to distinguish between specific organic nitrogen compounds measured in precipitation. Therefore, it is difficult to determine relative contributions of biogenic versus anthropogenic sources. Cornell et al. (1995) speculate from open ocean studies that a continental source is more likely given the higher concentrations in continental rains, the need for a relatively large sea-surface fractionation to sustain a marine source, and the isotope results. They also suggest that industrial combustion sources could contribute to DON formation via reactions of soot with NO_x and NH_3.

of nitrogen entering Chesapeake Bay and its tributaries" (Chesapeake Executive Council 1992).

Table IV-7
Chesapeake Bay Basin Nutrient Reduction and Loading Caps by Major Tributary
(in millions of pounds)

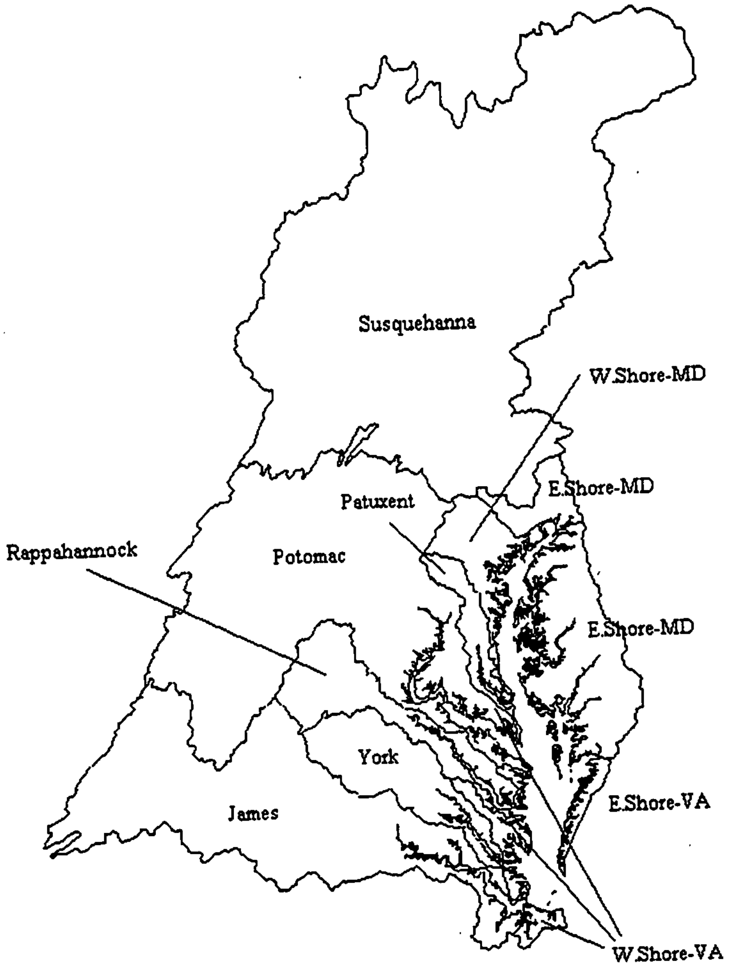
Major Tributary Watershed Basin	Nutrient	1985 Base Load	40% Target Reduction	Year 2000 Agreement Loading Cap
Susquehanna[a]	Nitrogen	116.80	18.30	98.50
	Phosphorus	5.95	2.22	3.73
Patuxent	Nitrogen	4.90	1.40	3.50
	Phosphorus	0.53	0.20	0.33
Potomac[b]	Nitrogen	68.70	18.70	50.0
	Phosphorus	5.32	1.71	3.61
Rappahannock	Nitrogen	8.30	2.60	5.70
	Phosphorus	0.86	0.32	0.54
York	Nitrogen	6.40	1.90	4.50
	Phosphorus	0.93	0.34	0.59
James[c]	Nitrogen	43.70	14.10	29.60
	Phosphorus	6.18	2.14	4.04
Western Shore MD	Nitrogen	26.50	9.70	16.80
	Phosphorus	1.70	0.67	1.03
Eastern Shore MD	Nitrogen	22.80	5.60	17.20
	Phosphorus	1.81	0.62	1.19
Western Shore VA	Nitrogen	4.20	1.20	3.00
	Phosphorus	0.50	0.19	0.31
Eastern Shore VA	Nitrogen	1.80	0.40	1.40
	Phosphorus	0.09	0.03	0.06

[a] Susquehanna loads include only loads from Pennsylvania and Maryland.
[b] Potomac loads include only loads from Pennsylvania, Maryland, Virginia, and the District of Columbia.
[c] James loads include only loads from Virginia.

Source: CBP 1992.

Any reductions in nitrogen loads brought about by the CAA Amendments are considered to be additional nutrient load reductions separate from the point and nonpoint source reductions identified in the tributary nutrient reduction strategies. CAA implementation is expected to reduce nitrogen loads in Chesapeake Bay beyond the tributary strategy reductions (CBP 1994a). However, these additional reductions may last only a short time; at some time after the year 2000, population growth and increased land development are expected to begin eroding the gains made by the CAA. This expected increase in nutrient loads may make it difficult to meet the caps on nutrient loads to the Bay. Indeed, it was acknowledged at the time the tributary strategies were developed that "achieving a 40 percent nutrient reduction goal, in at least some cases, challenges the limits of current point and nonpoint source control technologies" (Chesapeake Executive Council 1992). To maintain the restoration progress that is achieved by the year 2000, the technological limits of controls on reductions from point and nonpoint sources may have to be expanded to make further reductions in these areas economically attractive, or other sources of controllable nutrients may have to be considered to achieve cost effective ecosystem protection in the Bay.

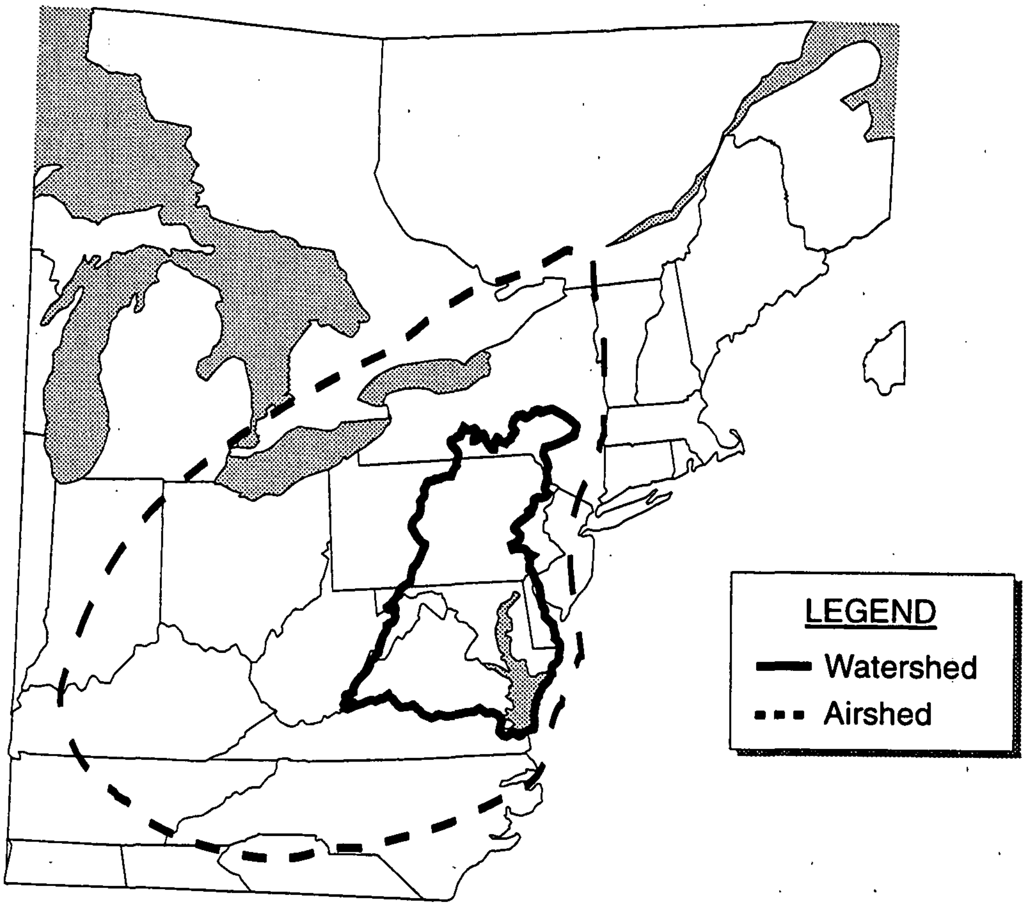
FIGURE IV-9
Major Tributary Basins of the Chesapeake Bay



MODELING AIR TO WATERSHED TRANSPORT: THE CHESAPEAKE BAY AIRSHED

A series of linked computer models have been developed by the Chesapeake Bay Program to simulate the transport of nitrogen from its emission sources to the Chesapeake Bay watershed and eventually into the tidal Bay waters. As a first step in establishing the air to tidal waters connection, the "airshed" of the Chesapeake Bay was defined. The boundaries of the airshed were defined as the contiguous areas whose sources "significantly" contributed (i.e., 75 percent) to nitrogen atmospheric deposition to the Bay and its surrounding watershed (Dennis 1996). These boundaries were delineated by running a series of scenarios on the Regional Acid Deposition Model (RADM) (described in more detail in Chapter III), using a predefined point of diminishing return (i.e., when a 50 percent reduction in emissions from large source regions would be expected to produce less than a 10 percent reduction in deposition onto the Bay watershed). The resulting 350,000 square mile airshed, shown in Figure IV-10, is about 5.5 times larger than the Bay's watershed and includes: all of Maryland, Virginia, Pennsylvania, Delaware, the District of Columbia, West Virginia, and Ohio; most of New York; half of New Jersey, North Carolina, and Kentucky; and parts of Tennessee, South Carolina, Michigan, Ontario, and Quebec (including Lakes Erie and Ontario).

FIGURE IV-10
Chesapeake Bay Airshed



According to the Bay airshed model, about 25 percent of the nitrogen that is deposited on the Bay and its surrounding watershed originates from sources within the Bay watershed. Sources located within the jurisdictions of the Bay Agreement signatories of Maryland, Virginia, Pennsylvania, and the District of Columbia (including those sources that are within the state boundaries but outside of the Bay watershed) contribute about 40 percent of the nitrogen that deposits on the Bay and its watershed (Dennis 1996).

As defined, the Bay airshed, which accounts for 30 percent of eastern U.S. and Canadian nitrogen emissions, accounts for 75 percent of the atmospheric nitrogen deposited onto the Bay and its watershed. The remaining 25 percent of the deposition originates from emission sources outside the defined airshed (Dennis 1996). Therefore, the Chesapeake Bay airshed as defined here is smaller than the actual areas of the United States and Canada that contribute to nitrogen deposition to the Bay watershed. A still unresolved portion of the airshed is the portion that contributes to atmospheric deposition to offshore ocean waters which, in turn, contributes to the influx of nitrogen from coastal waters into the southern Chesapeake Bay (CBP 1994b).

Through the Bay airshed model, researchers investigated the types of sources of nitrogen emissions and the contribution of these sources to nitrogen loads to the Bay. From the Bay watershed States of Maryland, Virginia, Pennsylvania, West Virginia, Delaware, and New York, and the District of Columbia, utility sources (e.g., power plants) contribute 37 percent of the total NO_x emissions, mobile sources (e.g., cars, trucks) contribute 35 percent, other area sources (e.g., ships, boats, lawn equipment) contribute 21 percent, and other point sources (e.g., industries) contribute 6 percent (Dennis 1996). All told, these emissions indicate that the contributions from utility and mobile sources to Chesapeake watershed and estuary deposition are roughly equal and make up the majority of emission sources. However, the patterns of nitrogen deposition from the two sources, which were simulated using RADM, are different. The model simulations suggest that utilities contribute a majority of the nitrogen that deposits on the western side of the Bay watershed and that deposition from utility emissions show a decreasing trend from the western to eastern portion of the watershed (see Figure IV-11). These simulations further suggest that mobile sources, associated with emissions from the Boston to Washington, D.C., metropolitan areas, contribute a majority of the nitrogen that deposits along the Delmarva Peninsula, the Chesapeake Bay itself, and the lower portions of the western shore tidal tributaries (see Figure IV-12). In contrast to utility sources, the simulated deposition from mobile sources shows a decreasing trend from the eastern to western portion of the basin. Model scenarios simulating the effects of a uniform 50 percent reduction in NO_x emissions from utilities alone and then from mobile sources alone show the same west to east, or east to west, gradients respectively (Dennis 1996).

FIGURE IV-11
Total Nitrogen Deposition from Utility Sources
(percent contribution of 1990 Base Case)

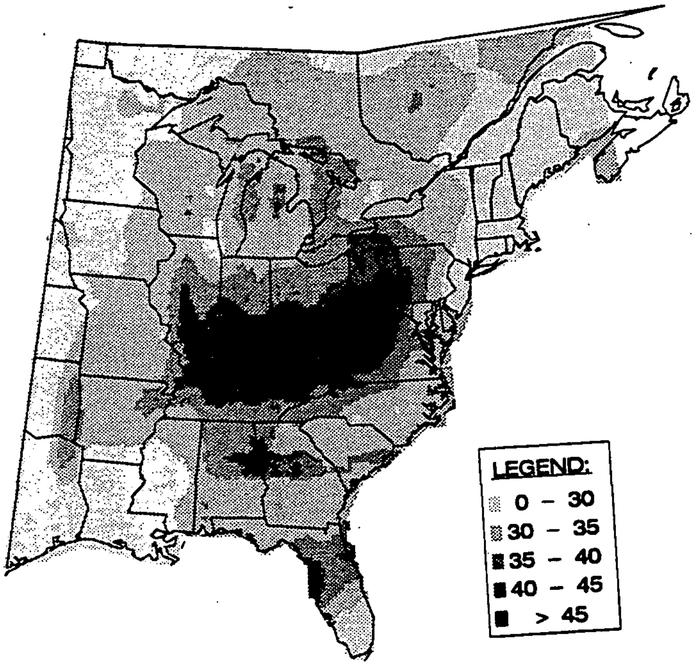
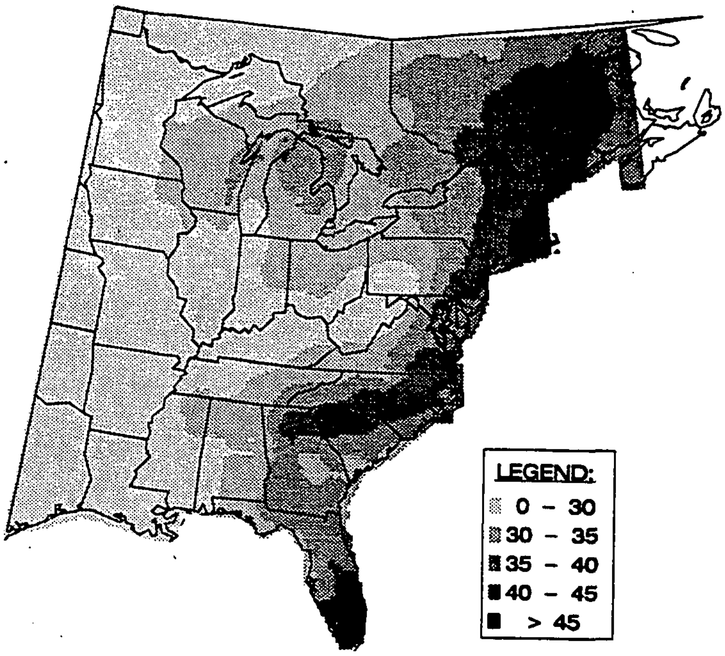


FIGURE IV-12
Total Nitrogen Deposition from Mobile Sources*
(percent contribution of 1990 Base Case)



ATMOSPHERIC NITROGEN LOADINGS TO CHESAPEAKE BAY

Atmospheric nitrogen loads from the airshed are transported to the Chesapeake Bay by three routes: direct deposition to the Bay tidal waters; indirect deposition to the watershed with subsequent runoff and river transport to the Bay; and deposition to adjacent offshore coastal waters with subsequent transport to the Bay through coastal currents, which is the least well understood route. The first two processes, direct and indirect deposition to the Bay, are discussed below, as are some estimates of total loadings to the Bay using both a mass balance approach and computer models.

Direct Loadings. The first estimates of atmospheric deposition to the tidal waters of Chesapeake Bay were made through spatial extrapolation of the National Atmospheric Deposition Program (NADP) sites in the Chesapeake watershed (Cerco and Cole 1994). The NADP is a long-term nationwide monitoring program that was started in the 1970s. Based on the annual loads reported by NADP, and an assumption that dry deposition of nitrate is equal to the long-term average of wet deposition of nitrate (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988), atmospheric deposition to the tidal waters of the Chesapeake were estimated as 14.1 million pounds of nitrate, 4.0 million pounds of ammonia deposition, and 15.0 million pounds of organic nitrogen. Studies have explored the idea that atmospheric deposition may contribute a significant proportion of phytoplankton nitrogen demands in coastal areas (Paerl 1985; Paerl 1988; Paerl et al. 1990); phytoplankton require nitrogen, both new and recycled, for growth. Fogel and Paerl (1991), for example, have estimated that 20 to 50 percent of annual new nitrogen demands for phytoplankton in Albemarle-Pamlico Sound, NC, may be supplied by direct atmospheric deposition to the water surface (wet and dry).

The NADP monitoring data provide an initial estimate of atmospheric deposition to the tidal Bay; however, it is not currently known if the over-land measurements of wet deposition accurately represent over-water wet deposition. To investigate this question, a daily precipitation chemistry site was established on Smith Island, Maryland, in late 1995. This site is providing the first time-series measurements of over-water deposition collected on the east coast. Estimates of wet deposition directly to the Chesapeake Bay tidal surface waters range from 7.3 to 9.3 million pounds of nitrate per year (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988).

Although the dry deposition to surface water loading rates of nitrogen compounds have been estimated for most nitrogen species over open ocean (Galloway 1985; Duce et al. 1991), these rates may not apply to coastal areas because of the different meteorological processes involved. Through the use of instrumented Chesapeake Bay Observing System (CBOS) buoys owned by the University of Maryland, estimates of nitrogen (HNO_3, NO_2, NH_4) dry deposition rates to the Bay tidal surface waters have been developed (Valigura 1995). These estimates corroborate those given by other investigators to some extent, but still cover a wide range of values, from 1.6 to 4.9 million pounds of nitrate per year. From this data set, calculations were performed to determine the effect of atmospheric dry deposition on phytoplankton dynamics. This analysis demonstrated that dry deposited nitrogen may provide 10 percent of the annual "new nitrogen" demands in Chesapeake Bay, and that individual events could supply up to 75 percent of the new demands for periods of several days (Malone 1992; Owens et al. 1992).

Indirect Loadings. Quantification of indirect loadings to Chesapeake Bay, which refers to the atmospheric deposition of nitrogen to the terrestrial watershed and subsequent transport of the nitrogen from the terrestrial watershed to Bay surface waters, is an important area of uncertainty in estimating atmospheric deposition to the Bay. NADP monitoring data provide an initial estimate of the atmospheric deposition to the Chesapeake Bay watershed. Generally, higher deposition levels are found in the northern portions of the basin. In fact, some of the highest readings for atmospheric deposition of nitrate in the NADP monitoring network come from the northern sections of the Chesapeake basin. Greatest uncertainty is in dry deposition of nitrogen, which is not routinely measured by NADP.

The amount of atmospheric nitrogen transferred to surface waters within a given watershed depends on land use, total nitrogen loading from atmospheric, fertilizer, animal waste, and biosolid sources, the amount of soil nitrogen, characteristics of the soil, site rainfall and temperature, the elevation and slope of the land, and the type, age, and health of the vegetative cover. These characteristics vary independently, making it difficult to determine the fate of atmospherically-deposited nitrogen over any area of significant size. However, several classification schemes for forested sites have been developed to evaluate a site's potential to retain and leach nitrogen (Melillo et al. 1989; Johnson and Lindberg 1992; Stoddard 1994).

Stoddard (1994) in particular has helped organize thinking about nitrogen retention by using a scheme to classify forest systems with regards to stages of nitrogen saturation (i.e., the extent to which the system is saturated with nitrogen; the more saturated a system, the more likely to leach nitrogen). The classifications range from Stage 0, where forest nitrogen transformations are dominated by plant and microbial assimilation (uptake) with little or no NO_3 export from the watershed during the growing season, to Stage 3, where nitrogen deposition is well in excess of assimilation and has reduced plant and microbial assimilation capacities resulting in greater export of atmospheric nitrogen as well as losses from mineralization of soil organic nitrogen). Study sites in the southern portions of the Chesapeake Bay watershed generally fall into the low nitrate export classification (Stages 0-1), while the northern portions have generally high to medium export classifications (Stages 1-2).

Total Loadings Estimates - The Mass Balance Approach. The role of atmospheric transport as an important path for nitrogen deposition to estuarine areas was first publicized in 1988 (Fisher et al. 1988). Based on a mass balance analysis using 1984 hydrology data, the authors estimated that one-third of the nitrogen entering the Chesapeake Bay is deposited from the atmosphere. Several subsequent efforts (Fisher and Oppenheimer 1991; Hinga et al. 1991; Tyler 1988) to quantify atmospheric nitrogen loadings to Chesapeake Bay produced "best-estimate loadings" ranging from 25 percent (Fisher and Oppenheimer 1991) to about 33 percent (Hinga et al. 1991) of the total anthropogenic loads to the Chesapeake.

The approach taken in these mass balance studies can be divided into two components: (1) estimating wet and dry deposition and (2) estimating nitrogen retention. A central difficulty in mass balance studies is the use of average land use values of nitrogen retention. Nitrogen retention assumptions used in three of the Chesapeake Bay studies are presented in Table IV-8.

TABLE IV-8
Nitrogen Retention Assumptions Used in Chesapeake Bay Loading Studies (in percentage of nitrogen loading)

Land Use	Tyler 1988	Fisher and Oppenheimer 1991[a]	Hinga et al. 1991[a]
Forest	95.2-100.0	80.0 (51.0-100.0)	80.0 (25.0-95.0)
Pasture	93.7-99.96	70.0 (51.0-90.0)	80.0 (25.0-95.0)
Cropland	76.0-99.97	70.0	60.0 (45.0-75.0)
Residential	62.0-95.3	35.0 (0.0-70.0)	25.0 (10.0-50.0)

[a] Numbers in parentheses indicate range tested.

Assembling an adequate understanding of long-term behavior when the processes involved are fundamentally episodic is another major challenge of contemporary models. Some studies indicate that the majority of the atmospheric wet deposition occurs during a few episodes (Dana and Slinn 1988; Fowler and Cape 1984), such that the wet-deposited nitrogen (as well as previously dry-deposited nitrogen) is deposited directly to, or flows quickly into, the surface waters without intermediate reduction in concentration. Despite these difficulties, mass balance studies provide a good first-order estimate of nitrogen loading to Chesapeake Bay.

Experimental manipulation at the watershed scale are being conducted at a few U.S. locations (Kahl and Norton 1993; Norton et al. 1994). Work from these sites is providing information on the cycling of nitrogen in forested catchments and is fully supportive of the conclusion that atmospheric deposition contributes to nitrogen loading of coastal waters through the export of atmospherically-derived nitrogen. Results of these long-term experiments are just beginning to be published. An example is the Bear Brook watershed in Maine. Divided into treatment and control catchments, the treated catchment received increased nitrogen loading in the form of labeled ammonia. The treated watershed response was an immediate increase in stream nitrate export (Norton et al. 1994, 1995).

Total Loadings Estimates - Chesapeake Watershed and Estuary Models. The Chesapeake Watershed Model (discussed in more detail below) is one approach to disaggregating the separate components of terrestrial and river nitrogen dynamics in the basin, along with the temporal effects of high loading during rainfall events. The most recent estimate of atmospheric nitrogen loads for both direct and indirect deposition is 27 percent of the annual nitrogen load delivered to the Chesapeake Bay. This estimate was developed using the 1992 Watershed Model (Linker et al. 1993) and is consistent with the range reported by Chesapeake Bay mass balance studies (25-33 percent).

Inherent Uncertainties in Mass Balance Approach

"It would not be difficult to make the [mass balance] calculations appear more elegant by subdividing the watersheds into more land use types, using a detailed data base of land uses, assembling more detailed lists of point source and agricultural inputs, and using some technique for contouring deposition over the watershed. None of these approaches are likely to make better calculations. More precise and reliable estimates of the magnitudes of inputs of atmospherically-deposited nitrogen to coastal waters will require significant advances in the understanding of many processes responsible for the behavior of nitrogen in terrestrial ecosystems and in rivers and streams."

Source: Hinga et al. 1991.

To estimate wet deposition, the Chesapeake Bay Program combined output from a regression model developed from National Atmospheric Deposition Program weekly and daily precipitation chemistry measurements with data from the NOAA rainfall network. This approach yields daily estimates of rainfall to 74 sub-basins of the Chesapeake Bay watershed. Dry deposition was assumed to be equal to wet deposition for over-land areas and 44 percent of wet deposition for over-water areas. Indirect atmospheric loadings from the over-land portion of the watershed were estimated using the Chesapeake Bay Program Watershed Model.

Comparison of Nitrogen Loadings Between Coastal Waters. Approximately 40 studies around the world have addressed at least one aspect of atmospheric nitrogen loadings, the majority of which were published after 1990. However, the measurement and modeling techniques used vary considerably between individual studies, making intercomparisons difficult. Table IV-9 presents a summary of selected studies performed along the eastern and Gulf coasts of the United States that are comparable in broad terms (the studies are ordered by Chesapeake Bay and tributaries, followed by the remaining coastal areas in order of decreasing tidal water area). The two criteria for selecting these studies, based on the approach and information content rather than scientific merit, were that the study results were either published in a credible peer-reviewed journal or advocated by a major management organization (e.g., an EPA National Estuary Program). These studies can be divided into two groups: those that considered both direct and indirect nitrogen loads and those that considered only direct loads. Data confirm the common belief that the amount of atmospheric nitrogen input is related to the size of the waterbody and its watershed (see Table IV-9).

The most recent estimate of a 27 percent contribution of atmospheric deposition to total nitrogen loadings to the Chesapeake Bay falls within the range reported for other estuaries. The studies shown in Table IV-9 for the several other major eastern and Gulf coast estuaries have provided atmospheric nitrogen loading estimates that range from 2 to 80

TABLE IV-9
Current Estimates of Atmospheric Nitrogen Loading to Selected Coastal Waters (in millions of kg)

Coastal Water	Surface Area (km^2)		Deposition to Watershed	Direct Deposition to Tidal Waters	Indirect Atmos. Load From Watershed	Total Atmospheric Load	Total Load From All Sources	% Load from Atmosphere	Reference[a]
	Watershed	Tidal Waters							
Chesapeake Bay (MD/VA)	165,886	11,400	175	16	29	45	170	27	5
Rhode River (MD)	33	4.9	—	0.005	—	0.005	0.012	40	6
Choptank River (MD)	1,779	361	—	0.57	—	0.57	1.54	37	11
Patuxent River (MD)	2,393	137	—	0.22	—	0.22	12.6	13	11
Potomac River (MD)	29,940	1,210	—	1.9	—	1.9	35.5	5	11
New York Bight (NY/NJ)	50,107	38,900	69	54	8	62	164	38	1
Albemarle-Pamlico Sound (NC)	59,197	7,754	~39	3.3	6.7	10	23	44	4
Long Island Sound (NY/CT)	43,481	4,820	43	5	6	11	60	18	3
Massachusetts Bays (MA)	—	3,700	—	1.6-6	—	1.6-6	22-30	5-27	15
Delaware Bay (DE)	36,905	1,846	53	3	5	8	54	15	2
Tampa Bay (FL)	6,216	1,031	—	1.1	—	1.1	3.8	28	14
Guadalupe Estuary (TX)	—	551	—	0.31	—	0.31	4.2-15.9	2-8	13
Narragansett Bay (RI)	4,708	328	4.2	0.3	0.3	0.6	5	12	1
			—	0.4	—	0.4	9	4	12
Newport River Coastal Waters (NC)	340	225-1,600	—	0.095-0.68	—	0.095-0.68	0.27-0.85	35-80	4
Sarasota Bay (FL)	524	135	—	0.16	—	0.16	0.6	26	10
Delaware Inland Bays (DE)	800	83	—	0.28	—	0.28	1.3	21	9
Flanders Bay (NY)	83	39	—	0.027	—	0.027	0.36	7	8
Waquiot Bay (MA)	~70	~8	0.062	—	0.0065	0.0065	0.022	29	7

[a] (1) Hinga et al. 1991; (2) Scudlark and Church 1992; (3) LIS 1996; (4) Paerl and Fogel 1994; (5) CBP 1996; (6) Correll and Ford 1982; (7) Valiela et al. 1996; (8) Peconic Bay NEP; (9) Delaware Bays NEP; (10) Sarasota Bay NEP; (11) Boynton et al. 1995; (12) Nixon et al. 1995; (13) Brock et al 1995; (14) Tampa Bay NEP; (15) Mass. Bays NEP.

percent of the total nitrogen load. (See Section IV.D for more discussion of atmospheric deposition to coastal waters.)

MODELING THE CHESAPEAKE BAY'S WATERSHED AND ESTUARY

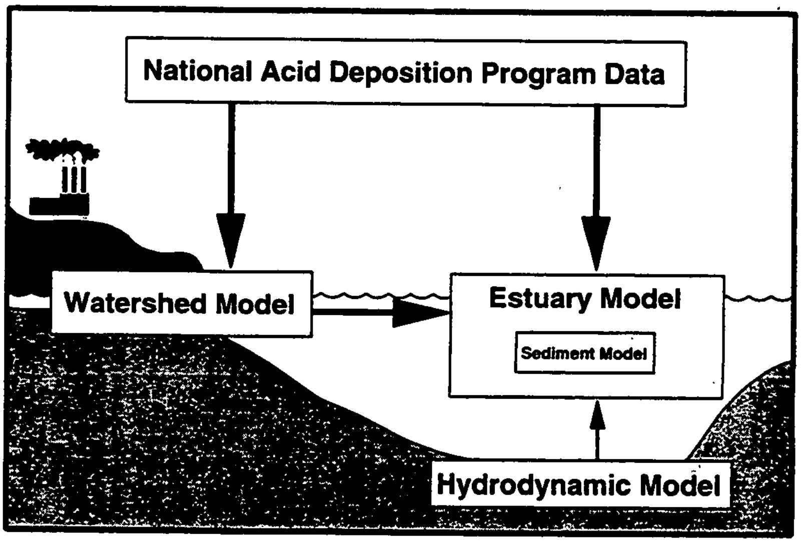
Water quality models of the Chesapeake Bay's watershed and estuary have been in use since the mid-1980s (Hartigan 1983; CBP 1987; Donigian et al. 1991; Thomann et al. 1994). The 1987 Bay Agreement's 40 percent controllable nutrient goal was based in part on findings from these models.

The first model of the Bay watershed was completed in 1982 and provided a basin-by-basin assessment of the relative importance of point source and nonpoint source controls of nutrients (NVPDC 1983). Subsequent refinements of the Watershed Model established the importance of animal waste management in the watershed (Donigian et al. 1991), the delivery to the Bay of atmospheric deposition loads from the watershed (Donigian et al. 1994), and the development of tributary allocation loads of nitrogen and phosphorus to achieve the 40 percent nutrient reduction goal (Thomann et al. 1994).

In a parallel effort, the first model of the Chesapeake estuary was completed in 1987 to evaluate the impact of nutrient reduction scenarios on the Bay's dissolved oxygen concentrations (CBP 1987). Results from the steady-state Estuary Model indicated that a 40 percent reduction in nutrient loads would significantly reduce anoxia (dissolved oxygen concentrations less than 1 mg/L) in the Bay mainstem during average summer (June-September) conditions (CBP 1988). The 40 percent controllable nutrient reduction goal, under the 1987 Bay Agreement, was based in large part on these findings.

A reevaluation of the Bay's nutrient reduction goal and a review of the progress made in reducing nutrients was scheduled for 1992. In advance of this reevaluation, researchers began refining and integrating the Watershed and Estuary models (Figure IV-13). For example, the Watershed Model was updated with greater detail of agricultural and atmospheric sources and was linked to the Estuary Model (Donigian et al. 1994). Providing a predictive framework for determining nutrient loads delivered to the tidal Bay under different source reduction scenarios, the Watershed Model simulated delivered nutrient loads with changes in land use practices and levels of wastewater treatment (Thomann et al. 1994). The Estuary Model was upgraded to add a sediment processes model and a hydrodynamic model, and was linked with the Watershed Model to accept Watershed Model nutrient loads as data input (Cerco and Cole 1994; DiToro and Fitzpatrick 1993; Johnson et al. 1991).

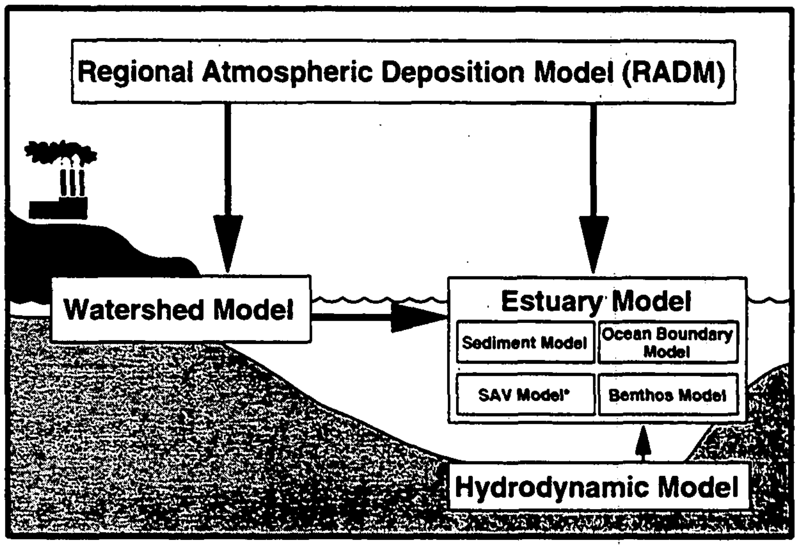
FIGURE IV-13
Watershed and Estuary Model Integration



The integrated Watershed and Estuary Model of the Chesapeake Bay was used to estimate water quality improvements that would be realized upon reaching the Bay Agreement goal of reducing controllable nutrients by 40 percent. Through the application of these models, the Bay Program partners established the Bay Agreement tributary nutrient allocations of nitrogen and phosphorus to be achieved by the year 2000 and maintained thereafter.

While this initial integrated model (Figure IV-13) could simulate the effects of atmospheric nitrogen deposition on water quality, it could not project the ultimate influence of changes in total nitrogen loadings (i.e., including loadings other than from the air) to the Bay. To provide for this predictive capacity, the Chesapeake Bay Program recently configured the Bay Watershed Model to accept daily atmospheric loadings by land use category (i.e., forest, pasture, cropland, and urban) (Linker and Thomann 1996). The Bay Watershed Model can now simulate the transport of increased or decreased atmospheric loadings to the Bay tidal waters along with nutrients from other land-based point and nonpoint sources. The Estuary Model is being upgraded to simulate basic ecosystem processes of underwater grasses, benthic microorganisms, and major zooplankton groups. In addition, EPA's RADM is being directly linked to the Watershed and Estuary models. This new integrated model, integrating the airshed, watershed, estuary, and ecosystem, is expected to be completed in early 1997. With these refinements, the integrated model (Figure IV-14) will simulate and evaluate the overall loads of controllable and uncontrollable nitrogen from the surrounding airshed and watershed, and the impact of these loads on the ecosystem. This model will be one of the tools used by Chesapeake Bay Program state and federal managers to formulate additional nitrogen reduction steps.

FIGURE IV-14
Integrated Model Improvements



* SAV = submerged aquatic vegetation

WATER QUALITY BENEFITS FROM REDUCING NITROGEN EMISSIONS

Using the Watershed Model, several scenarios were developed to examine the effectiveness of air emission controls compared to traditional point source and nonpoint source controls on the delivery of nutrient loads to the Chesapeake Bay. The Watershed Model scenarios were:

- Base Case Scenario: This scenario represents the base year 1985 loads to the Chesapeake Bay.

- Bay Agreement Scenario: This scenario represents the 40 percent controllable nutrient load reduction to be achieved by the year 2000 (as discussed under Nutrient Reduction Strategy in this section).

- Bay Agreement plus CAA Scenario: The scenario represents the controls on point and nonpoint source loads through the Bay Agreement, plus the atmospheric load reductions expected under full implementation of the CAA titles I (reductions in ground level ozone), II (mobile sources), and IV (utility sources).

- Bay Agreement plus Ozone Transport Commission (OTC) Scenario: The scenario evaluates reductions from the controls on point and nonpoint source loads through the Bay Agreement, plus the effects of implementation of CAA titles I, II and IV, as well as additional nitrogen reductions to reduce ground level ozone in the mid-Atlantic and New England metropolitan regions as called for by the Ozone Transport Commission.

- Limit of Technology Scenario: This scenario estimates the nutrient reductions that may be realized using the current practical limit of point and nonpoint source control technologies, including conservation tillage for all cropland implemented; the Conservation Reserve program fully implemented; nutrient management, animal waste controls, and pasture stabilization systems implemented where needed; a 20 percent reduction in urban loads; and point source effluent controlled to a level of 0.075 mg/L total phosphorus and 3.0 mg/L total nitrogen. This scenario is significant because it determines the limit of currently feasible control measures.

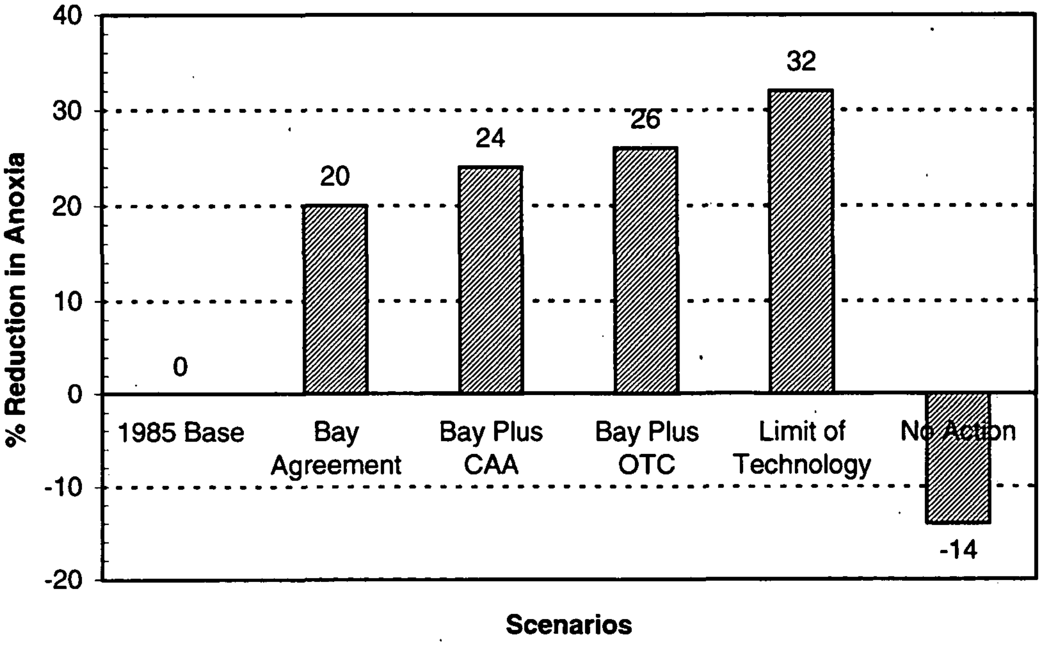
- No Action Scenario: This scenario represents loads that would occur in the year 2000 given population growth and projected changes in land use. The controls in place in 1985 were applied to the year 2000 point source flows and land use representing the loading conditions without the nutrient reductions stipulated in the Bay Agreement.

These reduction scenarios are part of an effort to evaluate options for achieving the 40 percent nutrient reduction goal. Land-based nonpoint source and wastewater treatment facility-based point source reduction actions, planned for implementation in many Chesapeake tributary watersheds, are approaching the limits of technology. Options for reductions in air emission are being explored for maintaining the targeted 60 percent nutrient loadings cap beyond the year 2000 in the face of a growing population and resultant development in the watershed. Different options will have different costs and effectiveness with regards to water quality improvements, and should be evaluated to find the best approach.

Figure IV-15 shows the water quality improvements from the expected reduction in nitrogen emissions under each scenario. The improvement in water quality reflects the estimated reductions in Bay bottom waters with no dissolved oxygen (i.e., reduction in Chesapeake anoxia or "dead waters"). Decreased nitrogen loadings will result in decreased water column nitrogen which will, in turn, decrease the growth of algae and improve the level of light penetration necessary to support the critically important underwater Bay grasses (Batiuk et al. 1992; CBP 1994c; Dennison et al. 1993; Thomann et al. 1994).

The CAA and OTC scenarios indicate that these controls provide for nitrogen loading reductions and water quality improvements above and beyond those provided by

FIGURE IV-15
Reductions in Anoxia Under Nutrient Reduction Scenarios



implementation of the Chesapeake Bay Agreement commitment of 40 percent reduction in controllable nitrogen. Relative reductions from controls on sources of atmospheric deposition vary by tributary, with the more sensitive tributaries being the Susquehanna and the Potomac. These basins receive the highest deposition loads in the Chesapeake watershed and are among the most responsive to reductions in atmospheric deposition. Table IV-10 shows in more detail the changes in nitrogen loading that would result from the CAA Scenario and the OTC Scenario.

TABLE IV-10
Reductions in Nitrogen Loadings to Chesapeake Bay and Water Quality Response

Control Scenario	Nitrogen Atmospheric Deposition Reduction		Nitrogen Load Reduction to Bay (in millions of pounds)	% Reduction in Anoxia
	To Bay	To Watershed		
Bay Agreement	0	0	75 (target)	20
CAA	9	14	14	4[a]
OTC	12	21	24	6[b]

[a] Determined by the difference between the Bay Agreement Scenario and the Bay Agreement plus CAA Scenario (Thomann et al. 1994).

[b] Extrapolation of Bay Agreement plus CAA Scenario to load reductions under OTC controls.

These numbers might seem small, but air emission controls could account for a fifth and a third of the baywide nitrogen load reduction goal through CAA implementation and OTC reductions, respectively. Such reductions could make achieving the 40 percent reduction target more feasible, and make maintaining a cap on any further increases in nutrient loadings beyond 2000 possible. These additional reductions are especially important in the face of increasing population and watershed development that studies predict will increase the significance of atmospheric deposition as a source of nitrogen loadings to the Chesapeake Bay in the coming decades (Fisher et al. 1988; Pechan 1991).

ADDRESSING AREAS OF UNCERTAINTY AND WORK UNDERWAY

The uncertainty in nitrogen retention, the relative loadings of ammonia and organic nitrogen, and dry deposition to water surfaces are remaining areas of significant uncertainty in estimating atmospheric nitrogen loads. Several specific examples of areas of uncertainty are discussed below.

- *Nitrogen retention* within watersheds makes a big difference in the proportion of the atmospheric contribution to nitrogen loading to the Bay. Different retention assumptions in mass balance analyses lead to an uncertainty in the estimate of this contribution by more than a factor of two.

- *Ammonia and organic nitrogen* contribute a large portion of nitrogen deposition, perhaps contributing more than 25 percent of the total atmospheric nitrogen load. However, it is unknown to what degree these sources are controllable; they may be changing with time. For example, airborne ammonia emissions from agricultural animal operations could increase.

- Estimates of the relative contribution of *dry deposition* to the total atmospheric deposition loadings range from 25 to 63 percent (Duce et al. 1991; Levy and Moxim 1987; Lindberg et al. 1986; Logan 1983; Lovett and Lindberg 1986; Sirois and Barrie 1988; Walcek and Chang 1987). Faced with this wide range of estimates, many investigators choose to set the dry deposition loading equal to the measured wet deposition loading. This assumption is known to be questionable. While site-specific data to refine the estimate are lacking, recent evaluations indicate that dry deposition to tidal water surfaces is about 25 percent of wet deposition to tidal water surfaces (Luke and Valigura 1996).

- *1990 baseline emission estimates* continue to be refined. Estimates of emissions from off-road vehicles have been significantly improved (compare U.S. EPA 1989 and U.S. EPA 1993). Ship emissions in harbors are suspected to be significantly underestimated (Booz-Allen & Hamilton 1991). While emissions from these sources are not large in the aggregate, they occur close to the Bay tidal surface waters, and thus have an influence greater than their national fractions would imply. Vehicle emissions continue to be a source of uncertainty in terms of nitrogen emissions.

- *Particulate nitrate* (which has a low deposition velocity) and *nitric acid* (which has a high deposition velocity) are currently indistinguishable by RADM, leading to modeling uncertainty.

- The contribution of atmospheric nitrogen deposition to *offshore ocean waters*, which exchange with waters of the Chesapeake Bay and thus can add to its nitrogen loads, is not yet characterized.

To reduce existing uncertainties in atmospheric loadings estimates, the Chesapeake Bay Program convened a workshop in June 1994, inviting scientists and managers with expertise and experience in understanding or managing atmospheric deposition. The challenge given to attendees was to construct a list of practical studies that would make the greatest impact on reducing the current uncertainty in estimates of the contribution of atmospheric deposition to declining aquatic ecosystem health. The resulting list is summarized below (CBP 1995a):

- *Priority 1*: Conduct intensive, coordinated, and integrated monitoring at special locations within the watershed that characterize wet deposition, dry deposition, and local catchment area.
- *Priority 2*: Improve existing atmospheric models (e.g., reduce grid size, account for the effect of mountains).
- *Priority 3*: Improve models of chemical retention in watersheds.
- *Priority 4*: Improve emissions inventories and projections.
- *Priority 5*: Conduct measurements to extend vertical and spatial meteorological and chemical concentration coverage in models.
- *Priority 6*: Establish an extensive array of less intensive measurement stations to improve spatial resolution for selected variables.

To improve the cross-media modeling capabilities and to reduce existing sources of uncertainty in atmospheric deposition loadings estimates, the following work is underway through cooperation between EPA, state and federal agencies, and universities:

- Conducting measurements of atmospheric organic nitrogen within the Chesapeake Bay watershed;
- Measuring dry deposition of nitrate to tidal surface waters of the Bay;
- Investigating the atmospheric deposition of dissolved organic nitrogen and its isotope (^{15}N);
- Linking daily atmospheric deposition and resultant nitrogen runoff from pasture, forested, and urban lands within the Chesapeake Bay Watershed Model;
- Decreasing the grid size of the RADM across the Bay watershed to increase the spatial scale of the resultant model scenario output;
- Linking the RADM with the Watershed, Estuary, and Water Quality models, including simulation of atmospheric deposition to offshore ocean waters and the exchange of the ocean waters with Chesapeake Bay waters.

The result of this and other work will become part of the integrated model of the Bay's airshed, watershed, estuary, and ecosystem (discussed above), which is expected to be completed in early 1997. A series of management scenarios, similar to those discussed above, are also expected to be completed in 1997 to examine the most feasible and cost-effective combination of point source, nonpoint source, and air deposition reductions to meet the Chesapeake Bay Agreement commitment to cap nutrient loadings in 2000 at 60 percent of 1985 controllable base loadings and to ultimately restore the water quality conditions necessary to fully support the Bay's invaluable living resources.

Toxic Contaminant Deposition to the Chesapeake Bay

CHESAPEAKE BAY BASINWIDE TOXICS STRATEGY

The 1987 Chesapeake Bay Agreement committed the signatories to "develop, adopt, and begin implementation of a basinwide strategy to achieve a reduction of toxics consistent with the Clean Water Act of 1987, which will ensure protection of human health and living resources" (Chesapeake Executive Council 1987). The resultant strategy, adopted in 1989, initiated a multi-jurisdictional effort to more accurately define the nature, extent, and magnitude of Chesapeake Bay chemical contaminant problems and to initiate specific chemical contaminant reduction and prevention actions (Chesapeake Executive Council 1989). Building on a two-year reevaluation of the strategy and increased understanding of the nature of the Bay's toxics problems, a revised, farther-reaching strategy was adopted in 1994. The 1994 *Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy* recognized the contribution of atmospheric deposition as a significant source of chemical contaminant loadings to the Bay. Within the basinwide strategy, the signatories committed to establishing a more comprehensive loadings baseline and setting an atmospheric deposition loading reduction target to be achieved over the next decade (Chesapeake Executive Council 1994).

Extent of Toxic Contamination in Chesapeake Bay

Prior to adoption of the 1994 *Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy*, the Chesapeake Bay Program conducted a two-year, in-depth evaluation of the nature, extent, and magnitude of toxic contaminant-related problems within the Chesapeake Bay. Through the evaluation, no evidence was found of severe baywide impacts from chemical contamination, unlike other problems facing the Chesapeake Bay, such as the impacts from excess nutrients. The Program did document severe, *localized* toxicity problems, adverse effects from chemical contamination on aquatic organisms in areas previously considered unaffected, and widespread low levels of chemical contamination in all Bay habitats sampled.

Existing state and federal regulatory and management programs continue to reduce the input of potential toxic chemicals to the Chesapeake Bay. Measured concentrations of many of these chemical contaminants in the Bay's bottom sediments, shellfish, fish, and wildlife have also generally declined, although elevated levels occur in several industrialized areas and some increasing trends have been observed. Progress in reducing the point sources of these chemical contaminants is offset by significant nonpoint source inputs of chemical contaminants (e.g., urban stormwater runoff, atmospheric deposition) from increasing development and urbanization of the Bay watershed.

In 1991, the Chesapeake Bay Program adopted its first Chesapeake Bay Toxics of Concern list, principally to identify and provide concise documentation on chemical contaminants that adversely impact the Bay or have a reasonable potential to do so. This list provided Chesapeake Bay region resource managers and regulators with a baywide consensus of priority chemicals and the information necessary to target these chemical contaminants for strengthened regulatory and prevention actions or additional research, monitoring, and assessment. In response to a commitment within the 1994 Basinwide Toxics Strategy, the Toxics of Concern list was evaluated and revised using a risk-based chemical ranking system incorporating source, fate, and exposure/effects ranking factors. In-depth analyses of the top-ranked chemicals led to the selection of the revised Toxics of Concern list (see sidebar). This list includes several pollutants of concern to the Great Waters program. In addition, a list of Chesapeake Bay Chemicals of Potential Concern was identified (see sidebar) and includes a pesticide of concern for the Great Waters (toxaphene) and one potential Great Waters pollutant of concern (atrazine). Clear evidence is lacking that the contaminants on this list actually or have reasonable potential to cause impacts, but they warrant enough concern to be carefully monitored and tracked. For example, a number of the chemicals listed as being a potential concern are either banned or restricted pesticides that have residues still remaining in the ecosystem at elevated levels but below thresholds of concern; others are chemicals of increasing concern due to use patterns or potential for toxicity to Bay resources.

Chesapeake Bay Toxics of Concern

Toxics of Concern: atrazine, cadmium, chlordane, chromium, copper, lead, mercury, PCBs, tributyltin, several PAHs (i.e, benz(a)anthracene, benzo(a)pyrene, chrysene, fluoranthene, and napthalene).

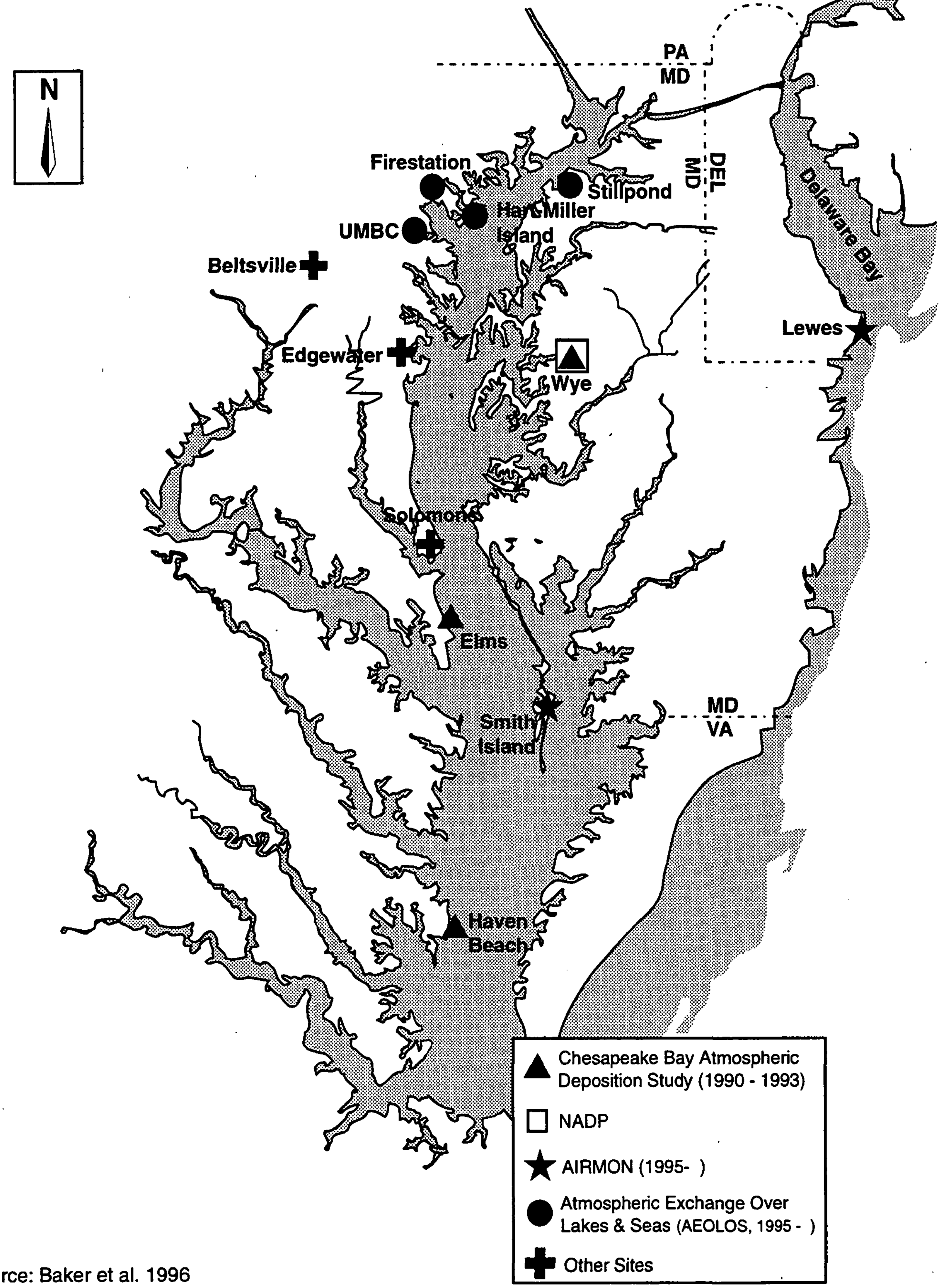
Chemicals of Potential Concern: alachlor, aldrin, arsenic, dieldrin, fenvalerate, metolachlor, permethrin, toxaphene, and zinc.

CHESAPEAKE BAY TOXIC CONTAMINANT ATMOSPHERIC DEPOSITION STUDIES

Studies conducted in the southern Chesapeake Bay in the early 1980s suggest that the atmosphere is a significant source of organic contaminants. For example, Velinsky et al. (1986) determined that organic carbon loadings in bulk atmospheric deposition were greater than the estimated tributary loadings of organic carbon to the Bay. Anthropogenic hydrocarbons, including PAHs, were detected in bulk deposition samples, with calculated loadings estimated to be comparable to hydrocarbon loadings from the municipal wastewater discharges to the Bay (Webber 1993). While these earlier studies demonstrated the potential importance of the atmosphere in supplying contaminants to the Chesapeake Bay, they were limited by their methodologies (i.e., bulk deposition sampling) and their relatively limited temporal and spatial scope. Accurate estimates of baywide annual loadings require determining the spatial and temporal variability in contaminant loads in the atmosphere and in depositional fluxes to the Bay.

To help quantify the atmospheric loadings and the depositional fluxes to the Bay, the Chesapeake Bay Atmospheric Deposition Study (CBADS) network was established by a team of scientists from the University of Maryland, Virginia Institute of Marine Sciences, University of Delaware, and Old Dominion University. The primary objective of the CBADS network was to provide the best estimate of total annual atmospheric loadings of a variety of trace elements and organic contaminants directly to the surface waters of the Chesapeake Bay. Based on previous studies of wet deposition in the region (e.g., Tyler 1988) and given the resources available for the network, three non-urban shoreline locations -- Wye Institute and Elms Institute, Maryland, and Haven Beach, Virginia -- were selected and sampled from 1990 to 1993. These three sampling site locations, as well as other monitoring sites around Chesapeake Bay, are shown in Figure IV-16.

FIGURE IV-16
Sampling Locations



Source: Baker et al. 1996

In establishing this initial network, the influence of urban areas was purposely avoided by locating the sites more than 50 kilometers from metropolitan areas (similar to the initial non-urban stations for the Great Lakes deposition network, IADN). By minimizing possible urban influences, the resulting CBADS loading estimates are considered to be minimum values. Although it is most likely that some fraction of the materials deposited from the atmosphere to the watershed of the Chesapeake Bay are ultimately transported to the surface waters, CBADS did not attempt to characterize loadings to the watershed in this study. Because deposition to the various land surfaces is likely much different than that to the water surface, fluxes measured at the shore-based stations cannot be extrapolated with confidence to the watershed. Additionally, the large uncertainty in the understanding of the fate of materials deposited to the land surface (i.e., the fraction transmitted to the receiving water) precludes the simple estimation of the indirect atmospheric loading of contaminants to the Chesapeake Bay in more than a rough estimate. The following information on concentrations in air, concentrations in precipitation, and wet and dry aerosol depositional fluxes were extracted from the CBADS results presented in Baker et al. (1996). Results are presented for several PAHs, PCBs, and several metals (including cadmium and lead). Mercury data are being collected but have not yet been compiled; data on other Great Waters pollutants of concern were not collected.

Concentrations in Air. The elemental composition of aerosol particles less than 10 µm in diameter was dominated by the crustal elements aluminum and iron, as well as sulfur (in the form of sulfate). During 1991 and 1992, concentrations averaged over the three sampling sites were 116, 111, and 2,123 ng/m^3 for aluminum, iron, and sulfur, respectively. Trace element concentrations averaged over the same period ranged from 0.16 ng/m^3 for cadmium to 12.6 ng/m^3 for zinc, with lead averaging 3.88 ng/m^3. The fraction of each element derived from non-crustal (e.g., combustion) sources was estimated based on the average concentration of elements in the Earth's crust (Turekian and Wedepohl 1961), and assuming all of the measured aluminum associated with aerosol particles is derived from erosion of soils. In the Chesapeake Bay region, non-crustal sources supply greater than 95 percent of most of the elements measured on aerosol particles (Wu et al. 1994). Arsenic, cadmium, lead, sulfur, and selenium are almost exclusively non-crustal, and are likely introduced into the atmosphere as a result of combustion of fossil fuels and incineration of municipal wastes.

The measured concentrations of trace elements were generally within a factor of two among the three sampling sites. On an average annual basis, concentrations slightly decrease from north (Wye) to south (Haven Beach), except sulfur, which is 15 percent higher at Elms (1991 and 1992) and Haven Beach (1992) than at the Wye site. The general decreasing trends observed from north to south, in light of increasing sulfate, may indicate higher levels of low sulfur combustion sources (e.g., incinerators, vehicles) in the northern reaches of the Chesapeake Bay. In general, the variability in the atmospheric concentrations of trace elements between sites (from north to south) is substantially lower than corresponding temporal trends (discussed below).

Semivolatile organic chemicals exist in the atmosphere as gases and also are associated with aerosol particles (Pankow 1987). In this study, baywide annual average concentrations of PAHs ranged from 16 ng/m^3 for dibenz[*a*,*h*]anthracene to 2,590 ng/m^3 for phenanthrene. Atmospheric concentrations were quite variable with individual measurements ranging from one-tenth to ten times the annual average concentrations. These variations likely result from sampling air masses coming from differing directions, from changes in local and regional emissions, and from differences in atmospheric degradation and deposition rates. For example, increased concentrations of gas-phase PAHs, such as pyrene, during the summer months may reflect both higher temperatures (i.e., enhanced volatilization) and increased coal and oil combustion to meet electrical demand for air conditioning. Increases in the atmosphere of particulate PAHs, such as benzo[*a*]pyrene, may result from local burning of yard wastes and of wood for home heating. Some variation in atmospheric levels of organic chemicals may result from the efficient removal of particulate PAHs by precipitation (Poster and Baker 1996a, 1996b). In general, the magnitude of fluctuations in atmospheric levels of organic chemicals is larger than the corresponding variations in trace elements and sulfur discussed above, suggesting the importance of local combustion sources. Air concentrations were not available for PCBs.

Concentrations in Precipitation. The overall volume-weighted mean concentrations of trace elements in precipitation collected at the three sites range from 0.03 µg/L for cadmium at Elms to 14.6 µg/L for iron at Haven Beach. For lead, overall volume-weighted mean concentrations were approximately 0.40 µg/L at the three sites. The relative proportion of trace elements in precipitation is nearly identical to that in the Chesapeake Bay aerosol particles, confirming that aerosol scavenging is the source of trace metals to wet deposition. Trace metal wet depositional fluxes are highly variable, changing more than ten-fold between consecutive months with little apparent seasonal dependence. This variability, which was similar at each of the three sites, results not only from fluctuations in the atmospheric inventories of trace metals, but also from changes in the amount of precipitation. On an annual basis, the volume-weighted mean concentrations of most trace metals did not systematically vary between the summer of 1990 and fall of 1993, again suggesting that these rural sites were most strongly influenced by the same regional background signal throughout the study.

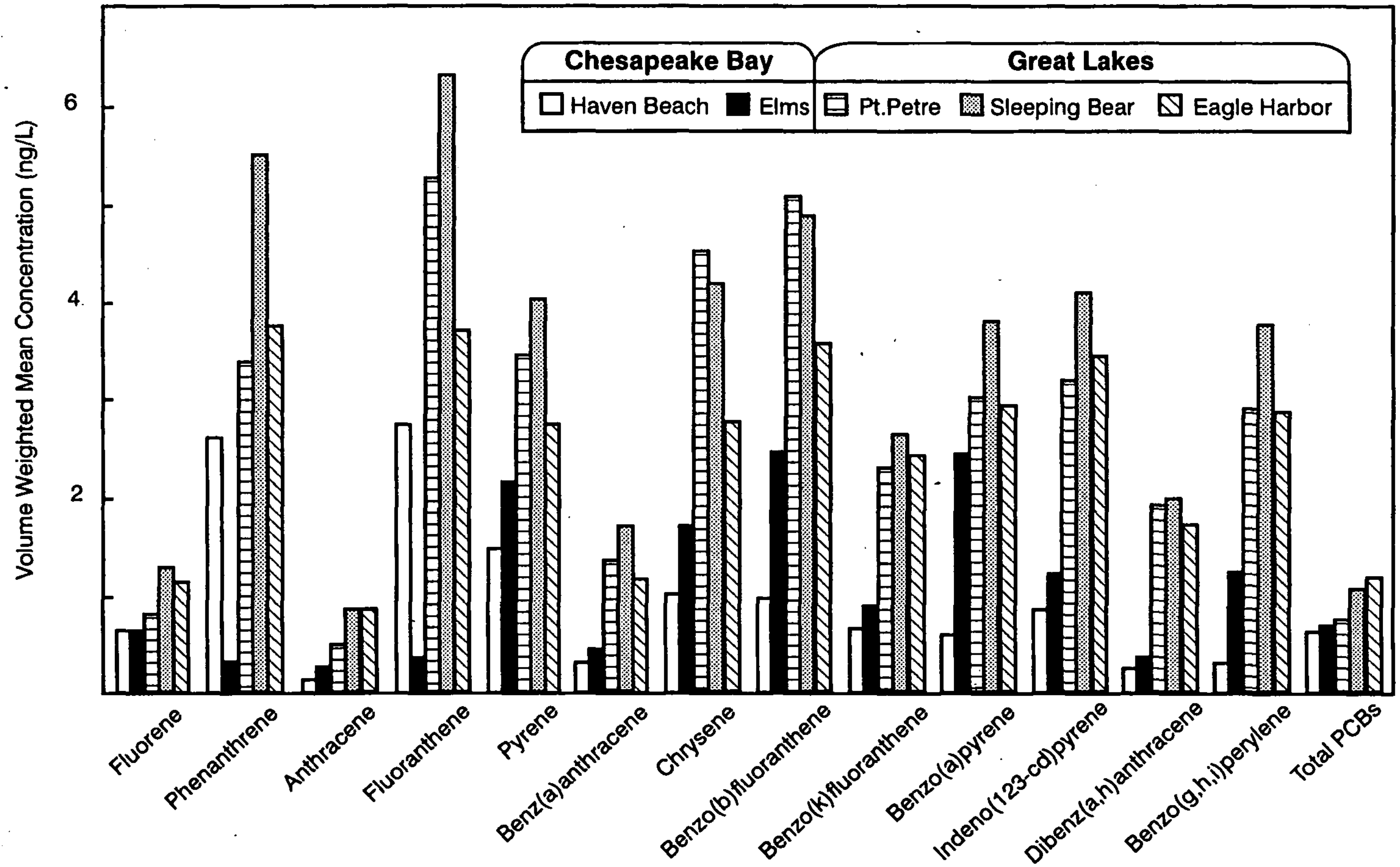
While individual precipitation events result in spikes in trace metal deposition at one site that are not observed at the other two locations, these isolated events average out over the year. Annual volume-weighted mean concentrations of trace metals in precipitation are generally within a factor of two among the three sites, with no clear systematic spatial trend observed for all metals.

Overall volume-weighted mean concentrations of PAHs in precipitation ranged from 0.3 ng/L for anthracene and dibenz[*a,h*]anthracene at the Havens Beach site to 9 ng/L for pyrene at the Elms site. Volume-weighted mean concentrations for total PCBs were 1.1 and 0.9 ng/L at the Elms and Haven Beach sites, and for a shorter period of time, 0.35 ng/L at the Wye site. As was observed for trace metals, wet depositional fluxes of organic contaminants varied considerably with time, and were dominated by episodic spikes at each location. Extremely high concentrations of some analytes, including pyrene, that were measured in both air and precipitation at the Elms site in the summer of 1990, may have resulted from local vegetation burning. Unlike PAH and PCB levels in the atmosphere, concentrations in precipitation did not systematically vary with season. The enrichment at the Elms site is especially pronounced for higher molecular weight PAHs, implicating a local combustion source (e.g., wood burning for residential heating).

Concentrations of organic contaminants in precipitation measured in this study are consistently lower than those observed in the Great Lakes region (see Figure IV-17). For example, PAH concentrations in precipitation at the Chesapeake Bay sites are one-third to one-half as high as at the three IADN sites located at rural, shoreline sites on Lakes Ontario, Michigan, and Superior (Gatz et al. 1994). In contrast, levels of the same PAHs in the air

FIGURE IV-17

Comparison of 13 PAHs and Total PCBs in Precipitation (1992) from Chesapeake Bay and Great Lakes Sampling Sites



Source: Baker et al. 1996 (Chesapeake Bay) and Gatz et al. 1994 (Great Lakes).

* * * DRAFT: Do Not Cite or Quote -- October 21, 1996 * * *

over Chesapeake Bay are equal to or perhaps more less than those measured over the Great Lakes. Whether the apparent enrichment in PAHs in Great Lakes precipitation relative to that in the Chesapeake Bay region, as shown in Figure IV-17, is due to more efficient scavenging by precipitation in the colder, drier region, or simply reflects methodological differences between the two networks is unclear.

Wet and Dry Aerosol Depositional Fluxes. Total annual fluxes in 1992 ranged from 0.07 mg/m^2 for cadmium at the Wye site to 121 mg/m^2 for aluminum at the Elms site; the highest flux for lead was 1.34 mg/m^2 at the Wye site. Not surprisingly, dry aerosol deposition comprises the majority of the total flux for the soil elements aluminum and iron which occur on coarse particles. Wet deposition contributes between one-third and one-half of the total depositional flux of the remaining trace elements. Naturally, spatial trends in total deposition result from variation in precipitation chemistry and amount, and the trace element inventories associated with aerosol particles (given the considerable uncertainty in the dry aerosol deposition calculation, the same deposition velocity was used at each site). Although a distinct north to south trend in precipitation amount occurred in 1992 (100, 107, and 122 cm, respectively), total annual depositional fluxes were remarkably similar among the three stations. Total depositional fluxes were also very similar between years, again indicating that the relatively rare spikes in concentration are dampened against the chronic regional background signal.

For PAHs, total annual fluxes for 1992 range from 0.2 µg/m^2 for anthracene at the Wye site to 10.8 µg/m^2 for benzo[*b*]fluorathene at the Elms site.[14] Both wet deposition and dry aerosol deposition contribute to total PAH deposition, with dry aerosol deposition becoming relatively more important for the higher molecular weight, less volatile compounds. Total depositional fluxes of PAHs decrease with time during this study, with the lowest fluxes measured during the first nine months of 1993. While some of this decrease is attributed to beginning with anomalously high measurement in the summer of 1990, we continued to see decreases in both wet and dry aerosol fluxes between 1992 and 1993. The total annual deposition flux of total PCBs is about 3.5 µg/m^2, with approximately equal contribution from wet and dry aerosol deposition.[15]

Overall, total annual fluxes of PAHs and PCBs are generally within 50 percent among the sites. Given the uncertainty in the dry aerosol deposition estimates, this percentage indicates little spatial variability when integrating over annual cycles. However, this study did not specifically address the possible influences of urban areas, such as the cities of Baltimore, Washington, and Norfolk, on atmospheric deposition, which may be important.

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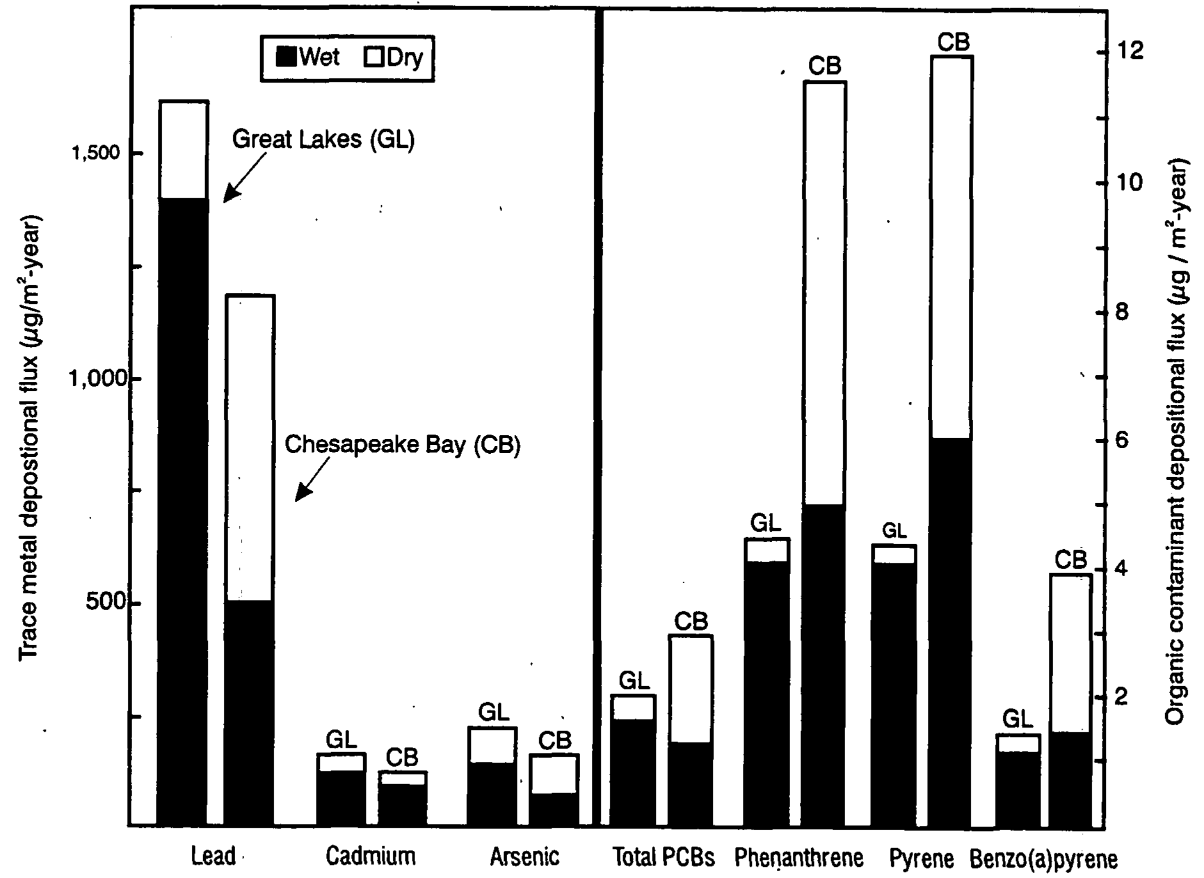
[14] It is important to note that these flux estimates do not include passive exchange of gaseous organic contaminants across the air-water interface. Other recent studies have shown that this is the *dominant* atmospheric deposition process for semi-volatile organic contaminants, including PCBs (Baker and Eisenreich 1990; Achman et al. 1993) and low molecular weight PAHs (Nelson et al. 1995). In those studies, the net direction of exchange is often from the water to the air and diffusive gas exchange is large enough to offset wet deposition and dry aerosol deposition. Therefore, the depositional fluxes presented here should be considered gross fluxes to the water surface, rather than net exchanges.

[15] Because aerosol particle-associated PCBs were present below analytical detection limits, estimates of PCB dry deposition were made using an aerosol PCB level calculated from the measured gaseous PCB concentration and the Junge-Pankow sorption model (Pankow 1987; Leister and Baker 1994).

FIGURE IV-18
Comparison of Chesapeake Bay and Great Lakes Atmospheric Depositional Fluxes



Source: Baker et al. 1996 (Chesapeake Bay) and Eisenreich and Strachan 1992 (Great Lakes).

Annual Great Lakes fluxes were estimated for each lake based upon several sources of data, including the initial results of the IADN program, and then averaged.

To place the atmospheric deposition fluxes measured in this study in perspective, they are compared to similar estimates made for the Great Lakes region (Figure IV-18). Wet depositional fluxes of trace elements are up to three times higher in the Great Lakes than in Chesapeake Bay, despite significantly less rainfall (80 versus 110 cm/year). In contrast, wet depositional fluxes of total PCBs and PAHs are remarkably similar between the two regions, as higher concentrations in Great Lakes precipitation (Figure IV-17) are offset by lower precipitation amounts. Dry aerosol depositional flux estimates are higher in the Chesapeake region, especially for organic contaminants.[16] In addition, measured aerosol-bound organic concentrations were generally higher than the values used in the Great Lakes dry aerosol deposition calculations (Eisenreich and Strachan 1992). Despite the differences, estimated

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[16] This is due, in part, to the choice of deposition velocities used for the two estimates (0.2 cm/sec for all species in the Great Lakes, 0.26 cm/sec for elements and 0.49 cm/sec for organics in the Chesapeake Bay). While all of these values are within the generally accepted range for dry aerosol deposition velocities, the impact of this uncertainty can be seen in Figure IV-18.

TABLE IV-11
Annual Loadings of Trace Metals and Organic Contaminants to the Chesapeake Bay[a]

	Wet Deposition (kg/year)			Dry Deposition (kg/year)			Total Deposition (kg/year)		
Pollutant	1991	1992	Overall	1991	1992	Overall	1991	1992	Overall
Aluminum[b]	160000	114000	137000	1300000	1093000	1200000	1460000	1207000	1340000
Arsenic	580	633	607	1000	1094	1050	1580	1727	1660
Cadmium	920	813	867	130	347	240	1050	1160	1110
Chromium	1100	951	1026	2000	2064	2030	3100	3020	3060
Copper	6100	5049	5575	3500	3745	3620	9600	8790	9200
Iron	120000	145600	132800	750000	582000	666000	870000	727000	799000
Manganese	12700	13700	13200	15000	12200	13600	27700	25900	26800
Nickel	3000	11370	7185	5300	7019	6160	8300	18400	13300
Lead	6700	4180	5440	5200	8960	7080	11900	13100	12500
Selenium	1500	1280	1390	3000	2860	2930	4500	4140	4320
Zinc	18000	35300	26000	23000	22600	22800	41000	57800	49400
PAHs									
Anthracene	7	3	6	8.9	3	6	16	7	13
Benz(*a*)anthracene	10	5	9	47	17	34	57	22	44
Benza[*a*]pyrene	18	18	17	52	24	36	70	42	53
Benzo[*b*]fluoranthene	31	21	36	138	58	98	169	79	134
Benzo[*e*]pyrene	21	17	21	90	49	67	111	66	88
Benzo[*ghi*]perylene	27	10	19	113	44	75	141	54	94
Benzo[*k*]fluoranthene	18	11	22	99	39	65	117	50	88
Chrysene	31	17	29	116	53	85	146	70	114
Dibenz[*ah*]anthracene	6.7	5	7	27	7	16	34	12	22
Fluoranthene	61	34	70	176	44	119	237	78	189
Fluorene	23	7	16	16	7	12	39	14	27
Indeno[123*cd*]pyrene	27	13	20	141	38	78	168	51	98
Phenanthrene	85	31	63	127	55	92	211	86	155
Pyrene	43	24	75	147	47	109	190	70	184
Total PCBs	17	9	13	20	NA	20	37	NA	37

[a] To estimate annual baywide loadings of elements and organic contaminants to the entire Chesapeake Bay, the annual site specific wet and dry aerosol fluxes were averaged and these two average fluxes were multiplied by the surface area of the bay (1.5 x 10^{10} m^2).

[b] Contribution of aluminum is considered to be entirely from natural sources (i.e., not emitted through human activities).

Source: Baker et al. 1996.

atmospheric depositional fluxes are generally within a factor of two between the Chesapeake Bay and the Great Lakes regions, which, given the numerous opportunities for error in these measurements and calculations, is quite good agreement.

CHESAPEAKE BAY TOXIC CONTAMINANT ATMOSPHERIC LOADINGS

Through basinwide measurements of toxic contaminant concentrations and known releases to air, land, and surface water, atmospheric loadings were estimated and compared with other sources of chemical contaminants. The atmospheric deposition loadings used in the comparison include only wet and dry deposition to the surface waters of the Bay and its tidal tributaries. An unquantified, yet probably significant, fraction of urban stormwater runoff actually represents atmospheric deposition to impervious surfaces (e.g., pavement).

Baywide atmospheric loadings of aluminum and iron are estimated at 1,340,000 and 799,000 kg/year, respectively (Table IV-11). Loadings of trace elements range from 1,110 kg/year for cadmium to 49,400 kg/year for nickel; lead loadings are estimated at 12,500 kg/year. Loading estimates are generally similar for 1991 and 1992, except for nickel and zinc due to elevated wet deposition measured at the Haven Beach site in 1992. Loadings of PAHs range from 13 kg/year for anthracene to 189 kg/year for fluoranthene. Total PCB loadings are estimated to be 37 kg/year. Interestingly, for many of the pollutants in Table IV-11, wet deposition and dry aerosol depositional fluxes appear to decrease between 1991 and 1992. Whether this reflects a real inter-annual variation or simply results from aggregating measurements from different locations is unclear.

To place these loadings in perspective, they are compared to recent estimates of trace metal and organic contaminant loadings delivered to the Chesapeake Bay by the Susquehanna River in Table IV-12 (Conko 1995; Foster 1995; Godfrey et al. 1995). The Susquehanna River is the largest tributary of the Chesapeake Bay, supplying approximately 60 percent of the freshwater inflow to the estuary. Annual riverine loadings of dissolved and particulate trace metals and organic contaminants were determined by analyzing flow-weighted samples collected at Conowingo, Maryland, between February 1994 and January 1995 (Conko 1995; Foster 1995). Atmospheric deposition directly to the surface waters of the Chesapeake Bay supplies PAH loads that are comparable to or greater than the loads of dissolved PAHs delivered by the Susquehanna River (Table IV-12). Dissolved total PCB loads from the river are approximately three times those from the atmosphere. Particulate-bound organic contaminants discharged from the river dominate the loading of PAHs, with a large contribution from the high sediment burden carried by the river during high flows. Atmospheric depositional fluxes of several elements, including lead, cadmium, and chromium, are within a factor of two of the dissolved load from the Susquehanna River. Again, particulate metal loads from the river dominate over both dissolved riverine loads and atmospheric deposition.

While it is interesting to compare the relative importance of riverine and atmospheric sources of trace elements and organic contaminants to the Chesapeake Bay, the results should be carefully interpreted. While the Susquehanna River delivers large quantities of these compounds to the Bay, much of this load is removed in the northern extreme of the Bay (Helz and Huggett 1987) and is delivered episodically during high river flows. Whether particulate-bound metals and organic contaminants are broken down in forms that can be taken up by aquatic organisms is quite unclear. In contrast, atmospheric deposition directly to the water's surface supplies these toxics directly to the water column, without any

TABLE IV-12
Relative Importance of Sources of Trace Metals and Organic Contaminants to Chesapeake Bay

Pollutant	Susquehanna River Load (kg/year)[a]		Atmospheric Deposition Load (kg/year)[b]
	Dissolved	Particulate	
Aluminum (x 10^{-3})	2560	64800	1340
Arsenic	12600	ND	1660
Cadmium	2130	26700	1110
Chromium	4130	111000	3060
Copper	47800	151000	9200
Iron (x 10^{-3})	4100	40000	799
Manganese (x 10^{-3})	3290	1530	26.8
Nickel	121000	65200	13300
Lead	6530	38600	12500
Zinc	77900	360000	49400
PAHs			
Benz[*a*]anthracene	12	364	44
Benzo[*a*]pyrene	5	436	53
Chrysene	15	316	114
Fluoranthene	108	1020	189
Fluorene	37	85	27
Phenanthrene	63	388	155
Pyrene	104	925	184
Total PCBs	97	68	37

[a] Annual loads entering the Chesapeake Bay via the Susquehanna River, measured at Conowingo, Maryland, between February 1994 and January 1995 by Foster (1995) for organic compounds and Conko (1995) for metals.

[b] Total atmospheric deposition loads directly to the surface of the Chesapeake Bay below the fall lines (1.15 x 10^{10} m^2), as measured by the Chesapeake Bay Atmospheric Deposition Study, 1990-1991.

Source: Baker et al. 1996.

comparable zone of efficient removal. However, it has recently been suggested that combustion-derived PAHs associated with aerosol particles washed into the surface waters by precipitation also may not be broken down (McGroddy and Farrington 1995). Finally, the distinction between riverine and atmospheric loadings is not clear. Some fraction of the pollutant input from the tributaries results from deposition of atmospheric pollutants to the watershed, with subsequent transmission through the vegetation and soil layers into surface waters; however, this input cannot yet be quantified.

ADDRESSING AREAS OF UNCERTAINTY

Building on the existing CAA requirements, the Chesapeake Bay Program's state and federal partners will focus their efforts on implementation of the Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy commitment to "establish a more complete baselineand source identification for atmospheric deposition...and set a reduction target from that baseline to be achieved over the next decade" (Chesapeake Executive Council 1994). However, there are several remaining areas of uncertainty to be addressed.

- *Dry Deposition.* Dry deposition is viewed as an important mechanism by which chemical contaminants are deposited onto the Bay's tidal surface waters and surrounding watershed. As is the case with nitrogen, there are no widely accepted techniques for direct measurement of dry deposition fluxes of metal or organic contaminants. Although no direct measurements of dry deposition directly to the Bay exist, depositional fluxes have been estimated based on a particle-size-dependent deposition velocity function applied to direct measurements of aerosol concentrations of metals and organic chemical contaminants. Given the absence of direct measures of dry deposition fluxes, there is much uncertainty associated with these loading estimates.

- *Transport through the Watershed.* Atmospheric deposition of a pollutant can be a direct input to the Bay surface waters or can be transported from the watershed by surface water and groundwater to the Bay. Transported loads are a component of the total fluvial (i.e., surface water) input from the watershed to the Bay. The degree of landscape retention for a given pollutant is related to the geomorphology, land use, basic hydrological characteristics unique for each watershed, and soil chemistry. Limited studies to date suggest that the degree of watershed throughput is relatively small (< 30 percent of the rate of rainfall volume). However, evidence to date suggests that watersheds serve as a "reservoir" for atmospherically deposited metals; organically-bound metals are sequestered but may be episodically mobilized by acidic precipitation. Because of the relatively large watershed to open water surface area ratio typical of coastal plain estuaries such as Chesapeake Bay (15:1), recent estimates for nitrogen and trace elements suggest that the indirect atmospheric loading may be as significant as the direct input. Thus, while it has been possible to quantify direct atmospheric flux with a fair degree of confidence, one of the primary uncertainties associated with resolving the total atmospheric loading to Chesapeake Bay is in gauging the indirect loading as it relates to the watershed transmission/retention for the myriad of sub-basins (Valigura et al. 1995).

To further improve existing estimates of the relative atmospheric deposition contribution to total chemical contaminant loadings to Chesapeake Bay, the following work is underway. In 1993, the University of Delaware, in cooperation with the U.S. Geological Survey, initiated an 18-month pilot study to investigate the transport of atmospherically deposited trace elements through a pristine, forested watershed in the headwaters of the Potomac River (Bear Branch). This study, funded by the Maryland Department of Natural Resources' Power Plant Research Program, has the following specific objectives: (1) to accurately determine the wet and dry atmospheric trace element loads into the watershed, (2) to compare the total atmospheric load versus fluvial output of trace metals and (3) to estimate the transport of atmospherically deposited trace elements through the watershed relative to the trace metals naturally relapsed during weathering of the soil and rock within the study area. The Bear Branch watershed was chosen as it has been well-characterized hydrologically, is representative of the land use in the Potomac basin (60 percent of which is forested), and possesses an unreactive quartzite lithology which simplifies its geochemical weathering behavior. Further watershed transmission studies are scheduled to commence in the spring of 1996 in the Appalachian Plateau of Western Maryland. While the results of these studies will represent an initial attempt to quantify the watershed retention/transport of atmospheric loads, further work is needed to extend the study to other regions with differing land use/geomorphology, in order to accurately determine an integrated, baywide watershed transport factor.

IV.D Coastal Waters

As stated previously, section 112(m) designates "coastal waters," for purposes of the Great Waters program, as EPA's National Estuary Program (NEP) and NOAA's National Estuarine Research Reserve System (NERRS) estuaries. These two programs and EPA's Gulf of Mexico Program are the three significant coastal waters programs, other than the Chesapeake Bay Program, established in the last decade.[17] Each of the three programs differs slightly in purpose and procedure, but they all serve to protect and restore the nation's valuable coastal water resources. Background information on each of these programs, plus a discussion of atmospheric deposition studies relevant to the program, where available, are presented below.

National Estuary Program

Congress established the National Estuary Program (NEP) in 1987 under Section 320 of the Clean Water Act. Through the NEP, states nominate estuaries of national significance that are threatened or impaired by pollution, development, or overuse. EPA evaluates the nominations and selects those estuaries that show evidence of political support, citizen and government involvement (local, state, regional, federal), and available scientific and technical information to address the problem. EPA convenes management conferences with representatives from all concerned groups (e.g., industry, agriculture, environmental organizations, state agencies) to more fully characterize problems and seek solutions through a collaborative decision-making process. Through these conferences, Comprehensive Conservation and Management Plans (CCMPs) are developed, for which EPA provides up to 75 percent of the funding. The management conference must complete development of the CCMP within three to five years of the date the conference was convened. Upon approval of the CCMP, action plans are carried out by implementation agencies involved with development of the plan.

The purpose of the NEP is to identify nationally significant estuaries, protect and improve their water quality, and enhance their living resources. The NEP currently includes 28 estuaries representing a wide spectrum of environmental conditions (see Figure IV-19 for the location of the NEP estuaries). Because there are over 150 estuaries in the United States and only a small fraction can be targeted for action through the NEP, the NEP is intended to act as a national demonstration program, such that results and lessons learned in the NEP estuaries are shared and applied by parties concerned with other estuaries throughout the country. It should be emphasized that the NEP is a management program rather than a research program and relies on the research of other agencies and institutions to support its work. The development of support networks and cooperation between local, state, regional, and federal agencies is one of the program's greatest assets.

A number of NEP estuaries have identified atmospheric deposition as a concern and are interested in pursuing studies to investigate the role of atmospheric deposition as a source of pollutants. Others have completed studies that examine atmospheric deposition loadings of nitrogen and other pollutants. To date, the Tampa Bay and Galveston Bay NEPs

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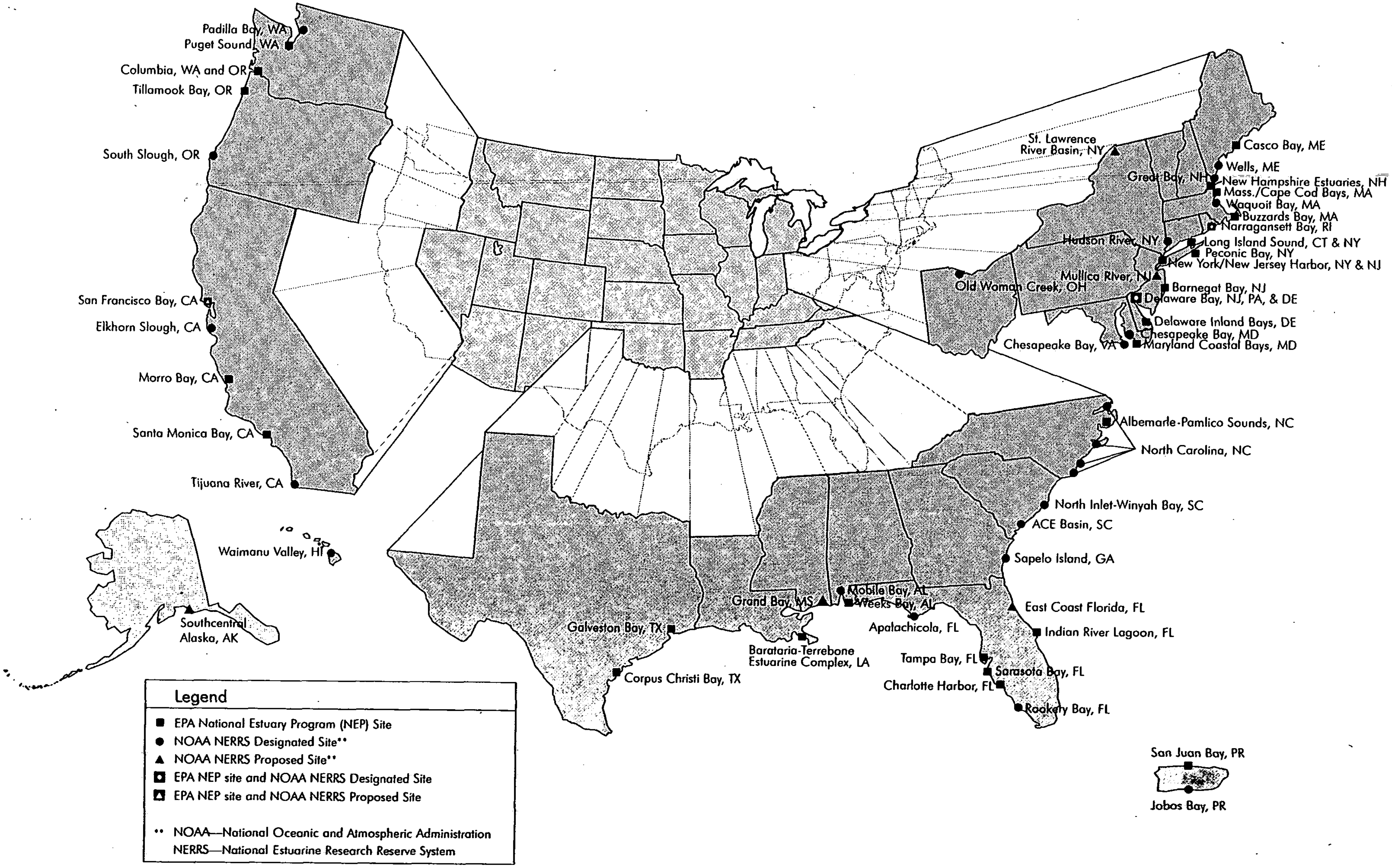
[17] Although the Gulf itself is not a designated Great Water under the CAA, many of the estuaries of the Gulf are NEP or NERRS estuaries. In addition, issues of importance for the Great Waters estuaries are similarly important for the Gulf.

FIGURE IV-19
Locations of NEP and NERRS Sites



have received CAA section 112(m) funds from EPA to conduct such work. Studies and activities related to atmospheric deposition in NEP estuaries, as well as future research needs, are discussed in the remainder of this section.

STUDIES OF ATMOSPHERIC DEPOSITION IN NEP WATERS

The largest sources of pollutants to coastal waters have historically been considered to be municipal and industrial wastewater discharges and urban runoff/storm sewer inputs. Recently, however, researchers have begun to investigate the role of atmospheric deposition as a source of pollutants in a coastal waters. Assessing the impact of atmospheric deposition of pollutants has become a priority for many NEP estuaries. There is a clear need to characterize the types, quantities, and sources of pollutants that are being directly and indirectly deposited from the atmosphere into these estuaries. Recent studies on atmospheric deposition to coastal waters are discussed briefly below and are presented in Table IV-13. In general, these studies have evaluated the relative loadings of nitrogen and other pollutants of concern, and do not attempt to identify the particular emission sources contributing to atmospheric deposition. Initial observations from the limited scope and number of studies suggest that direct and indirect air deposition may be significant sources of nitrogen and toxic pollutants to NEP estuaries.

As mentioned above, research on atmospheric deposition to Tampa Bay, Florida, and Galveston Bay, Texas, has been conducted under the Great Waters program. The Tampa Bay and Galveston Bay studies are discussed first, followed by a description of other studies of atmospheric deposition to NEP estuaries (i.e., those that have been funded through sources other than EPA's Great Waters program).

Tampa Bay. As recently as 1991, atmospheric deposition of nutrients and pollutants was assumed to have a minimal effect on water quality in Tampa Bay. However, a recent study of nitrogen loading conducted for the Tampa Bay NEP suggests that direct deposition of nitrogen is the second largest source of nitrogen entering the Bay, with a contribution of up to 27 percent of the total nitrogen load (Zarbock et al. 1994). The largest source of nitrogen is urban stormwater runoff, a portion of which originates from atmospheric deposition to impervious surfaces. When atmospheric deposition to the entire watershed is taken into account, initial studies indicate that as much as 67 percent of the nitrogen load to the Bay may originate from atmospheric deposition (Patwardhan and Donigian 1996). These findings prompted the Tampa Bay NEP to revise its CCMP to consider atmospheric deposition issues.

In another study, atmospheric deposition was found to be a significant source of cadmium, chromium, and copper, and to contribute to loadings of iron, lead, mercury, and zinc (Frithsen et al. 1995a). Atmospheric deposition was the only pathway that contributed a measurable amount of PCBs and it also contributed to chlordane, DDT, and dieldrin loadings. On-going monitoring in Tampa Bay by the Tampa Bay NEP and local governments will better define the spatial distribution of atmospheric deposition of pollutants throughout the watershed. This effort will also aid in determining the relationship between emission sources (local, regional, or global) of airborne pollutants and their subsequent deposition in Tampa Bay.

An ongoing cooperative study will conduct air transport and deposition monitoring and modeling to help respond to two issues: (1) how much of the nitrogen and toxic

TABLE IV-13
Atmospheric Deposition to NEP Waters

Waterbody	Pollutants of Concern Evaluated[a]	Brief Summary of Results Related to Atmospheric Deposition of Pollutants of Concern	Reference
Massachusetts Bays (MA)	Nitrogen, PAHs, PCBs, cadmium, lead, mercury	Direct atmospheric loadings were estimated: nitrogen, 5-16%; PAHs, 10-40%; PCBs, 85%; cadmium, 17-31%; lead, 39-45%. Permitted wastewater discharges account for higher load of nitrogen and PAHs.	Menzie-Cura & Associates 1991
	Nitrogen	Direct atmospheric deposition contributes 6-8% of nitrogen load.	Zemba 1995
Narragansett Bay (RI)	PCBs, PAHs	PAH and PCB concentrations differ seasonally; wet deposition accounts for 50% of deposition of both pollutants; atmospheric deposition not a significant contributor of PCBs (3%) but is for PAHs (12%).	Latimer 1996
Long Island Sound (NY/CT)	Nitrogen	Direct atmospheric deposition is a significant source of nitrogen (10%); nitrogen deposition to watershed also considered a major source.	LIS 1994; Miller et al. 1993
Peconic Bay (NY)	Nitrogen	Atmospheric deposition estimated to contribute 5% of nitrogen load to Peconic River and Flanders Bay.	Suffolk County 1992
New York-New Jersey Harbor Estuary and Bight (NY/NJ)	Cadmium, lead, mercury, PCBs, dioxins, PAHs, various pesticides, nitrogen	Atmospheric deposition found to contribute significant portion of total pollutant loading for lead and nitrogen.	NY-NJ Harbor NEP 1995
Delaware Bay (DE/NJ/PA)	Nitrogen	Atmospheric deposition found to account for 14% of dissolved inorganic nitrogen loading; seasonal component noted.	Scudlark and Church 1993
	Lead, mercury, PCBs, various pesticides, and VOCs	Atmospheric deposition found to be a significant source of mercury (80%) and PCBs (34%). For lead, atmospheric deposition contributed less than 5%. For the remaining pollutants, atmospheric deposition was either insignificant or estimates were not developed.	Frithsen et al. 1995b
Delaware Inland Bays (DE)	Nitrogen	Tributaries are largest source of nitrogen loading, followed by atmospheric deposition; in summer, atmospheric deposition can be largest source.	Cerco et al. 1994

TABLE IV-13
Atmospheric Deposition to NEP Waters
(continued)

Waterbody	Pollutants of Concern Evaluated[a]	Brief Summary of Results Related to Atmospheric Deposition of Pollutants of Concern	Reference
Tampa Bay (FL)	Nitrogen	Atmospheric deposition accounts for 27% of current mean annual nitrogen loading.	Zarbock et al. 1994
	Cadmium, lead, mercury, chlordane, DDT, dieldrin, PCBs, PAHs	Atmospheric deposition found to be significant contributor of PCBs, cadmium, and lead.	Frithsen et al. 1995a
	Cadmium, lead, mercury, chlordane, DDT, dieldrin, PCBs, PAHs	On-going study - no results yet.	U.S. EPA 1995g
Galveston Bay (TX)	PAHs, PCBs, selected pesticides, lead, cadmium, mercury, and nitrogen	On-going study - no results yet.	U.S. EPA 1995h
Tampa Bay (FL) and Galveston Bay (TX)	Nitrogen	Nitrogen loads from atmospheric deposition are 67% (largest source) in Tampa Bay and 49% (point sources 48%) in Galveston Bay.	Patwardhan and Donigian 1996
Santa Monica Bay (CA)	PAHs, metals, nitrogen, chlorinated organics	Atmospheric deposition a significant source of lead and PAHs.	SMBRP 1994

[a] Pollutants other than those on the Great Waters list of pollutants of concern may have been evaluated; see reference for further details.

pollutants emitted annually from sources within the Tampa Bay watershed are subsequently deposited in the Bay or its watershed; and (2) what are the relative contributions of remote versus local sources of nitrogen and toxic pollutants to deposition in the watershed. Projects that have been initiated to address elements of the deposition program include:

- Initiation of intensive daily wet and dry deposition monitoring for measurement of nitrogen and toxic pollutants;
- Application of a regional air mass movement model focused on the Tampa Bay area (with Florida State University Center for Tropical Meteorology);
- Integration of NOAA's Physical Oceanographic Real-Time System (PORTS) meteorological data collected at several in-bay stations (with NOAA and the University of South Florida); and
- Initiation of stormwater collection at several gauged sub-basins co-located with the intensive deposition site (with local county stormwater divisions and the Florida Department of Transportation).

An important element of the Tampa Bay atmospheric deposition program is the participation and coordination of local and federal government programs and state agencies with the Great Waters program and the Tampa Bay NEP.

Galveston Bay. The Great Waters program conducted a screening atmospheric deposition monitoring program in Galveston Bay, Texas, which was chosen as the site to establish monitoring for the Texas Regional Integrated Atmospheric Deposition Study (TRIADS) as a representative of a Gulf of Mexico estuary. Monitoring at the TRIADS site began in February 1995. To facilitate comparability, the sampling and analytical design of TRIADS is similar to that of existing Great Waters program sites in the Great Lakes and Chesapeake Bay. The goals of this study are to evaluate the contribution of atmospheric deposition of selected contaminants to the Bay and to evaluate long-range transboundary transport of contaminants. Pollutants measured include PAHs, PCBs, selected pesticides, lead, cadmium, mercury, and nitrogen. Results from TRIADS complement and add to data from other investigations in Galveston Bay, including studies by the Galveston Bay NEP, EPA's Environmental Monitoring and Assessment (EMAP) program, NOAA National Status and Trends (NS&T) program, and special urban-pollutant studies in Houston, Texas. Data from these programs and the TRIADS data will be used to estimate the cumulative, direct and indirect impacts of atmospheric deposition to pollution of Galveston Bay.

Although atmospheric deposition has been shown to contribute a large percentage of the nitrogen load (49 percent) to Galveston Bay (Patwardhan and Donigian 1996), this loading is not expected to be a significant concern for Galveston Bay because, in general, the Bay is not as sensitive as other estuaries, such as Chesapeake Bay and Tampa Bay, to nitrogen inputs. For example, all three of these estuaries have experienced declines in submerged aquatic vegetation (SAV); however, in Galveston Bay this vegetation is not as important a component of the ecosystem as it is in Chesapeake Bay and Tampa Bay, and unlike the other two Bays the decline of SAV in Galveston Bay does not appear to be due primarily to nutrient loading (though it is a possible factor). In addition, while Chesapeake Bay has a significant baywide problem with hypoxia attributable to excessive nutrient loads, Galveston Bay's hypoxia problems are very localized and are influenced by factors other than nutrient loading (e.g., biological oxygen demand loading).

In contrast to nitrogen, research has indicated that atmospheric deposition of toxic pollutants may be a significant contributor to potential health risks. One study documented the presence of PAHs, PCBs, pesticides, mercury, lead, and dioxin/furans in fish and shellfish from Galveston Bay (Brooks et al. 1992). The study was not able to determine the source of these pollutants, but ancillary data suggested that atmospheric deposition may be a significant source. During the present monitoring program, all of the contaminants listed above have been detected in atmospheric samples collected from the TRIADS site. Continued monitoring will enable scientists and managers to more fully evaluate this problem and determine the relative effect of atmospheric deposition versus point and nonpoint source inputs into Galveston Bay.

Casco Bay. The primary pollutants of concern for atmospheric deposition to Casco Bay include PAHs, PCBs, nitrogen, phosphorus, sulfates, pesticides, and mercury and other trace metals. Recent sediment studies have found elevated concentrations of some pollutants (i.e., PAHs, PCBs, lead, mercury, silver, zinc, and cadmium) near population centers and waste discharges, but also observed elevated levels in rural eastern Casco Bay away from these known sources (Wade et al. 1995). A circulation model study of the Bay did not clearly indicate any possible sources for these pollutants, suggesting atmospheric deposition as a significant source (Gong et al. 1995). While elevated levels of lead found in Casco Bay sediments were relatively near potential sources, elevated levels of cadmium were found far from any known local source. A deposition study would provide empirical verification of processes occurring at Casco Bay.

Massachusetts Bays. As part of a Massachusetts Bays NEP study, Menzie-Cura & Associates (1991) estimated that direct atmospheric deposition contributes 5 to 16 percent of the total nitrogen load to Massachusetts Bays. In another Massachusetts Bays NEP study, direct atmospheric deposition of nitrogen was estimated to account for 6 to 8 percent of total nitrogen loadings to the Bays (Zemba 1995). Different methodologies were used to estimate nitrogen loadings in these two studies. The estimate made by Zemba (1995) is more precise and was made using literature values and ten years (1981-1991) of wet deposition data from the National Atmospheric Deposition Program (NADP).

Atmospheric deposition is also a significant contributor of organic compounds and trace metals to Massachusetts Bays. Menzie-Cura (1991) estimated that direct atmospheric deposition was a significant source of PAHs (10-40 percent), PCBs (85 percent), cadmium (17-31 percent), and lead (39-45 percent). A subsequent Massachusetts Bays NEP study generally corroborated the Menzie-Cura (1991) metal deposition results, although lead deposition rates were slightly lower (Golomb et al. 1995). The lower lead deposition estimates may be due to the fact that data used in the Menzie-Cura study were obtained prior to the phase out of leaded gasoline. Golomb (1995) also indicated that PAH deposition may have been underestimated and that PCB concentrations were below detection limits and, therefore, atmospheric deposition rates for PCBs were not calculated. Because PCB concentrations were below the detection limit, more precise field measurements of wet and dry deposition of PCBs are necessary to verify the initial estimates and to determine the relative impact of atmospheric deposition of PCBs to Massachusetts Bays.

Delaware Bay. In Delaware Bay, studies have shown that direct and indirect atmospheric deposition provide 14 percent of the annual nitrogen input, increasing to 25 percent in late spring and early summer (Scudlark and Church 1993). The relative nitrogen loading is slightly lower than observed in nearby Chesapeake Bay (27 percent), and much lower than in the Delaware Inland Bays (Rehoboth and Indian River Bays) where direct atmospheric deposition alone contributes 27 percent of the total nitrogen load (Cerco et al. 1994). The contribution to the Delaware Bay is lower because of higher point source nitrogen loading to Delaware Bay and the influence of a highly urbanized watershed.

As part of a Delaware Estuary Program study to estimate contaminant inputs, atmospheric deposition was found to be a significant source of mercury (80 percent) and PCBs (34 percent) (Frithsen et al. 1995b). As is the case in other regions, more research is warranted on atmospheric inputs of mercury and the resulting effects on estuarine and human health. To evaluate the effect atmospheric deposition of mercury has on the Delaware, Rehoboth, and Indian River estuaries, a precipitation monitoring station was established at Lewes, Delaware, in 1995 in conjunction with EPA's National Atmospheric Deposition Program Mercury Deposition Network.

Long Island Sound. A chronic problem in Long Island Sound is the low oxygen levels (hypoxia) that are observed during the summer. A 1994 study noted that excess nitrogen loading was a major cause of hypoxia and estimated that atmospheric loading directly to the water surface contributed about 10 percent of the total nitrogen delivered to the Sound (LIS 1994). Using direct measurements from two sites along the Connecticut shore, Miller et al. (1993) estimated atmospheric deposition rates. These data were used to refine the previous estimates and suggest that sources of nitrogen from atmospheric deposition (direct and indirect) may be responsible for 17 to 24 percent of the dissolved oxygen depression in the Sound. Further work is necessary to quantify the relationships between air quality, direct and indirect atmospheric deposition, and runoff concentrations to receiving waters of the Sound.

Peconic Bay. Nitrogen from atmospheric deposition to the Peconic River and Flanders Bay is estimated to be 160 lbs/day, or about 5 percent of the total nitrogen loading to that area (Suffolk County 1992). The impact of atmospheric deposition on eutrophication in Peconic River and Flanders Bay is considered to be relatively small in relation to other point and nonpoint sources. Atmospheric deposition is believed to be much more significant in terms of relative eutrophication impacts to Peconic Estuary surface waters east of Flanders Bay. Detailed loading estimates for these eastern areas, as well as for specific subwatersheds, are being developed for the Peconic NEP waters, as is an assessment through computer modeling of the relative eutrophication impacts of sources.

FUTURE RESEARCH NEEDS IN NEP WATERS

Research on atmospheric deposition to coastal waters has been very limited, and most studies have focused on identifying and determining the concentration of pollutants of concern in water and sediment, and measuring concentrations of pollutants in precipitation. Due to limited funding, many preliminary NEP studies are restricted to using historical data to estimate atmospheric deposition. Some NEP studies have used a rough mass-balance approach to determine the relative loading of each pollutant to the estuary, but quantitative mass balances requires accurate atmospheric data.

Establishing the total contribution of pollutants and their sources is an important part of developing and implementing CCMPs, and the lack of knowledge about the concentrations, deposition, and potential sources of airborne pollutants makes sound policy formation for the estuaries difficult. The question of the magnitude of pollutant deposition from the air has become more important as other sources of pollution to rivers, lakes, streams, and coastal waters have been identified and significantly reduced.

Research questions for the NEP estuaries include:

- What are the concentrations and loadings of pollutants that are being supplied by atmospheric deposition?
- What are the relative contributions of these inputs to the total load of pollutants entering the estuary?
- What are the locations of sources affecting the estuaries?
- Does atmospheric deposition (direct and indirect) of contaminants cause or contribute to biological harm in benthic (bottom-dwelling) or pelagic (swimming) communities and in turn affect human health?
- What economically and technically feasible methods are available to reduce airborne pollutants and their effects on estuaries?

The NEP estuaries provide an excellent opportunity to evaluate the effects and contribution of atmospheric deposition of contaminants to a varied set of ecological, environmental, and anthropogenic conditions. The NEP also provides a "grassroots" forum for addressing and correcting regional and national air quality issues as they pertain to our coastal waters. Recommendations for further atmospheric deposition research in coastal waters to help answer the above questions include:

- Utilize existing databases and ongoing work of established research programs and coordinate research initiatives with these programs;
- Protect and enhance existing monitoring programs;
- Establish long-term water and air quality monitoring programs that incorporate sampling for atmospheric deposition of contaminants for a subset of NEP estuaries representing various geographical regions and environmental conditions;
- Use sampling data from monitoring programs to track trends and spatial variability to develop more accurate loading estimates;
- Coordinate efforts between NEP estuaries and other Great Waters program studies to identify local, regional, and national sources of airborne pollutants;
- Pursue detailed atmospheric chemistry and deposition models for estimating atmospheric deposition to NEP estuaries;

- Develop a multi-party effort to identify and demonstrate appropriate pollution prevention techniques; and
- Apply existing atmospheric circulation models to fill in the data gaps between measured and estimated atmospheric deposition and to aid in tracing the pollutants in the estuaries back to their probable sources.

This research is needed not only to assist decision-makers, but to form a comprehensive picture of the atmospheric deposition problem across the United States. In addition, coordinated use of other mechanisms, such as voluntary pollution prevention, can help control the negative impacts of atmospheric deposition to water quality in NEP estuaries, especially at the local and regional level.

National Estuarine Research Reserve System

Another program established to recognize the importance of estuaries is the National Estuarine Research Reserve System (NERRS), which was created by Congress in 1972 under the Coastal Zone Management Act and which operates under the authority of NOAA. The mission of NERRS is to establish and manage, through the cooperation of federal, state, and community efforts, a national system of estuarine research reserves that are representative of various regions and estuary types in the United States, in order to provide opportunities for long-term research, education, and stewardship.

The process for designating and maintaining a NERRS site includes five main activities, all of which may be partially funded by NOAA: (1) the predesignation phase (includes selection of the site by the state and, after approval of the site by NOAA, preparation of a draft and final management plan and environmental impact study and completion of basic characterization studies); (2) acquisition of land and development activities; (3) after designation as an NERRS site, implementation of research, educational, and research programs detailed in the research reserve management plan; (4) estuarine research and monitoring; and (5) educational and interpretive activities.

Currently, 22 areas are designated as NERRS sites, including portions of Chesapeake Bay and associated lands in Maryland and Virginia (see Section IV.A for a detailed discussion of Chesapeake Bay). Six additional NERRS sites have been proposed or are in the beginning stages of development. See Figure IV-19 for the location of the NERRS estuaries.

No studies on atmospheric deposition to NERRS waters are available at this time (except for NERRS sites that are also NEP sites). However, research in other estuaries, such as NEP waters, is expected to be useful for research efforts in and protection of NERRS waters.

Gulf of Mexico Program

The Gulf of Mexico is a very important resource to all of North America. Its surface area is about 619,000 square miles, large enough to cover one-fifth of the continental United States. The U.S. portion of the Gulf's shoreline measures over 1,600 miles from the Florida Keys to the Rio Grande. Taking into account the shoreline length of all the bays, estuaries and other coastal features of the Gulf, its effective shoreline length is about ten times that amount. The 21 major estuaries along the Gulf coast account for 24 percent of all estuarine area in the 48 contiguous states, and 55 percent of the marshes. The watershed of the Gulf includes more than two-thirds of the continental United States (plus one-half of Mexico and parts of Canada, Guatemala, and Cuba), with the Mississippi River watershed alone draining about 40 percent of the continental United States.

The Gulf of Mexico Program (GMP) was established in 1988 in response to citizens' concerns over declines in the Gulf's fish, shellfish, and wildlife; the quality of life in many coastal communities; the need to protect the remaining valuable resources and prevent problems before they occur; and to forge a positive relationship between the ecological health and the economic vitality of the Gulf region. The GMP is a unique organization that involves representatives from government agencies (federal, state, and local), business and industry, non-profit organizations and educational institutions, and interested individuals in the process of setting environmental goals and implementing actions to achieve those goals. The aim is to foster synergy among these organizations in order to reduce costs and coordinate actions.

The GMP is not a regulatory program, but rather an approach to environmental protection, similar to the Chesapeake Bay Program, that is founded on the principles of:

- Partnership among government agencies, private, and non-government interests to define and characterize concerns and implement solutions;
- Sound science and information as the basis of informed decision-making to guide actions; and
- Public involvement to determine goals, identify solutions, and generate the consensus needed for action.

Since its beginning, the GMP has made significant progress in effectively involving a broad spectrum of the public in defining goals and objectives and in characterizing fundamental issues. The fundamental goals of the GMP are to:

- Protect human health and the food supply;
- Maintain and improve Gulf habitats that support living resources (fish, shellfish, and wildlife); and
- Maintain and enhance the sustainability of the Gulf's living resources.

In the past few decades, the Gulf of Mexico has been degraded, largely due to nutrient enrichment and habitat loss. The contribution of atmospheric nitrogen to nutrient enrichment is possibly the least understood and a significant concern. Fed by nutrient-enriched waters of the Mississippi River, a large area of near-bottom waters commonly become depleted in oxygen, or hypoxic. At its peak, this area (known as the "dead zone") can extend from the coastal waters of the Mississippi River Delta of Louisiana to those of eastern Texas. Stresses to the benthic (bottom-dwelling) community have been observed in this zone, including mortality of larger non-swimming benthic organisms. This and other possible disruptions to the food chain threaten to impact the commercial and recreational fish species within the hypoxic area. In addition to the Louisiana Shelf hypoxic zone, 18 other coastal areas in the Gulf have experienced hypoxia due to increasing nutrient concentrations or loads. Atmospheric deposition research relevant to the Gulf is discussed in the next section.

STUDIES OF ATMOSPHERIC DEPOSITION TO THE GULF OF MEXICO

At the current time, some basic estimates of the amount of atmospheric nitrogen deposited into the Mississippi Drainage Basin have been made using NADP data. Using these data, two different studies have estimated the nitrogen loading to the Gulf. Dr. Scott Dinnel of the University of Southern Mississippi calculates that, on an average basis, about 3.08 million tons per year of nitrogen is deposited from the atmosphere (Dinnel 1996). Donald A. Goolsby of the USGS rates the nitrogen inputs as follows: commercial fertilizer, 6.93 million tons; animal manure, 3.08 million tons; legumes (net input) 1.21 tons; domestic and municipal waste, 0.66 million tons; and atmospheric deposition, 0.55 million tons (Goolsby et al. 1996).

Hypoxia Conference

In response to the hypoxia problem in the Gulf of Mexico, a recent conference discussed the issue. Topics addressed included:

- Characterization of the hypoxic zone;
- Economic impacts and trends in fisheries attributable to the hypoxia;
- Causes of the hypoxic zone;
- Sources and delivery of nutrients in the watershed, including atmospheric deposition;
- Current efforts to control nutrient loads; and
- Information and policy required for action.

Source: GMP 1996.

There is significant discrepancy between the two estimates of nitrogen deposited to the watershed. Using Dinnel's figure, atmospheric nitrogen is essentially on par with animal manure as the second highest source of nitrogen input into the watershed, instead of being fifth in importance as indicated by Goolsby. These conflicting estimates point to the need for further and more refined estimates of nitrogen inputs to the Gulf.

IV.E Summary of the Major Waterbodies

Since the First Report to Congress, quantitative information has continued to be gathered on the atmospheric levels and deposition of pollutants in the Great Waters. Research has been conducted to provide waterbody-specific data on deposition and water quality, as well as to develop and improve measurement and modeling methods. This chapter confirms the findings of the First Report that, for studied Great Waters, toxic pollutants and excessive nitrogen may affect the environmental conditions of these waterbodies, with the potential to produce adverse ecological and economic impacts to the surrounding area. Moreover, the contribution of atmospheric deposition is significant for many waterbodies. For the freshwaters such as the Great Lakes and Lake Champlain, metals, organic compounds, and pesticides released into the atmosphere have been measured in significant quantities near these waterbodies, and their deposition measured or calculated. For the Chesapeake Bay and many other U.S. coastal waters, the impact is not only from toxic pollutant releases, but also eutrophication as a result of excessive inputs of nitrogen compounds. The data collected at these waterbodies address specific concerns related to known toxic pollutants, but cannot account for the impact of cumulative or combined effects of pollutants and may not protect against unregulated (and potentially more toxic) chemicals entering the waterbody.

In the **Great Lakes**, atmospheric deposition is an important contributor to toxic pollution to the waters. Fish consumption advisories still exist for a number of Great Waters pollutants of concern -- mercury, chlordane, HCB, PAHs, PCBs, and toxaphene. Both local and long-range emission sources are believed to contribute to atmospheric deposition in the Great Lakes. There are limited data on the relative contribution from specific emission sources, but information on the potential sources of contamination continues to be collected. Recent atmospheric monitoring results from IADN (the binational monitoring network assessing magnitude and trends of atmospheric deposition to the region) indicate that atmospheric contamination is still a significant concern in the Great Lakes. Such findings support conclusions from the First Report. More definitive information on pollutant trends is anticipated as additional data are collected over the next few years.

As described in Section IV.A, many of the projects being performed on the Great Lakes are state-of-the-art research that will not only benefit the Great Lakes system, but will also provide methodologies and data that can be applied to other waterbodies. The Lake Michigan Mass Balance Modeling study, initiated in 1994, is designed to evaluate the pollutant loadings and movements of toxic substances from all pathways to and within Lake Michigan. Comprehensive sampling is performed, with measurements of pollutants in the atmosphere, tributaries, lakewater, sediments, and food chain. Once analysis of the collected data is complete, modeling concepts and approaches developed in this study will be applied to future efforts in other waterbodies. The Great Lakes Emissions Inventory is also underway, collecting information on emission sources of HAPs in the eight Great Lakes states.

Water quality criteria have recently been developed for the Great Lakes to target pollution reduction and contamination controls. However, the reduction of atmospheric contamination in the Great Lakes requires nationwide and binational efforts because sources of air pollutants arise from other regions of the United States, as well as Canada. The United States and Canada recognize the importance of coordinating efforts to examine issues related to maintaining the natural resources of the Great Lakes system. For example, the Great

Lakes Water Quality Agreement between the United States and Canada has set a goal to virtually eliminate persistent pollutants from the Great Lakes. The Binational Virtual Elimination Strategy sets reachable objectives for achieving that goal through voluntary and regulatory efforts. Both nations encourage and support voluntary programs to reduce generation, use, and release of toxic contaminants to the Great Lakes.

Recent accomplishments in the Great Lakes -- gathering data on pollutant deposition, particularly through IADN, and developing emission inventories and modeling tools to determine potential sources of atmospheric pollution deposition -- support the mandate in section 112(m) to assess and identify atmospheric contamination in the Great Lakes. Reduction goals have been developed and, in some instances, have been implemented through voluntary efforts and regulations.

In **Lake Champlain**, atmospheric contamination of mercury and PCBs is of concern. There are fish consumption advisories for these pollutants and only a limited number of direct dischargers and other identifiable local sources. As presented in Section IV.B, research on atmospheric loading of mercury is currently underway in the basin. Early data show that mercury levels and deposition are comparable to those measured around the Great Lakes. No studies specifically address atmospheric deposition of other toxic pollutants in Lake Champlain; however, the States of New York and Vermont are currently performing more broad-scale programs to assess toxic contamination concerns within their states, including measurements of atmospheric pollutant levels from sites located around the Lake Champlain basin. The major program for responding to environmental issues in Lake Champlain is the Lake Champlain Basin Program (LCBP). This program addresses issues important to protecting the unique resources of Lake Champlain. A pollution prevention, control, and restoration plan for the Lake was recently developed by LCBP. One of the components of the plan is prioritizing strategies or efforts to reduce toxic contaminants through pollution prevention programs, and to address both science and policy issues. Although the Plan has yet to be implemented, it provides comprehensive goals for responding to toxic contamination in the Lake.

With an increasing focus on binational efforts, it is expected that greater results will be seen due to better cooperation between the United States and Canada. For Lake Champlain, research on characterizing atmospheric deposition of mercury, a major pollutant of concern in the basin, has been a major focus. Strategies to achieve toxic pollutant reductions in the lake have been proposed by LCBP, and implementation of these recommendations are anticipated in the future with federal, state, local, and/or private involvement.

As discussed in Sections IV.C and IV.D, nitrogen and toxic contaminants are a concern in the **Chesapeake Bay** and **other coastal waters**. As discussed in the First Report, excessive nitrogen loading can accelerate eutrophication and its adverse effects, such as nuisance algal blooms and fish kills. Chesapeake Bay, the largest U.S. estuary, has developed a strategy for reducing the nitrogen load to the Bay and has made extensive progress in the modeling and understanding of the type and geographic origin of airborne nitrogen to the Bay. Significant data has also been collected on rates and amounts of nitrogen deposition (including direct versus indirect deposition, wet versus dry deposition), and models have been developed to evaluate the impact of several nitrogen reduction scenarios on the Bay's water quality. The modeling of nitrogen reduction scenarios shows that implementation of Clean Air Act and other controls on atmospheric nitrogen emissions can significantly improve Bay water quality, beyond the improvments realized from controlling water discharges.

Since the First Report, studies of several other coastal wasters, at National Estuary Program waters in particular, have investigated the significance to their waters of atmospheric deposition of nitrogen compounds. As shown in Table IV-9 and discussed in Section IV.D, atmospheric deposition can contribute a significant portion of the nitrogen load to coastal waters -- from 2 to 80 percent. To improve understanding and reduction of nitrogen deposition to Chesapeake Bay and other coastal waters, the Chesapeake Bay Program, various National Estuary Programs, and the Gulf of Mexico Program continue to develop and refine modeling and monitoring efforts by addressing uncertainties such as nitrogen retention in watersheds, the differences in transport and fate of various nitrogen compounds, and the contribution of nearshore ocean waters to the nitrogen inputs to estuaries.

Toxic contaminants also are a concern in the Chesapeake Bay and other coastal waters, as evidenced by the fish advisories in effect for several pollutants of concern in many coastal areas. The Chesapeake Bay Program has developed a strategy for addressing the reduction of toxic contamination to the Bay. Continued monitoring of several pollutants on the Bay's list of toxics of concern has measured air and precipitation concentrations of these pollutants and their depositional fluxes (both wet and dry). Through these measurements and emissions data, atmospheric loadings have been estimated. A comparison of these loadings to data from the Great Lakes is presented in Figure IV-18. Recent studies of several other coastal waters have investigated the significance of atmospheric deposition to toxic pollutant loadings, and have shown that atmospheric deposition can contribute a significant portion of pollutant loadings to coastal waters such as for lead, mercury, PAHs, and PCBs.

Continued monitoring efforts in Chesapeake Bay and other coastal waters are needed to better identify the toxic contaminants that are a concern for atmospheric deposition. Other uncertainties to be addressed include quantifying the contribution of dry deposition to the total pollutant load from the atmosphere, determining the mechanisms and contribution of pollutants by transport through the watershed, and identifying pollutant sources.

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APPENDIX B
FISH CONSUMPTION ADVISORIES

Table B-1 presents detailed fish advisory information for the Great Lakes, Lake Champlain, and Chesapeake Bay and other selected coastal waters. This information was used to develop the fish advisory tables in Section III.A of this report. For each waterbody, Table B-1 presents the state issuing the advisory, the relevant pollutants of concern, the species affected by the advisory, and the advisory type. The four advisory types are:

- Advisories to the general population to not consume particular species;
- Advisories to subpopulations potentially at risk (e.g., pregnant or nursing women, small children) to not consume particular species;
- Advisories to the general population to restrict the size and frequency of meals of particular species; and
- Advisories to subpopulations potentially at risk to restrict the size and frequency of meals of particular species.

Advisories for a particular waterbody include advisories for the entire waterbody (e.g., Lake Huron), portions of the waterbody (e.g., Saginaw Bay), and major tributaries (e.g., Saginaw River). Consequently, the advisories reported in Table B-1 do not necessarily apply to the entire waterbody. Table B-2 lists the names of the waterbodies, waterbody portions, and tributaries included in Table B-1.

TABLE B-1
Fish Consumption Advisories in the Great Waters

Waterbody[a]	State	Pollutant	Fish Species[b]	Advisory Type[c]
Great Lakes				
Lake Erie	MI	PCBs	Carp, catfish, white bass > 11"	NCGP
	NY	PCBs	Carp	NCGP, NCSP
	OH	HCB, PAHs, PCBs	All species	NCGP
		PCBs	Carp, channel catfish, chinook salmon > 19", coho salmon, freshwater drum, lake trout, smallmouth bass, steelhead trout, walleye, white bass, white perch	RGP
	PA	Chlordane, PCBs	Carp, channel catfish, lake trout	NCGP
Lake Huron	MI	Chlordane	Lake trout > 26" Lake trout	NCGP NCSP, RGP
		Dioxins	Lake trout < 26"	NCSP, RGP
		Dioxins, PCBs	Carp, channel catfish, lake trout > 26" All fish except banned species (i.e., those under NCGP advisory)	NCGP RGP, RSP
		PCBs	Brown trout > 21", lake trout, rainbow trout	NCSP, RGP

TABLE B-1
Fish Consumption Advisories in the Great Waters (continued)

Waterbody[a]	State	Pollutant	Fish Species[b]	Advisory Type[c]
Lake Michigan	IL	Chlordane, PCBs	Alewife, brown trout > 23", carp, catfish, chinook salmon > 32", lake trout > 23"	NCGP
			Brown trout < 23", chinook salmon 21-32", coho salmon > 26", lake trout 20-23"	NCSP, RGP
	IN	PCBs	Fish	NCGP
			Brown trout < 23", chinook salmon 21-32", coho salmon > 26", lake trout 20-23"	NCSP, RGP
	MI	Chlordane	Carp, lake whitefish > 23"	NCGP
		Dioxins	Carp > 30"	NCGP
			Carp < 30"	RGP, RSP
		Mercury	Carp < 30", northern pike, redhorse sucker > 17", sturgeon, walleye	RGP, RSP
		PCBs	Brook trout > 15", brown trout, carp, catfish, chinook salmon > 25", lake trout > 23", largemouth bass, northern pike, rainbow trout > 22", smallmouth bass, splake trout > 16", sturgeon, sucker, walleye > 20", white bass	NCGP
			Fish	NCSP
			Fish except banned species (i.e., those under NCGP advisory)	RGP
			Carp < 30"	RSP
	WI	Chlordane	Lake trout > 23"	NCGP
			Lake trout 20-23"	NCSP, RGP
		Mercury	Rock bass < 10", sturgeon > 50", walleye 15-26"	NCSP, RGP
			Walleye < 18"	RSP
		PCBs	Brook trout > 15", brown trout > 12", carp, catfish, chinook salmon > 25", drum, lake trout > 23", northern pike > 28", rainbow trout > 22", splake trout > 16", walleye > 15", white bass	NCGP
			Brown trout < 23", bullhead catfish, chinook salmon 21-32", coho salmon > 26", lake trout 20-23", northern pike, splake trout < 16", walleye 15-18", white sucker	NCSP, RGP
Lake Ontario	NY	Dioxins, PCBs	Fish	NCGP
			Fish	NCSP
			Brown trout < 20", coho salmon < 21", rainbow trout < 25", white perch, white sucker	RGP
		PCBs	Channel catfish	RGP

TABLE B-1
Fish Consumption Advisories in the Great Waters (continued)

Waterbody[a]	State	Pollutant	Fish Species[b]	Advisory Type[c]
Lake Superior	MI	Chlordane, mercury, PCBs, toxaphene	Ciscowet	NCGP
		Dioxins	Lake trout	NCSP, RGP
		PCBs	Lake trout > 30"	NCGP
			Brown trout > 21", carp, lake trout	NCSP, RGP
	MN	Dioxins	Carp 20-25"	NCGP
			Carp 5-15", northern pike 15-30", walleye 5-30"	RGP
		Mercury	Channel catfish 20-25", walleye 15-30"	NCSP
			Black bullhead catfish 5-15", black crappie 5-15", carp 5-15", channel catfish 5-25", northern pike 5-30", redhorse 15-20", rock bass 5-15", shorthead redhorse 5-20", smallmouth bass 5-15", walleye 5-30", white sucker 5-20", yellow perch 5-15"	RGP, RSP
			Bluegill sunfish 5-15", pumpkinseed sunfish 5-15"	RSP
		PCBs	Carp 15-25", ciscowet > 20"	NCGP
			Brown trout, chinook salmon, coho salmon, ciscowet < 20", lake herring, lake trout, lake whitefish, rainbow trout	RGP
	WI	Chlordane, PCBs	Ciscowet > 20"	NCGP
		Mercury	Walleye 26-30"	NCGP
			Walleye 18-26"	NCSP, RGP
			Walleye < 18"	RSP
		PCBs	Lake trout > 30"	NCGP
Connecting Channels (Great Lakes)				
Clinton River	MI	PCBs	Carp	NCSP, RGP
Detroit River	MI	Mercury	Freshwater drum > 14"	RGP, RSP
		PCBs	Carp	NCGP
Lake St. Clair	MI	Mercury	Muskellunge, sturgeon	NCGP
			Bluegill sunfish > 8", brown bullhead catfish > 14", carpsucker > 18", freshwater drum > 14", largemouth bass, northern pike > 26", rock bass > 8", smallmouth bass > 18", walleye > 20", white bass > 13", white perch > 10"	RGP, RSP
		PCBs	Channel catfish > 22"	NCGP
			Carp > 22"	NCSP, RGP
Niagara River	NY	Dioxins, PCBs	American eel, brown trout > 20", carp, channel catfish, chinook salmon, coho salmon > 21", lake trout, rainbow trout > 25", white perch	NCGP
			American eel, brown trout, carp, channel catfish, chinook salmon, coho salmon, lake trout, rainbow trout, smallmouth bass, white perch, white sucker	NCSP
			Brown trout < 20", coho salmon < 21", rainbow trout < 25", smallmouth bass, white sucker	RGP
		PCBs	Carp	RGP

TABLE B-1
Fish Consumption Advisories in the Great Waters (continued)

Waterbody[a]	State	Pollutant	Fish Species[b]	Advisory Type[c]
Rouge River	MI	PCBs	Carp, catfish, largemouth bass, northern pike, smallmouth bass, white sucker All fish All fish except banned species (i.e., those under NCGP advisory)	NCGP NCSP RGP
St. Clair River	MI	Mercury	Freshwater drum > 12"	RGP, RSP
		PCBs	Carp Gizzard shad > 10"	NCGP NCSP, RGP
St. Lawrence River	NY	Dioxins, PCBs	Fish Fish Brown trout < 20", coho salmon < 21", rainbow trout < 25", white perch	NCGP NCSP RGP
St. Mary's River	MI	Mercury	Walleye > 19"	RGP, RSP
Lake Champlain				
Lake Champlain	NY	Mercury	Walleye > 19"	NCSP, RGP
		PCBs	American eel, lake trout > 25"	NCSP, RGP
	VT	Mercury	Walleye	NCSP, RGP
		PCBs	Lake trout > 25"	NCSP, RGP
Selected Coastal Waters				
Chesapeake Bay	DC	Chlordane, PCBs	American eel, carp, channel catfish	RGP
	MD	Chlordane	American eel, black crappie, carp, channel catfish	RGP
Delaware Bay	DE	PCBs	Channel catfish, striped bass, white catfish, white perch Channel catfish, striped bass, white catfish	NCGP RGP, RSP
	PA	Chlordane, PCBs	American eel, channel catfish, white perch	NCGP
Galveston Bay	TX	Dioxins	Blue crab, catfish	NCSP, RGP
Long Island Sound	CT	PCBs	Bluefish > 25", striped bass	NCSP, RGP
Narragansett Bay	RI	PCBs	Bluefish > 25", striped bass	NCSP, RGP
NY/NJ Harbor	NJ	Chlordane, PCBs	Crustaceans, fish, shellfish American eel, bluefish, striped bass, white catfish, white perch	NCGP, NCSP NCSP, RGP
		Dioxins	Crustaceans, fish, shellfish	NCGP, NCSP
	NY	PCBs	All fish, american eel All species, snapping turtle	NCGP NCSP, RGP

TABLE B-1
Fish Consumption Advisories in the Great Waters (continued)

Waterbody[a]	State	Pollutant	Fish Species[b]	Advisory Type[c]
San Francisco Bay	CA	Dioxins, DDT and other pesticides, mercury, PCBs	Striped bass > 35" Sharks > 24", striped bass > 27" All fish except anchovy, herring, salmon, and smelt	NCGP NCSP RGP, RSP

[a] The advisories for each waterbody include advisories for the entire waterbody, portions of the waterbody, and major tributaries (including tributaries into which migratory species enter).

[b] In some cases, different parts of a waterbody have the same advisory except that the advisory applies to a different subset of a species (e.g., lake trout < 23" versus lake trout < 12"). In these cases, the more conservative advisory (i.e., the one that includes a larger universe of fish (e.g., lake trout < 23") is reported in the table.

[c] NCGP = advises against consumption by the general population
NCSP = advises against consumption by subpopulations potentially at risk (e.g., pregnant or nursing women, small children)
RGP = advises the general population to restrict size and frequency of meals of the particular species
RSP = advises subpopulations potentially at risk to restrict size and frequency of meals of the particular species

Source: U.S. EPA 1995b.

TABLE B-2
Names of Waterbodies Included in Table B-1

Waterbody	Waterbodies Included for Fish Advisory Data
Lake Erie	Lake Erie
	Maumee River
	River Raisin
	Black River
	Cuyahoga River
	Ashtabula River
	Buffalo River
Lake Huron	Lake Huron
	Saginaw River
	Saginaw Bay
Lake Michigan	Lake Michigan
	Manistique River
	Kalamazoo River
	Little Bay de Noc
	Lower Menominee River
	Green Bay
	Lower Fox River
	Grand Calumet River
	White Lake
Lake Ontario	Lake Ontario
	Oswego River
	18 Mile Creek
Lake Superior	Lake Superior
	St. Louis River
	Thunder Bay
Connecting Channels	St. Lawrence River
	St. Mary's River
	St. Clair River
	Clinton River
	Detroit River
	Lake St. Clair

TABLE B-2
Names of Waterbodies Included in Table B-1 (continued)

Waterbody	Waterbodies Included for Fish Advisory Data
Connecting Channels (cont'd)	Niagara River
	Rouge River
Lake Champlain	Lake Champlain
Chesapeake Bay	Anacostia River/Potomac River
	Back River
	Baltimore Harbor
	Lake Roland
Delaware Bay	Delaware Estuary
Galveston Bay	Houston Ship Channel
Long Island Sound	Long Island Sound
Narragansett Bay	All Rhode Island waters
NY/NJ Harbor	Hudson River
	Arthur Kill
	Kill van Kull
	Raritan Bay
	Harlem River
	East River
	Hackensack River
	Passaic River
	Raritan River
	Newark Bay
	Sandy Hook Bay
San Francisco Bay	San Francisco Bay delta region