United States
Environmental Protection
Agency

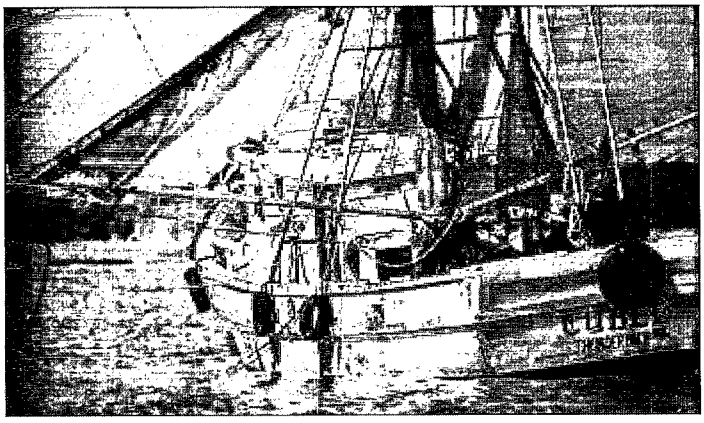
Office of Air Quality
Planning and Standards
Research Triangle Park, NC 27711

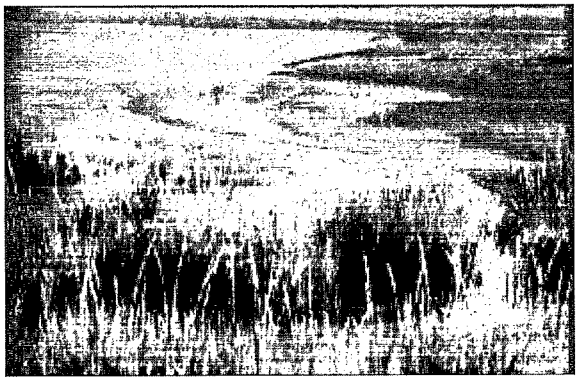
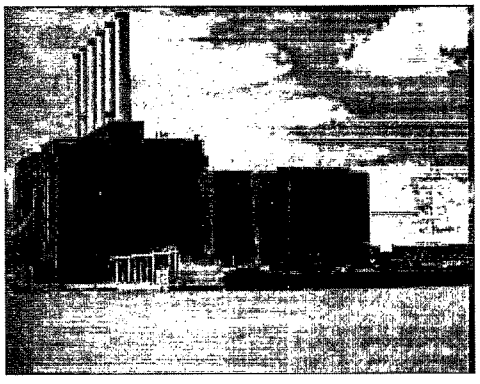
EPA-453/R-00-005
June 2000

EPA

Deposition of Air Pollutants to the Great Waters

Third Report to Congress



ACKNOWLEDGMENTS

The report was prepared by the Office of Air Quality Planning and Standards of the U. S. Environmental Protection Agency (EPA). The Project Leads were Gail Lacy and Dale Evarts. Completion of this report could not have been possible without significant contributions from the wider community of scientists and environmental experts involved with the programs and issues described in the report. In particular, EPA would like to thank those persons who provided peer review of the draft report:

Mary Ann Allan	Electric Power Research Institute, Palo Alto, California
Dr. Caroline Currin	National Marine Fisheries Service, Beaufort, North Carolina
Elisabeth Galarneau	Atmospheric Environment Service, Ontario, Canada
Dr. Louis Guillette	Department of Zoology, University of Florida
Dr. Michael Murray	National Wildlife Federation, Ann Arbor, Michigan
Dr. Hans Paerl	Institute of Marine Sciences, University of North Carolina
Dr. Joseph Scudlark	College of Marine Sciences, University of Delaware

In addition, EPA would like to acknowledge the substantive review and contributions provided by Richard Artz and Margaret Kerchner of the Air Resources Lab of the National Oceanographic and Atmospheric Administration, by Bob Larson of the National Atmospheric Deposition Program, and by many staff of State, local and tribal agencies.

Within EPA, we would like to acknowledge the contributions of the Agency work group members and others involved in the development of the report, including staff (too numerous to name) in the Offices Air and Radiation, Water, Research and Development, Solid Waste and Emergency Response, Prevention, Pesticides, and Toxic Substances, and General Counsel, as well as in the Great Lakes National Program Office, the Chesapeake Bay Program Office, the Gulf of Mexico Program Office, EPA Regions IV and V, and the National Estuary Program.

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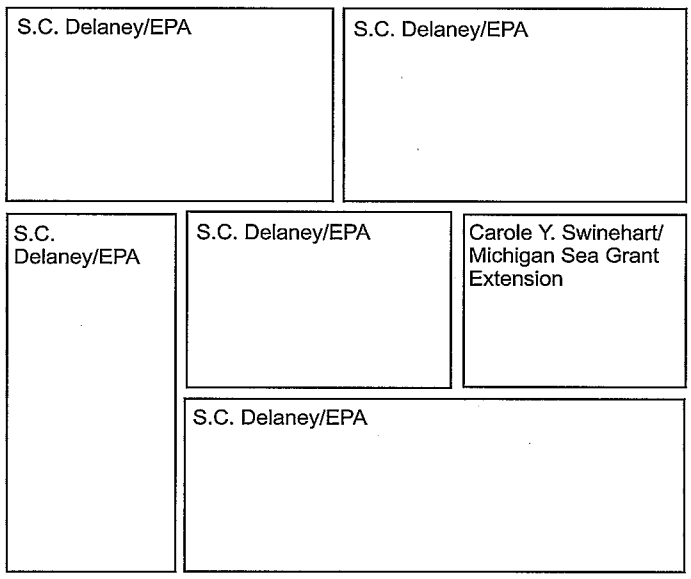

ERRATA

Page II-7, Table II-1, Rows 8 and 12:

"Chlor-alkali" row (Row 8) should be removed because it is a duplicate with Row 5, "Chlor-alkali Production." "Others (<1 percent each)" row (Row 12) should be revised to read 10 tons/year and 6 percent. The corrected table is included below.

Table II-1
National Anthropogenic Mercury Air Emissions
(Based on 1994-1995 Inventory; U.S. EPA 1997e)

Source Category	1994-1995 Anthropogenic Air Emissions (tons/year)	Percent Contribution to Total U.S. Anthropogenic Air Emissions
Utility boilers: coal combustion, oil, and natural gas	52	33
Municipal waste combustion	30	19
Commercial/industrial boilers: coal and oil	28	18
Medical waste incineration	16	10
Chlor-alkali production	7	4
Hazardous waste combustors	7	4
Portland cement, excluding hazardous waste-fired	5	3
Residential boilers: oil and coal	4	2
Pulp and paper manufacturing	2	1
Others (<1percent each) [a]	10	6
Total U.S. Anthropogenic Mercury Air Emissions	158[b,c]	100[b,c]

[a] A list of the source categories that contribute less than 1 percent to total U.S. emissions is provided in Appendix B.
[b] This value represents anthropogenic mercury air emissions in the U.S. only.
[c] Values do not add exactly due to rounding.

Page II-51, Paragraph 3, Sentences 4 and 5:

Replace original sentences with revised sentences provided below. Also, one new reference should be added, and one reference should be revised.

Original Text: "It should be noted, however, that ADN accounts for roughly only 1 percent of nitrogen loadings in the Mississippi River basin, which has highly diverse sources of anthropogenic nitrogen (Goolsby et al. 1998, as cited by Paerl 1999). The ADN is also

likely to be a relatively less important contributor to nitrogen loadings in Pacific coast Great Waters than is the case in the east and Gulf coast areas, due to the prevailing westerly winds over Pacific coastal waters and their watersheds, originating from unpolluted ocean areas."

Revised Text: "In the Mississippi River basin, ADN accounts for at least 10 percent of nitrogen loadings in the Mississippi River basin, which has highly diverse sources of anthropogenic nitrogen (Goolsby et al. 1999; Alexander et al. 2000). The ADN is likely to be a relatively less important contributor to nitrogen loadings in Pacific coast Great Waters than is the case in the east and Gulf coast areas, due to the prevailing westerly winds over Pacific coastal waters and their watersheds, originating from unpolluted ocean areas."

New Reference:

Alexander, R.B., R.A. Smith, and G.E. Schwarz. 2000. Effects of stream channel size on the delivery of nitrogen to the Gulf of Mexico. *Nature*. 403: 758-761.

Revised Reference:

Goolsby, D.A., W.A. Battaglin, G.B. Lawrence, R.S. Artz, B.T. Aulenbach, R.P. Hooper, D.R. Kenney, and G.J. Stensland. 1999. Flux and sources of nutrients in the Mississippi-Atchafalaya River Basin. Topic 3 Report for the Integrated Assessment on Hypoxia in the Gulf of Mexico: NOAA Coastal Ocean Program, Decision Analysis Series No. 17.

EXECUTIVE SUMMARY

With section 112(m) of the 1990 Clean Air Act (CAA), Congress directed the U.S. Environmental Protection Agency (EPA), in cooperation with the National Oceanic and Atmospheric Administration (NOAA), to identify and assess the extent of atmospheric deposition of air pollutants to the Great Lakes, the Chesapeake Bay, Lake Champlain, and coastal waters, collectively known as the Great Waters. Further, section 112(m) directed EPA to report its findings to Congress in periodic reports. This is EPA's third Report to Congress on the deposition of air pollutants to the Great Waters. The first report was published in May 1994, and the second report was published in June 1997.

The goals of the *Third Great Waters Report to Congress* are to discuss the current state of knowledge regarding atmospheric deposition of pollutants to the Great Waters based on new research and program activities undertaken since the *Second Report to Congress* and to describe any necessary revisions to requirements, standards, and limitations under the CAA or other Federal laws. This report is not intended to be a comprehensive summary of all relevant scientific research and activities. Instead, it summarizes and highlights major trends and key findings, and builds on conclusions presented in the *First* and *Second Reports to Congress*.

How does deposition of air pollutants affect public health and the health of the Great Waters ecosystems?

A rapidly growing number of atmospheric deposition monitoring and modeling studies confirm that, along with runoff and discharges of pollution into waterways, atmospheric deposition is a significant pathway of pollutant inputs to the Great Waters. These studies show that the contribution of atmospheric deposition to overall pollutant loadings varies greatly by pollutant and location. For example, studies show that atmospheric deposition contributes from less than 5 to 100 percent of dioxins and furans entering the Great Lakes, depending on the location of the monitoring site, and from 2 to 38 percent of the nitrogen load to certain coastal waters. Given this variability in contributions, the improvement of the quality of the Great Waters environments requires an understanding of all of the sources of the pollutants into a waterbody, including runoff from urban areas and farms, discharge from point sources, and seepage from contaminated sediments, as well as atmospheric deposition.

Great Waters Pollutants of Concern

Cadmium and cadmium compounds
Chlordane
DDT/DDE
Dieldrin
Hexachlorobenzene
α-Hexachlorocyclohexane
Lindane (γ-hexachlorocyclohexane)
Lead and lead compounds
Mercury and mercury compounds
Polychlorinated biphenyls
Polycyclic organic matter
Tetrachlorodibenzo-p-dioxin (dioxins)
Tetrachlorodibenzofuran (furans)
Toxaphene
Nitrogen compounds

Like the *First* and *Second Reports to Congress*, this report focuses on 15 pollutants of concern, including certain pesticides, metal compounds, chlorinated organic compounds, and nitrogen compounds (see sidebar). Some of these pollutants are single compounds while others represent categories of several or even hundreds of individual compounds. They are emitted into the air by a wide range of sources, including industries and other human activities, natural sources, and re-emissions of these pollutants from soil and water. What is known about their emission rates, concentrations in the environment, transformation processes, deposition rates and pathways, and health and environmental effects varies widely. Nevertheless, recent research has added to our knowledge of the adverse human health and ecological effects of these pollutants. At certain levels, these pollutants are associated with adverse

effects on many target organs in humans and animals, including the liver, kidney, nervous system, endocrine system, reproductive organs, and immunological system. Research since the *Second Report to Congress* provides additional evidence that some of the pollutants are likely "endocrine disruptors," meaning they may interfere with the action of hormones in wildlife and humans.

A number of regional and national assessments and monitoring programs described in this report suggest that, while environmental conditions are improving in general, the current concentrations of pollutants of concern continue to impair the ecological health of many Great Waters. For example, the NOAA Estuarine Eutrophication Surveys indicate that approximately 89 percent of the Great Waters that are coastal estuaries show some degree of adverse effects associated with amounts of nitrogen in excess of natural levels. (Nitrogen is a natural component of these waterbodies, but excess amounts can also be introduced through natural and human activities such as runoff from urban and agricultural areas and atmospheric deposition.) Because of long-range atmospheric transport and certain chemical properties (e.g., persistence, mobility, bioaccumulation, and bioconcentration), the pollutants of concern may contribute to ecological impairment far from known emission sources and long after releases.

At current levels of contamination, pollutants of concern in the Great Waters pose potentially the greatest health risks to individuals who consume fish from contaminated waters for subsistence or cultural reasons, women of child-bearing age, the developing fetuses of pregnant women, and young children who consume fish from contaminated waters. For mercury in particular, exposures do not appear to pose a health risk to people consuming average amounts of fish, but sensitive sub-populations (e.g., young children, and pregnant women and their developing fetuses) with higher than typical fish consumption may be at risk. Also at risk are subsistence fish-eating populations who consume large amounts of fish. The extent of risk for these groups depends on the amount of fish consumed and the mercury concentrations present in the fish.

What are the recent and anticipated trends in emission and deposition of Great Waters pollutants of concern and their concentrations in the Great Waters environment?

Where monitoring trends information exists, either nationally or locally, atmospheric deposition of pollutants of concern to the Great Waters has declined or remained relatively constant in recent years, as described in Chapter II. Specifically, deposition of lead, cadmium, polycyclic organic matter (POM) – which includes a group of compounds known as polycyclic aromatic hydrocarbons (PAHs) – polychlorinated biphenyls (PCBs), and some banned or restricted use pesticides is declining in the Great Lakes. Other Great Waters have also shown decreasing trends of some of the pollutants of concern, such as lead in Long Island Sound. Deposition of nitrogen in the U.S. has remained fairly constant. These trends may reflect the results of emission reduction programs established under the CAA, pesticide bans, as well as other local, tribal, State, Federal, and international pollution control efforts, many of which are described in this report. There is considerable uncertainty in the trends estimates, however, because limited monitoring capability, technological barriers, and variable collection and analysis methods make it difficult to adequately characterize historical or current conditions. In addition, these estimates do not address all Great Waters waterbodies for all pollutants of concern.

Emissions of mercury to the atmosphere come from human-made sources, natural emissions, and re-emission from biologic and geologic processes. Emissions of mercury have been on a downward trend since 1990, due chiefly to the phase-out of mercury in many products. Nevertheless, monitoring suggests that atmospheric deposition is a significant contributor of mercury to the Great Waters. At EPA's current reference dose for mercury, levels in some lakes and streams remain sufficiently high to pose adverse

human health and ecological risks and to result in fish consumption advisories for mercury in many Great Waters.

Lead air emissions, ambient air concentrations, and deposition levels in the Great Lakes region have also decreased in recent years. Cadmium emissions in the Great Lakes region, on the other hand, increased in the 1980s and have not shown a trend since then, while cadmium deposition in the Great Lakes region is believed to have decreased in recent years. In the Chesapeake Bay, trends in atmospheric deposition of lead and cadmium are difficult to discern with current information, but total inputs of both lead and cadmium in some eastern parts of the bay have increased in recent years due to the increase in population and industrial activities. In some areas of the Chesapeake Bay, levels of these metals may be high enough in sediments to cause adverse ecological effects.

Air emissions of dioxins, furans, and POM (including PAHs) in the U.S. are from both natural and human-made combustion and incineration processes. Emissions have declined for dioxins and furans, while trends for POM are not known. A large degree of variation has been observed in deposition levels of dioxins and furans over time within and between the Great Lakes. Deposition levels of these compounds are related to the pattern of industrialization and population density. In many instances, long term monitoring indicates that concentrations of dioxins and furans in biota in the Great Waters have declined over time. Analytical results indicated that levels in aquatic species are declining steadily and in several Great Waters, no dioxin was found in fish samples taken between 1987 and 1994.

Although CAA programs have had a major impact on nitrogen oxide (NO_x) emissions, the emission reductions have been balanced approximately equally with emission increases attributable to economic growth, resulting in a relatively flat trend since 1980. Deposition monitoring data suggest that the deposition rates of inorganic nitrogen (e.g., NO_x, ammonia) to many of the Great Waters watersheds have also been relatively constant for the past two decades, though some increases have been noted downwind of areas where population or livestock operations are growing. The EPA expects that additional NO_x controls to be implemented under the CAA will slightly outpace emission increases associated with economic growth, resulting in a net decreasing trend in NO_x emissions through 2005. Emissions are expected to remain steady at that level until 2010. Research is ongoing into the lesser known, but apparently important, forms of nitrogen deposition (e.g., dry deposition, organic nitrogen). Current and anticipated nitrogen deposition rates are significantly greater than natural rates, and combined with nitrogen inputs from runoff from farms and cities, have the potential to overwhelm the capacities of surface waters to assimilate the additional nitrogen.

Manufacture of PCBs in the U.S. no longer occurs; however, releases into the environment continue to occur because of PCBs in electrical transformers and capacitors that are still in use, releases from soils and sediments contaminated with PCBs, and releases during some combustion processes. The number and magnitude of PCB sources in the U.S. has decreased 20-fold in the past 20 years. Furthermore, deposition of PCBs in the Great Lakes has decreased and a net loss of PCBs has been observed in the Chesapeake Bay. Overall, PCB concentrations in the environment appear to have declined but are still present and continue to result in the need for fish consumption advisories in many Great Waters.

Based on recent academic research on atmospheric pollutant concentrations in the Great Lakes region, DDT and DDE, followed by dieldrin and chlordane, are estimated to fall below current detection limits in the atmosphere between 2010 and 2020. Hexachlorocyclohexane and hexachlorobenzene are projected to be eliminated in the atmosphere by 2030 and 2060, respectively. These estimates assume current rates of long-range transport of these pollutants into the region and do not mean that concentrations would be eliminated in deposited media (water and sediments) by these dates. However,

these estimates indicate that reduction strategies in the Great Lakes, along with the original bans and restrictions on the use of these substances, are having the intended effect.

What actions are EPA and others taking to address atmospheric deposition?

Chapter III of this report describes more than 60 programs under way from the local to international levels that directly or indirectly contribute to reducing atmospheric deposition of pollution to the Great Waters or to understanding its effects. Although EPA leads or supports many of these programs, which often use multimedia and cross-program approaches to control pollution, other Federal agencies, State and tribal organizations, industry groups, and Canada have also initiated and implemented many important activities. Examples of EPA's cross-program and multimedia approaches include the pulp and paper industry "cluster rule" that for the first time integrates, coordinates, and streamlines applicable requirements of the Clean Water Act and the CAA. The EPA's air and water programs are also working together to address the contribution of air deposition into water quality protection under total maximum daily load (TMDL) determinations. This also includes coordinated activities such as the Persistent Bioaccumulative Toxics Initiative, Clean Water Action Plan, and Contaminated Sediment Management Strategy. Several State organizations have successfully worked with industry and municipalities to develop pollution prevention programs and programs to collect banned and restricted use pesticides. Tribes are working to reduce exposure of indigenous populations to pollutants of concern through a variety of efforts, and industry has initiated efforts to reduce the use of pollutants of concern in products and processes. Canada has implemented a variety of programs, some of which are joint efforts with the U.S., like the Binational Toxics Strategy which seeks to eliminate pollutants of concern from the Great Lakes environment.

These coordinated Agencywide efforts demonstrate EPA's commitment to implement the strategic directions discussed in the *First* and *Second Reports to Congress* and in the section 112(m)(6) adequacy determination, and to pursue all authorities available for addressing atmospheric deposition and Great Waters pollutants of concern. In addition, developing and implementing these and other programs and initiatives described in this report have not required revisions to requirements, standards, and limitations in accordance with the CAA and other Federal laws which provide protection of human health and the environment from atmospheric deposition to the Great Waters.

What are EPA's findings and conclusions from this Third Report to Congress?

The new scientific and programmatic information presented in this report supports and builds on the three broad conclusions presented in the *First* and *Second Reports to Congress*.

- Atmospheric deposition from human activities can be a significant contributor of toxic chemicals and nitrogen compounds to the Great Waters. The relative importance of atmospheric loading for a particular chemical in a given waterbody depends on many factors, including characteristics of the waterbody, properties of the chemical, and the kind and amount of atmospheric or water discharges.

- A plausible link exists between emissions into the air of Great Waters toxic pollutants of concern, the atmospheric deposition of these pollutants (and their transformation products), and the concentrations of these pollutants found in water, sediments and biota, especially fish and shellfish. For mercury, fate and transport modeling and exposure assessments predict that the

anthropogenic contribution to the total amount of methylmercury in fish is, in part, the result of anthropogenic mercury releases from industrial and combustion sources increasing mercury body burdens (i.e., concentrations) in fish. Furthermore, the consumption of fish is the dominant pathway of exposure to methylmercury for fish-consuming humans and wildlife. However, what is known about each stage of this process varies with each pollutant (for instance, the chemical species of the emissions and its transformation in the atmosphere).

- Airborne emissions from local as well as distant sources, both within and outside the U.S., contribute pollutant loadings to waters through atmospheric deposition. Determining the relative roles of particular sources -- local, regional, national, and possibly global, as well as anthropogenic, natural, and re-emission of pollutants -- contributing to specific waterbodies is complex, requiring careful monitoring, atmospheric modeling, and other analytical techniques.

Actions taken by EPA and others to control sources of Great Waters pollutants of concern appear to have positively affected trends in pollutant concentrations measured in air, water, sediment, and biota. Overall deposition rates of pollutants of concern have declined slightly or remained constant. Several pollutants of concern continue to enter the Great Waters primarily through atmospheric deposition. In addition, long-range transport of pollutants of concern from other U.S. regions or other countries is estimated to contribute significantly to atmospheric loadings to the Great Waters. For example, the global reservoir of mercury (which includes mercury from both U.S. and foreign sources) is estimated to contribute about 40 percent of the total mercury deposition to U.S. lands and waters.

Concentrations of some pollutants of concern in the water, sediment, and biota of the Great Waters declined in recent years, whereas others were constant or variable. Concentrations of most pollutants of concern still pose potential adverse ecological and human health effects. For example, approximately 5 percent of the Nation's coastal and inland watersheds include "areas of probable concern," meaning a watershed that is associated with a certain number of monitoring sites with sediment contamination at levels likely to cause adverse effects. Water quality data also indicate that water quality standards in place for drinking water supplies in the Great Waters are not being exceeded for the pollutants of concern, but that surface water quality guidance and criteria are being exceeded for some of the Great Waters. In addition, nationally, fish consumption advisories were in place for 39 of 56 Great Waters waterbodies as of 1997.

Based on current trends, EPA expects atmospheric deposition to remain a significant source of several pollutants of concern to the Great Waters for the foreseeable future. In addition, because of the ability of these pollutants to persist and bioaccumulate, they are expected to remain in the water, sediments and biota for much longer.

Implementation of existing EPA regulations is expected to further reduce emissions of mercury, NO_x, POMs, dioxins and furans, cadmium, and hexachlorobenzene. The EPA continues to implement programs under CAA authorities and expects that pollutant emissions will be further controlled by several rules scheduled to take effect in coming years. As a result, atmospheric deposition and loadings of these pollutants may be significantly reduced. In addition, actions taken to voluntarily reduce chemical use, implement pollution prevention initiatives, advance technology (e.g., alternative fuel vehicles), and implement pollution control laws issued by States and other nations will further reduce pollutant loadings to the Great Waters.

What future directions will EPA follow to implement section 112(m)?

The EPA developed six key recommendations that will assist in meeting the objectives and requirements of the CAA related to the Great Waters program.

- The EPA will continue to support the maintenance and expansion of efforts to monitor Great Waters pollutants of concern in order to evaluate the relative contributions of local, regional, and long-range transport to deposition in the U.S., as well as natural versus human-made sources.

- The EPA will continue to develop and implement regulations and pollution prevention programs regionally and nationally, including multimedia programs, in order to reduce the impact of sources of Great Waters pollutants of concern within the U.S.

- For Great Waters pollutants emitted by sources outside the U.S., EPA will work within international frameworks to reduce sources of these pollutants.

- The EPA will support model development and research that establish and clarify the linkages from emissions to atmospheric deposition to waterbody loadings to adverse public health and the environmental effects of Great Waters pollutants of concern in order to enable effective risk management decisions.

- The EPA will encourage and support the establishment of common baselines and measures of progress in order to better assess trends and health of Great Waters and other waterbodies affected by atmospheric deposition.

- The EPA will work to increase public awareness of risks of exposure to Great Waters pollutants.

These key recommendations build on the strategic themes identified in the *First* and *Second Reports to Congress*: (1) continued implementation of the CAA to directly control emissions of Great Waters pollutants of concern; (2) use of an integrated multimedia approach throughout the Agency, including coordination of clean air programs with programs available under other Federal laws (e.g., Clean Water Act); and, (3) continued support of research activities that address the goals of the Great Waters program.

In support of the first strategic theme, EPA will develop or assess the need for new rules and programs under the CAA, including maximum achievable control technology (MACT) standards, section 112(c)(6) standards, as well as standards that could stem from the residual risk program, the utility air toxics determination, and the integrated urban air toxics strategy. As appropriate, EPA will consider the impacts of atmospheric deposition in developing standards under these authorities. In addition, EPA will ensure the timely implementation of NO_x control programs already in place and will encourage innovative, nonregulatory approaches to reducing NO_x emissions and other sources of atmospheric nitrogen.

This report describes a number of multimedia and cross-program initiatives (e.g., Clean Water Action Plan, Persistent Bioaccumulative Toxics Initiative) that are in line with EPA's second strategic theme for the Great Waters program. Some of these initiatives, such as the pulp and paper cluster rule, will produce tangible environmental benefits within the near future. Other initiatives, such as development of multimedia models in support of TMDL determinations and completion of the

Contaminated Sediment Non-point Source Inventory, will provide tools and data resources that will help EPA target future pollution control activities.

There has been substantial progress in research activities relevant to the Great Waters program since the *Second Report to Congress*. However, important information gaps remain, and there are critical limitations to current atmospheric monitoring and modeling capabilities. Consistent with the third strategic theme, therefore, EPA will initiate and continue to support scientific research to fill these critical gaps, including the following activities:

- Support research to examine and quantify the ecological effects of atmospheric deposition and to better quantify the water quality benefits of air pollution controls;
- Expand the geographic coverage and consistency of waterbody monitoring to enable more accurate characterizations of the extent of contamination and ecosystem effects due to atmospheric deposition;
- Encourage and support interagency coordination to better quantify the indirect loadings of atmospheric deposition to the Great Waters through the development of tools that can quantify watershed transport of pollutants of concern;
- Continue to support the development of modeling tools which address the transport and fate of pollutants in ecosystems and characterize risk, including research to clarify mechanisms of mercury methylation so as to better predict and manage ecosystems at risk;
- Develop reliable approaches for quantifying and monitoring nitrogen dry deposition and wet organic nitrogen deposition;
- Support joint work with States and industry to fill gaps in emissions information for MACT source categories and further refine emissions measurement methods, inventories, and modeling for Great Waters pollutants of concern;
- Support research on viable prevention and controls for sources of pollutants of concern;
- Encourage and support greater coordination and expansion of monitoring networks to assess deposition of pollutants to coastal waters, to assess the contribution of long-range transport to deposition in the U.S., and to evaluate the impact of agricultural and urban sources;
- Develop standard methods to monitor pollutants of concern to enable the comparison of data and trends analyses;
- Support international efforts to quantify the transboundary contributions of pollutants of concern and to share technology, information, and expertise with other countries on reducing releases to the environment and on cost-effective alternatives to their use;
- Working closely with other EPA and inter-governmental efforts to address, in particular, persistent bioaccumulative toxic pollutants, and to identify and evaluate additional pollutants which may be of concern to the Great Waters;
- Continue research to identify additional endocrine disrupting chemicals and their associated effects; and,

- Continue coordination and support of efforts to improve the consistency of fish consumption advisories and the awareness and understanding of advisories among populations most at risk to exposure to the Great Waters pollutants of concern.

The EPA is committed to continuing to address air deposition of pollutants into the Nation's waters as a priority matter. To that end, and to assure continued coordination of the many related tasks involved and outlined in this report, EPA will develop a detailed biennial work plan for implementation actions beginning this year and updated every two years. As EPA develops and implements plans, programs and initiatives with NOAA and its other Federal, State, tribal, industry and community partners, we expect to make significant, measurable progress toward our goal of assuring the protection of human health and the environment from adverse effects attributable to atmospheric deposition of pollution to the Great Waters.

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ABBREVIATIONS AND ACRONYMS

ADN	Atmospheric deposition nitrogen
AEOLOS	Atmospheric Exchange Over Lakes and Ocean Surfaces
AIEO	American Indian Environmental Office
AILESP	American Indian Lands Environmental Support Project
AIRMoN	Atmospheric Integrated Research Monitoring Program
AOC	Areas of concern
APC	Areas of probable concern
AQC	Air Quality Committee
ARL	Air Resources Laboratory
ATSDR	Agency for Toxic Substances and Disease Registry
BACT	Best available control technology
BASMAA	Bay Area Stormwater Management Agencies Association
BNS	Binational Toxics Strategy
BRACE	Bay Regional Atmospheric Chemistry Experiment
CAA	Clean Air Act
CASTNet	Clean Air Status and Trends Network
CBADS	Chesapeake Bay Atmospheric Deposition Study
CBEP	Community-Based Environmental Program
CBP	Chesapeake Bay Program
CBPO	Chesapeake Bay Program Office
CEC	Commission on Environmental Cooperation
CENR	Committee on Environment and Natural Resources
CHPAC	Children's Health Protection Advisory Committee
CMAQ	Community multi-scale air quality
CTDEP	Connecticut Department of Environmental Protection
CVAFS	Cold vapor atomic fluorescence spectrometry
CWA	Clean Water Act
CZM	Coastal zone management
DC	District of Columbia
DDD	1,1'-(2,2-dichloroethylidene)bis(4-chlorobenzene)
DDE	1,1'-(dichloroethenylidene)bis(4-chlorobenzene)
DDT	1,1'-(2,2,2-trichloroethylidene)bis(4-chlorobenzene)
DIN/SRP	Dissolved inorganic nitrogen to soluble reactive phosphorus
DNREC	Department of Natural Resources and Environmental Conservation
EAGLE	Effects on Aboriginals from the Great Lakes Environment
EDSTAC	Endocrine Disruptor Screening and Testing Advisory Committee
EEGLE	Episodic Events/Great Lakes Experiment
EGU	Electric generation units
EMAP	Environmental Monitoring and Assessment Program
EPCHC	Environmental Protection Committee of Hillsborough County
EOM	Extractable organic matter
EPA	Environmental Protection Agency
EPRI	Electric Power Research Institute
FAMS	Florida Atmospheric Mercury Study
FCP	Fish Contamination Program
FDEP	Florida Department of Environmental Protection
FIFRA	Federal Insecticide, Fungicide, and Rodenticide Act
FIP	Federal implementation plan
FR	Federal Register

g	Gram
GIS	Geographic Information System
GLNPO	Great Lakes National Program Office
GLWQA	Great Lakes Water Quality Agreement
GMPO	Gulf of Mexico Program Office
GPRA	Government Performance and Results Act
HAP	Hazardous air pollutant
HCB	Hexachlorobenzene
HCH	Hexachlorocyclohexane
HMIWI	Hazardous/medical/infectious waste incinerators
HNO_3	Nitric acid
HYSPLIT	Hybrid single particle Lagrangian integrated trajectory
IADN	Integrated Atmospheric Deposition Network
IAQAB	International Air Quality Advisory Board
IBI	Index of Biotic Integrity
IDEM	Indiana Department of Environmental Management
IJC	International Joint Commission
INC	International Negotiating Committee
kg	Kilogram
LaMP	Great Lakes Lakewide Management Plans
LAER	Lowest achievable emission rate
LCBP	Lake Champlain Basin Program
LISS	Long Island Sound Study
LMMBS	Lake Michigan Mass Balance Study
LRTAP	Europe Long-Range Transboundary Air Pollution
MACT	Maximum achievable control technology
MCL	Maximum contaminant level
MCM	Mercury cycling model
MDEQ	Michigan Department of Environmental Quality
MDN	Mercury Deposition Network
MOU	Memorandum of Understanding
MPCA	Minnesota Pollution Control Agency
MTRL	Maximum tissue residue level
MWC	Municipal waste combustors
N	Nitrogen
NAAEC	North American Agreement on Environmental Cooperation
NAAQS	National ambient air quality standard
NADP	National Atmospheric Deposition Program
NADP-MDN	National Atmospheric Deposition Program - Mercury Deposition Network
NADP-NTN	National Atmospheric Deposition Program - National Trends Network
NAFTA	North American Free Trade Agreement
NCSU	North Carolina State University
NDAMN	National Dioxin Air Monitoring Network
NEP	National Estuary Program
NERRS	National Estuarine Research Reserve System
NESCAUM	Northeast States and Eastern Canadian Provinces Mercury Study
NH_3	Ammonia
NH_4^+	Ammonium
NH_x	Reduced nitrogen compounds (e.g., ammonia, ammonium)
NLFWA	National Listing of Fish and Wildlife Advisories

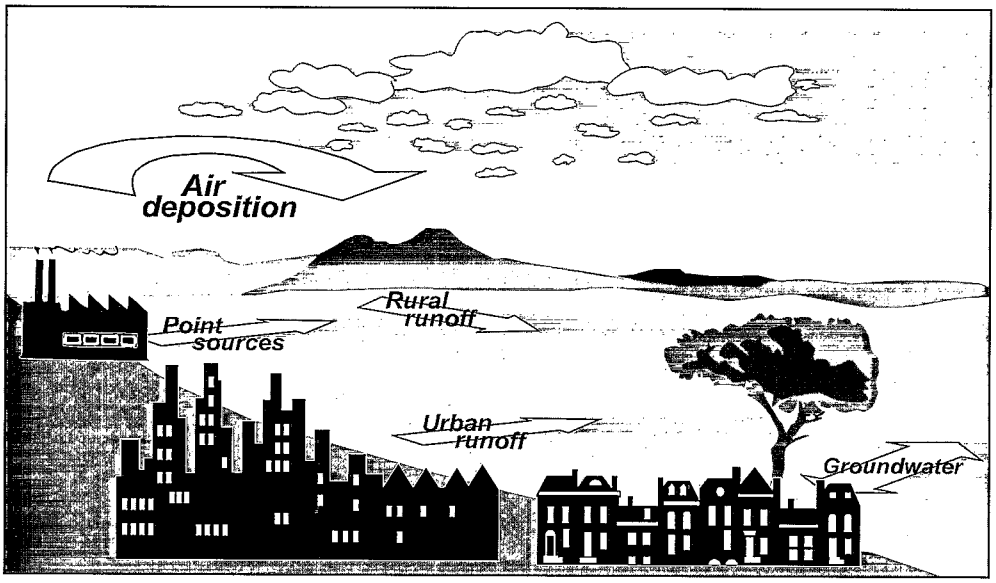
NLEV	National low emission vehicle
NO	Nitric oxide
NO_2	Nitric dioxide
NO_3	Aerosol nitrate
NOAA	National Oceanic and Atmospheric Administration
NOEL	No observed effects level
NO_x	Oxides of nitrogen
NS&T	National Status and Trends
NSI	National Sediment Inventory
NSPS	New source performance standards
NSQS	National Sediment Quality Survey
NTI	National Toxics Inventory
NORBIC	North Business-Industrial Council
OAP	Office of Atmospheric Programs
OAQPS	Office of Air Quality Planning and Standards
OAR	Office of Radiation
OC	Organochlorine chemical
OCHP	Office of Children's Health Protection
OMS	Office of Mobile Sources
OPPTS	Office of Pollution Prevention and Toxics
ORD	Office of Research and Development
OTAG	Ozone Transport Assessment Group
OTAQ	Office of Transportation and Air Quality
OTC	Ozone Transport Commission
OTR	Ozone Transport Region
OW	Office of Water
PAH	Polycyclic aromatic hydrocarbon
PBT	Persistent bioaccumulative toxic
PCB	Polychlorinated biphenyl
PCDD	Polychlorinated dibenzo-p-dioxin
PCDF	Polychlorinated dibenzofuran
PEL	Probable effects level
PM	Particulate matter
POM	Polycyclic organic matter
POP	Persistent organic pollutant
RACT	Reasonably available control technology
RADM	Regional Atmospheric Deposition Model
RAP	Regional action plan
RELMAP	Regional Lagrangian Model of Air Pollution
R-EMAP	Regional Environmental Monitoring and Assessment Program
REMSAD	Regulatory Modeling System for Aerosols and Deposition
SAB	Science Advisory Board
SAV	Submerged aquatic vegetation
SFEI	San Francisco Estuary Institute
SFEP	San Francisco Estuary Project
SIP	State implementation plan
SMOC	Sound management of chemicals
SoFAMMS	South Florida Atmospheric Mercury Monitoring Study
SVOC	Semi-volatile organic compound
SWFWMD	Southwest Florida Water Management District

SWMP	System-Wide Monitoring Program
TBNEP	Tampa Bay National Estuary Program
TEQ	Toxic equivalent quantity
TLRI	Toxics Loading and Release Inventory
TMDL	Total maximum daily load
tpy	Tons per year
TRANSCO	Transfer coefficient
TRI	Toxic Release Inventory
TRIM	Total Risk Integrated Methodology
TSCA	Toxic Substances Control Act
UNC-CH	University of North Carolina at Chapel Hill
UN-ECE	United Nations Economic Commission for Europe
UNEP	United Nations Environment Program
US	United States
USGS	U.S. Geological Survey
VOC	Volatile organic compound
WLSSD	Western Lake Superior Sanitary District
WMNP	Waste Minimization National Plan
WMPT	Waste Minimization Prioritization Tool
WQB	Water Quality Board
WRDA	Water Resources Development Act
yr	Year

CHAPTER I
INTRODUCTION AND BACKGROUND

Although water quality has improved in past years, ongoing research indicates that many of the Nation's waterbodies, including the Great Waters, are impacted by degraded water quality and associated adverse health and ecological effects. These impacts are due, in part, to inputs of pollutants from a variety of sources and through multiple pathways, including atmospheric deposition (Figure I-1). The role of air pollution as an important contributor to water pollution has long been recognized and, in recent years, has been the subject of growing scientific study and concern to regulatory agencies. Section 112 of the CAA provides the statutory basis for hazardous air pollutant (HAP) programs directed by EPA. 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*, in the 1990 Amendments to the CAA to establish research, reporting, and potential regulatory requirements related to atmospheric deposition of HAPs to the "Great Waters."

Figure I-1
Pollutant Transport Pathways to Waterbodies



This report fulfills the requirements in section 112(m)(5), which direct EPA, in cooperation with NOAA, to periodically submit a Report to Congress including information on pollutant sources and emissions, loadings, effects, and exceedances of guidelines in the Great Waters. The biennial report is required under section 112(m) of the CAA to cover the following:

1. The contribution of atmospheric deposition to pollution loadings in the Great Waters;

2. The environmental and public health effects of any pollution attributable to atmospheric deposition to these waterbodies;

3. The sources of any pollution attributable to atmospheric deposition to these waterbodies;

4. Whether pollution loadings in these waterbodies cause or contribute to exceedances of drinking water or water quality standards or, with respect to the Great Lakes, exceedances of the specific objectives of the Great Lakes Water Quality Agreement; and,

5. Descriptions of any revisions of the requirements, standards, and limitations of relevant CAA and Federal laws to ensure protection of human health and the environment.

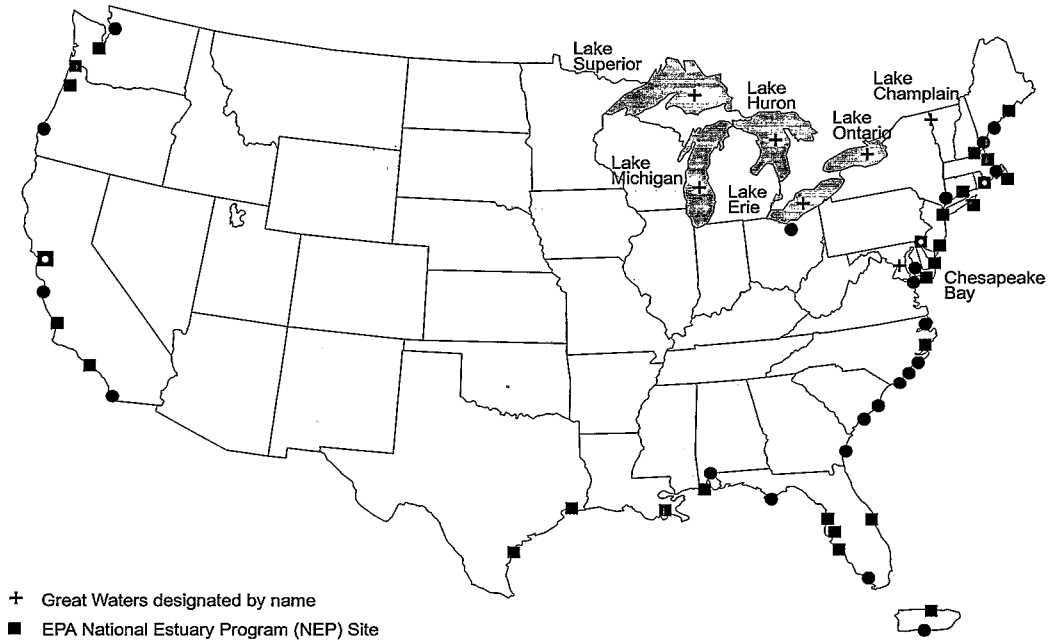
The *First* and *Second Great Waters Reports to Congress* on atmospheric deposition to the Great Waters were published in May 1994 (U.S. EPA 1994) and June 1997 (U.S. EPA 1997b). The first two reports presented the programmatic background and covered the scientific issues that are addressed by the Great Waters program. The *Third Great Waters Report to Congress* provides an update to the information presented in previous reports and specifically highlights progress made since the *Second Report to Congress*, including changes in pollutant emissions, deposition, and effects, as well as recent advancements in the scientific understanding of relevant issues. In addition, the report discusses recent activities and accomplishments of the many different initiatives that help protect the Great Waters from pollutants deposited from the atmosphere.

I.A OVERVIEW OF THE GREAT WATERS PROGRAM

DEFINITION OF GREAT WATERS

The waterbodies collectively referred to as the "Great Waters" in this report are the Great Lakes, Lake Champlain, Chesapeake Bay, and specific coastal waters defined in the statute as coastal waters designated through the National Estuary Program (NEP) and the National Estuarine Research Reserve System (NERRS). Figure I-2 displays the locations of these waterbodies. Examples of some of the coastal waters designated as Great Waters include Tampa Bay, Santa Monica Bay, San Francisco Bay, Puget Sound, Galveston Bay, Casco Bay, the New York Bight, and Albemarle-Pamlico Sounds.

Figure I-2
Locations of the Great Waters



PROGRAM PARTICIPANTS AND ORGANIZATIONS

The EPA's Great Waters program was established to implement the provisions of section 112(m) of the CAA. In partnership with NOAA, this program is charged with examining the sources and impacts of air pollutant deposition to the Great Waters and recommending potential solutions in Reports to Congress. In addition to these two agencies, other international, national, regional, and local organizations are engaged in activities that seek to reduce sources and quantities of pollution to the Great Waters and also contribute to the body of science relevant to the Great Waters program. This Report to Congress, like the first two reports, describes the most recent activities of these programs, both within and outside of EPA.

Increasingly, EPA is utilizing cross-program teams and partnerships with other agencies and organizations to study and solve complex, "multimedia" environmental issues (i.e., issues that cut across air, water, land, and biological resources). For example, the Clean Water Action Plan, issued by EPA in 1998, which includes many activities relevant to the Great Waters program, is a coordinated Federal effort involving EPA and the Departments of Agriculture, Defense, Interior, and Commerce, all of which have authorities affecting the quality of the Nation's water resources. This Report to Congress describes several such collaborative efforts that support targeted data gathering and decision making for multiple objectives, including those of the Great Waters program.

Because deposition of atmospheric pollutants to the Great Waters is a multimedia problem, several media-based and geographically-based programs are part of EPA's integrated efforts to address pollution in the Great Waters, including the following.

- **Office of Air and Radiation (OAR)** administers the Great Waters program along with other programs under the CAA that affect the Great Waters. For example, air regulations and programs developed by OAR reduce emissions of air toxics and nitrogen from many mobile and stationary sources of Great Waters pollutants of concern. Offices within OAR responsible for air pollution control programs include (1) the **Office of Air Quality Planning and Standards (OAQPS)**, which has overall responsibility for the Great Waters, air toxics, and air quality management programs; (2) the **Office of Atmospheric Programs (OAP)**, which manages the acid rain and global climate change programs; and, (3) the **Office of Transportation and Air Quality (OTAQ;** formerly the Office of Mobile Sources or OMS**)**, which is responsible for regulating on- and off-road vehicles and fuels.

- **Office of Research and Development (ORD)** undertakes and coordinates research and monitoring programs that provide information for assessment of human health and environmental risks associated with exposures to pollutants of concern in the Great Waters as well as for risk management decisions. The ORD is the lead EPA office in developing a mercury research strategy and is working closely with EPA's Office of Prevention, Pesticides and Toxic Substances (OPPTS) in preparing a dioxin reassessment.

- **Office of Water (OW)** leads numerous activities that are integral to the Great Waters program. For example, OW provides data on trends in fish and sediment contamination in the Great Waters and is working to factor atmospheric deposition into waterbody-specific water pollution standards. In addition, OW manages the Non-Point Source Control Program under section 319 of the Clean Water Act. Within OW, the **Office of Wetlands, Oceans and Watersheds** is responsible for, among other things, coastal protection (e.g., the National Estuary Program) and the TMDL determination. The **Office of Science and Technology** provides critical tools to help understand and reduce water pollution, and, in cooperation with NOAA and other Federal and State and tribal agencies, reviews and monitors fish and shellfish contamination across the U.S.

- **National Estuary Program (NEP)**, which is managed by OW, was established by EPA in 1987 to implement section 320 of the Clean Water Act. The purpose of the NEP is to restore and enhance nationally significant estuaries that are threatened or impaired by pollution, development, or overuse. Management decisions are made by conferences composed of EPA and local stakeholder groups. The NEP currently includes 28 estuaries, several of which have identified air deposition as a concern and have received Great Waters program funding to address the problem.

- **Chesapeake Bay Program Office (CBPO)** represents EPA as a partner in the Chesapeake Bay Program. This program was created in 1983 by an agreement between EPA, the State of Maryland, the Commonwealths of Virginia and Pennsylvania, and the District of Columbia. The Chesapeake Bay Program uses a cooperative, watershed approach to research and resolve public health and environmental issues affecting the bay.

- **Great Lakes National Program Office (GLNPO)** is responsible for EPA activities specific to the Great Lakes, including responsibilities defined by the Great Lakes Water Quality Agreement with Canada, Great Lakes Program requirements under section 118 of the Clean Water Act, and the Great Lakes Critical Programs Act of 1990. The GLNPO also supports EPA's OAQPS and Region V activities specific to the Great Lakes for the air toxics program under section 112 of the CAA. In addition, GLNPO conducts and funds monitoring research in support of the Great Waters program.

- **Gulf of Mexico Program Office (GMPO)** is a nonregulatory organization that sets environmental goals and implements projects to protect the environmental resources of the Gulf of Mexico. Partners in the GMPO include EPA, other governmental agencies (Federal, State, and local), private and non-profit sector stakeholders, and interested citizens. The Gulf of Mexico shoreline includes a substantial percentage of the Nation's estuaries and other significant coastal ecosystems.

- **EPA Regional Offices (Regions I-X)** are responsible for working directly with States, tribes, and regional organizations to implement national environmental programs, including the Great Waters program. For instance, Region I and Region II work with the States in the Northeast to address pollution in the Lake Champlain Basin, as well as the National Estuary Programs from Maine to New Jersey.

In addition to the EPA Offices listed above, many others contribute to the integrated efforts as well. For example, the Office of Solid Waste and Emergency Response (OSWER) provides Agencywide policy, guidance, and direction for the EPA's solid waste program and emergency response program. The Office of Enforcement and Compliance Assurance is responsible for leading the Agency in ensuring compliance with the rules that limit pollution. The OPPTS leads the Agency in pollution prevention activities, among many other activities. Chapter III discusses many of the programs led by these and other offices.

The NOAA administers the 23 National Estuarine Research Reserves System (NERRS) sites – all are coastal Great Waters – which were created in 1972 by the Coastal Zone Management Act. The NERRS sites, which were selected to be representative of various regions and estuary types, are managed cooperatively with the States to provide opportunities for long-term research, education, and stewardship. Research conducted at NERRS sites includes studies of the impacts of toxic pollutants deposited from the atmosphere.

The NOAA also conducts or funds several other monitoring and research programs that contribute to the Great Waters program. For example, the Atmospheric Integrated Research Monitoring Program (AIRMoN) conducts daily deposition monitoring, the National Status and Trends Program gathers and maintains data on the environmental quality of U.S. estuaries and coastal waters, and the NERRS System-wide Monitoring Program currently gathers water quality and weather data. In addition, NOAA plays a critical role in working with EPA to jointly assess the transport and deposition of nitrogen and other pollutants on surface waters.

Other involved participants include State, tribal, and local agencies, non-profit organizations, private sector groups, industry and industry groups, universities, other countries, and international research organizations. Relevant research and other activities of these partners are described throughout the report. The combined activities of all of these partners are responsible for progress to date in identifying, assessing, and resolving Great Waters problems, including problems associated with pollutants deposited from air pathways.

GOVERNMENT GOALS SUPPORTED BY THE GREAT WATERS PROGRAM AND ITS PARTNERS

The EPA's Strategic Plan (U.S. EPA 1997c) defines ten strategic goals used by the Agency for planning and resource allocation, as required by the Government Performance and Results Act (GPRA). The efforts by EPA, NOAA, and partners support the following GPRA goals:

1. **Clean Air** -- "The air in every American community will be safe and healthy to breathe . . . Reducing air pollution will also protect the environment, resulting in many benefits, such as restoring life in damaged ecosystems and reducing health risks to those whose subsistence depends directly on those ecosystems."

2. **Clean and Safe Water** -- "All Americans will have drinking water that is clean and safe to drink. Effective protection of America's rivers, lakes, wetlands, aquifers, and coastal and ocean waters will sustain fish, plants, and wildlife, as well as recreational, subsistence, and economic activities . . . "

3. **Preventing Pollution and Reducing Risk in Communities, Homes, Workplaces and Ecosystems** -- "EPA will safeguard ecosystems and promote the health of natural communities that are integral to the quality of life in this Nation . . ."

4. **Reduction of Global and Cross-Border Environmental Risks** -- "The U.S. will lead other nations in successful, multilateral efforts to reduce significant risks to human health and ecosystems . . . "

5. **Sound Science, Improved Understanding of Environmental Risk, and Greater Innovation to Address Environmental Problems** -- "EPA will develop and apply the best available science for addressing current and future environmental hazards, as well as new approaches toward improving environmental protection."

These efforts support these goals either directly or by contributing information on atmospheric deposition helpful to EPA's place-based (i.e., geographically-based programs, such as GLNPO) or media-oriented programs (e.g., programs in OW that focus on water resources). Specifically, the Great Waters program and its partners help the Agency achieve atmospheric deposition objectives outlined in the clean air and clean and safe water goals and the atmospheric transport and deposition component related to international pollution transport goal. Furthermore, the aspects of the work devoted to investigation and assessment of atmospheric deposition support EPA's efforts under the sound science goal to provide the best available science on which to base decisions on how best to reduce the presence and effects of this pollution. As shown in later sections of the report, progress toward each of these goals involves and emphasizes multimedia and multiprogram collaboration.

I.B POLLUTANTS OF CONCERN

This report focuses on the 15 pollutants of concern addressed in the *First* and *Second Great Waters Reports to Congress*. The general types of emission sources and uses (and use restrictions) of these pollutants are summarized in Table I-1. In general, sources of these pollutants can include U.S. short-range and long-range transport from stack emissions, natural emissions, re-emitted emissions from natural and human-made sources, long-range transport from foreign countries, and cycling within the global pool of each pollutant.

As in the two previous reports, these pollutants are organized into the following pollutant groups (with one change):

- **Mercury and Mercury Compounds**

- **Other Metals**

  -- Cadmium and cadmium compounds
  -- Lead and lead compounds

- **Combustion Emissions**

  -- Polycyclic organic matter (POM), including polycyclic aromatic hydrocarbons (PAHs)
  -- Dioxins (tetrachlorodibenzo-p-dioxin, 2,3,7,8-TCDD) and furans (tetrachlorodibenzofuran, 2,3,7,8-TCDF)
  -- Hexachlorobenzene (HCB)[1]

- **Nitrogen Compounds** (including NO_x, ammonia and ammonium, and organic nitrogen)

- **Banned and Restricted Use Substances**

  -- Polychlorinated biphenyls (PCBs)
  -- Pesticides
    - Chlordane
    - DDT/DDE
    - Dieldrin
    - Hexachlorobenzene (HCB)[1]
    - Hexachlorocyclohexane (α-HCH)[2]
    - Lindane (γ-hexachlorocyclohexane, or γ-HCH)[2]
    - Toxaphene

This last revised grouping is based on the fact that the use and manufacture of these pollutants have either been banned or severely restricted under other Federal statutes, such as the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA) and the Toxic Substances Control Act (TSCA).

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[1] HCB is a product of incomplete combustion as well as a pesticide. In Chapter II of this report, HCB is discussed under the pesticides pollutant group.

[2] There are different isomers of HCH of concern that can transform into other HCH isomers that have different persistence and toxicity characteristics.

With these pollutants, in general, historical contamination (e.g., in sediments and landfills), and not current national releases from U.S. industrial or agricultural activities, appears to be the problem. For the most part, the sources of these pollutants to the Great Waters today include resuspension of contaminated sediments, releases from contaminated soils, atmospheric deposition from long-range transport of pollutants from foreign countries where uses are still permitted, and accidental releases from stockpiles or intentional dumping. The revised grouping helps to organize research into these pollutants of concern and management of the reduction of the risks they pose to the Great Waters (e.g., work under international frameworks to reduce global releases, activities to remediate contaminated sediments).

Many programs and activities address these pollutants of concern either directly or indirectly and through statutory requirements or voluntary initiatives. The above pollutants are of concern to multiple programs because they share the problematic characteristics of toxicity, persistence, and/or potential for long-range transport in the environment (e.g., via the atmosphere). Because of these properties, many of these contaminants are ubiquitous in the environment and may accumulate to potentially harmful concentrations in water, sediments, soil, or the food web. This presents difficult challenges and places a premium on multimedia, interdisciplinary approaches for solving problems associated with these pollutants.

Table I-2 provides examples of how the Great Waters pollutants of concern are being addressed in the U.S. by closely related multimedia programs and other activities authorized by the CAA. All of the programs and activities identified in this table, along with several others, are discussed later in the report. Collectively, these programs and activities demonstrate EPA's holistic perspective and steps toward approaching the goals of clean air, clean water, and healthy ecosystems in an integrated manner.

Building strong partnerships with other governmental agencies and stakeholders is a guiding principle of EPA's Strategic Plan and one of five key attributes of EPA's reinvention plan (U.S. EPA 1997c). This report describes several ways in which EPA's progress toward the Great Waters program goals can be attributed to the collective efforts of many partners at the local, tribal, State, Federal, and international levels. Table I-3 identifies some of the programs at different levels that address Great Waters pollutants of concern. Each of these programs and activities is described later in this report.

Table I-1
Uses/Sources and U.S. Regulatory Status of Great Waters Pollutants of Concern

Pollutant	Examples of Uses/Sources
Mercury and Compounds	Naturally occurring element often used in thermometers, electrical equipment (such as batteries and switching equipment), industrial control instruments, and industrial processes (e.g., chlor-alkali plants). Released during combustion of fossil fuels (e.g., coal, oil); incineration of municipal, medical, and hazardous waste; and from numerous manufacturing and natural processes (e.g., volcanic activity). Banned as a paint additive in U.S. in both interior (1990) and exterior (1991) paint. Being phased out of batteries. Removed from catalysts, turf products, and explosives.
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.
Lead and Compounds	Naturally occurring element historically used in gasoline and paint additives, and still used in 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.
POM (includes PAHs)	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.
Dioxins and Furans	By-product of combustion of organic material containing chlorine, chlorine bleaching in pulp and paper manufacturing, and diesel-fueled vehicles. Also a contaminant in some pesticides.
Nitrogen Compounds	Naturally occurring substances from a variety of processes (e.g., microbial activities, lightning, biomass combustion) and by-products of power generation, industrial, and motor vehicle fossil fuel combustion processes (NO_x). Also, compounds used in fertilizers and released from agricultural animal manures (NH_3).
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 U.S.
Chlordane	Insecticide used widely in the 1970s and 1980s. All U.S. uses except termite control canceled in 1978; use for termite control voluntarily suspended in 1988. Use of existing stocks permitted.
DDT/DDE	Insecticide used widely from introduction in 1946 until significantly restricted in U.S. in 1972. Still used in other countries. Used in U.S. for agriculture and public health purposes only with special permits.
Dieldrin	Insecticide used widely after introduction in late 1940s. Used in U.S. for termite control from 1972 until registration voluntarily suspended in 1987.
Hexachlorobenzene	Fungicide used as seed protectant until 1985. 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.
Hexachlorocyclohexane	Component of technical-HCH, an insecticide for which use is restricted in U.S., but which is used widely in other countries.
Lindane	An insecticide used on food crops and forests, and to control lice and scabies in livestock and humans. Currently used primarily in China, India, and Mexico. U.S. production stopped in 1977. Use was restricted in 1983; many uses are still registered, but are expected to be voluntarily discontinued in the future.
Toxaphene	Insecticide used widely on cotton in the southern U.S. until the late 1970s. Most U.S. uses banned in 1982; remaining uses canceled in 1987.

Table I-2
EPA Multimedia and Cross-program Management of the Great Waters Pollutants of Concern

Great Waters Pollutant of Concern	Selected Multimedia and Air Programs				
	Multimedia Activities			Air Program Activities	
	PBT Initiative[a]	Clean Water Action Plan[b]	Contaminated Sediment Management Strategy[c]	CAA Section 112 Activities[d]	Other Major CAA Programs[e]
Mercury and Compounds	✔	✔	✔	• Standards for Specific Pollutants • MACT Program • Urban Area Source Program • Utility Study • Mercury Study • NIEHS Fish Mercury Study	• Solid Waste Combustion
Cadmium and Compounds		✔	✔	• MACT Program • Urban Area Source Program • Utility Study	• Solid Waste Combustion
Lead and Compounds	✔	✔	✔	• Standards for Specific Pollutants • MACT Program • Urban Area Source Program • Utility Study • Prevention of Accidental Releases	• Criteria Pollutant Program • Solid Waste Combustion • Fuels Program
POM (includes PAHs)	✔	✔	✔	• Standards for Specific Pollutants • MACT Program • Urban Area Source Program • Utility Study	
Dioxins and Furans	✔	✔	✔	• Standards for Specific Pollutants • MACT Program • Urban Area Source Program • Utility Study	• Solid Waste Combustion
Nitrogen Compounds		✔	✔	• Prevention of Accidental Releases	• Criteria Pollutant Program (NO_x, O_3, particulate matter) • Mobile Source Program • Acid Rain Program (NO_x) • Solid Waste Combustion
PCBs	✔	✔	✔	• MACT Program •Urban Area Source Program	
Chlordane	✔	✔	✔	• MACT Program	
DDT/DDE	✔	✔	✔		
Dieldrin	✔	✔	✔		
Hexachlorobenzene	✔	✔	✔	• Standards for Specific Pollutants • MACT Program •Urban Area Source Program • Utility Study	

Table I-2 (continued)
EPA Multimedia and Cross-program Management of the Great Waters Pollutants of Concern

Great Waters Pollutant of Concern	Selected Multimedia and Air Programs				
	Multimedia Activities			Air Program Activities	
	PBT Initiative[a]	Clean Water Action Plan[b]	Contaminated Sediment Management Strategy[c]	CAA Section 112 Activities[d]	Other Major CAA Programs[e]
Hexachlorocyclohexane		✔	✔		
Lindane		✔	✔	• MACT Program	
Toxaphene	✔	✔	✔	• MACT Program	

a As discussed in Chapter III of this report, the Persistent Bioaccumulative Toxic (PBT) Chemicals Initiative is a cross-office EPA effort to coordinate actions associated with these high-priority pollutants.

b Because the Clean Water Action Plan does not address a specific set of pollutants, any of the Great Waters pollutants of concern may be addressed (see Chapter III of this report for more detail).

c Because the Contaminated Sediment Management Strategy does not address a specific set of pollutants, any of the Great Waters pollutants of concern may be addressed (see Chapter III of this report for more detail).

d
- Standard for specific pollutants = §112(c)(6); Sources accounting for 90 percent of the emissions of these seven pollutants (i.e., alkylated lead compounds, POM, HCB, mercury, PCBs, dioxins and furans) are subject to regulation.
- MACT Program = §112(d)
- Urban Area Source Program = §112(k)
- Utility Study = §112(n)(1)(A); Emissions of certain pollutants from electric utility steam generating units are evaluated for potential regulation.
- Mercury Study = §112(n)(1)(B); Emissions of mercury from electric utilities, municipal waste combustors, and other sources studied for potential regulation.
- NIEHS Fish Mercury Study = §112(n)(1)(C); Report by the National Institute of Environmental Health Sciences on the "threshold" level for human health effects from mercury.
- Prevention of Accidental Releases = §112(r)

e
- Criteria Pollutant Program = §108
- Mobile Source Program = Subchapter II
- Solid Waste Combustion = §129
- Acid Rain Program = Subchapter IV-A

Table I-3
Great Waters Pollutants of Concern Addressed by Selected Waterbody, Regional, and International Activities[a]

Pollutant	Regional and Waterbody-specific Programs				International Activities				
	Great Lakes Programs[b]	Lake Champlain Basin Program[c]	Chesapeake Bay Program[d]	Estuarine Initiatives[e]	Binational Strategy	International Joint Commission	LRTAP	NAFTA	UNEP
Mercury and Compounds	✔	✔	✔	✔	✔	✔	✔	✔	
Cadmium and Compounds	✔	✔	✔	✔			✔		
Lead and Compounds	✔	✔	✔	✔	✔	✔	✔		
POM (includes PAHs)	✔[f]	✔	✔	✔	✔	✔[f]	✔[g]		✔[g]
Dioxins and Furans	✔	✔		✔	✔	✔	✔[g]	✔	✔[g]
Nitrogen Compounds		✔	✔	✔			✔		
PCBs	✔	✔	✔	✔	✔	✔	✔[g]		✔[g]
Chlordane	✔	✔	✔	✔	✔		✔[g]	✔	✔[g]
DDT/DDE	✔	✔		✔	✔	✔	✔[g]		✔[g]
Dieldrin	✔	✔	✔	✔	✔	✔	✔[g]		✔[g]
Hexachlorobenzene	✔	✔		✔	✔	✔	✔[g]		✔[g]
Hexachlorocyclohexane	✔	✔		✔			✔[g]		✔[g]
Lindane	✔	✔		✔			✔[g]	✔	✔[g]
Toxaphene	✔	✔	✔	✔	✔	✔	✔[g]		✔[g]

a See Chapter III of this report for information on the status of all of these and other programs addressing the Great Waters pollutants of concern.
b Integrated Atmospheric Deposition Network (IADN); Great Lakes Emissions Inventories; Lake Michigan Mass Balance Study; Great Lakes Lakewide Management Plans (LaMPs) and regional action plans (RAPs)
c Includes Group 1 through Group 4 Toxic Substances of Concern for the Lake Champlain Basin Program (LCBP). Ammonia, a Group 3 Toxic Substance of Concern, is included in the table as a nitrogen compound.
d Includes Toxics of Concern and Chemicals of Potential Concern for the Chesapeake Bay Program.
e Any of the Great Waters pollutants of concern could be a concern at a specific NEP or NERRS site.
f Benzo[a]pyrene
g Persistent organic pollutants (POPs) are included as pollutants of concern under the United Nations Economic Commission for Europe Long-Range Transboundary Air Pollution (LRTAP) initiative, as well as the United Nations Environment Program (UNEP) POPs initiative. This constituent is a POP.

I.C GOALS OF THE THIRD REPORT TO CONGRESS

The goals of the *Third Report to Congress* are to discuss the current state of knowledge regarding atmospheric deposition of pollutants to the Great Waters based on the information available and program activities since June 1996 (the end of the timeframe for information presented in the *Second Report to Congress*) and to describe any necessary revisions to requirements, standards, and limitations pursuant to the CAA and other Federal laws. The report is organized into four chapters that summarize the following:

- Observed and anticipated **spatial and temporal changes** in airborne emissions, loadings, and effects of pollutants of concern (Chapter II);
- **Programs and activities** that have been undertaken to address pollutants of concern since the *Second Report to Congress* (Chapter III);
- **Advancements in scientific research methods, environmental models, and data sources** that improve our understanding of and abilities to address the public health and environmental risks posed by the pollutants of concern in the Great Waters (Chapter IV); and,
- **Conclusions and recommendations** to help further national, regional, and waterbody-specific activities related to the Great Waters program (Chapter V).

In addition, this report identifies important remaining uncertainties, such as those that limit EPA's ability to assess the links between pollutant sources, loadings, and effects. The research findings, conclusions, and recommendations presented in this report will be used to further understand and promote reductions of overall contaminant loadings to the Great Waters.

The report focuses on the scientific and programmatic progress that has occurred since the previous report and presents information according to the major issues of concern (e.g., trends in loadings and effects, program progress, scientific tools) from a national perspective. Regional or waterbody-specific examples are presented in the context of a national-level picture of progress toward the goals of the Great Waters program. For more detail on specific regions or waterbodies, readers are referred to the many other publications or Internet sites that specifically address those areas (see Appendix A). In addition, the report includes an index of waterbodies to enable readers to identify all information related to a specific waterbody that is presented in this report.

The information in this report was collected from several sources. The references cited are generally from published peer-reviewed journals, government reports, and conference proceedings. The report uses a variety of sources that provide relevant new information on Great Waters issues and generally addresses information released in the past 2 years. In some cases, large summary reports were used and, therefore, primary sources of information are not cited. In general, literature published from the summer of 1996 to the fall of 1998 is included.

In addition to literature searches for updated information, EPA obtained current scientific and programmatic information about atmospheric deposition to the Great Waters through personal interviews with and direct input from various researchers and program participants. Many of the waterbody-specific activities highlighted in this report, for example, were provided by the EPA offices that coordinate investigation, restoration, and maintenance efforts of the waterbody (e.g., Great Lakes National Program Office, EPA Region V Air and Radiation Division, Chesapeake Bay Program Office, Gulf of Mexico Program Office).

CHAPTER II
ENVIRONMENTAL PROGRESS

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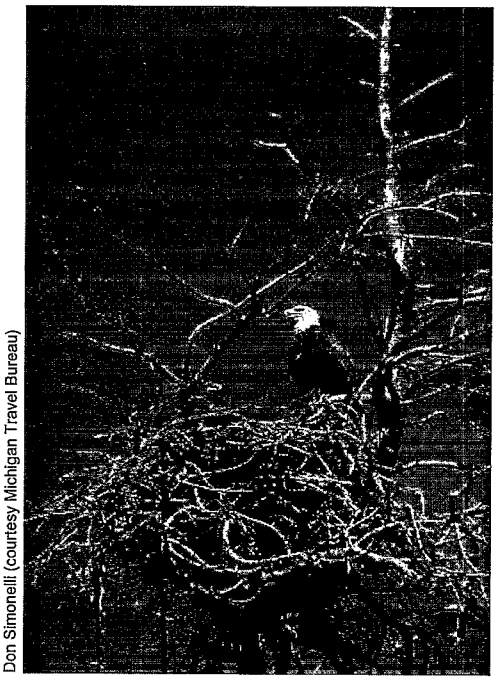


Don Simonelli (courtesy Michigan Travel Bureau)

This chapter attempts to answer the question: *Is the problem associated with atmospheric deposition of the pollutants of concern in the Great Waters getting better or worse?* It is not intended to summarize the myriad of regulatory and nonregulatory activities and accomplishments designed to solve the problem, a discussion which is reserved for Chapter III. Likewise, this chapter is not intended to cover new scientific research or analytical tools that help improve the understanding of pieces of the problem, such as the details of pollutant behavior in the atmosphere or the ways pollutants are exchanged between air and water. This more science-oriented update is provided in Chapter IV, although some of the results of this research are included in this chapter.

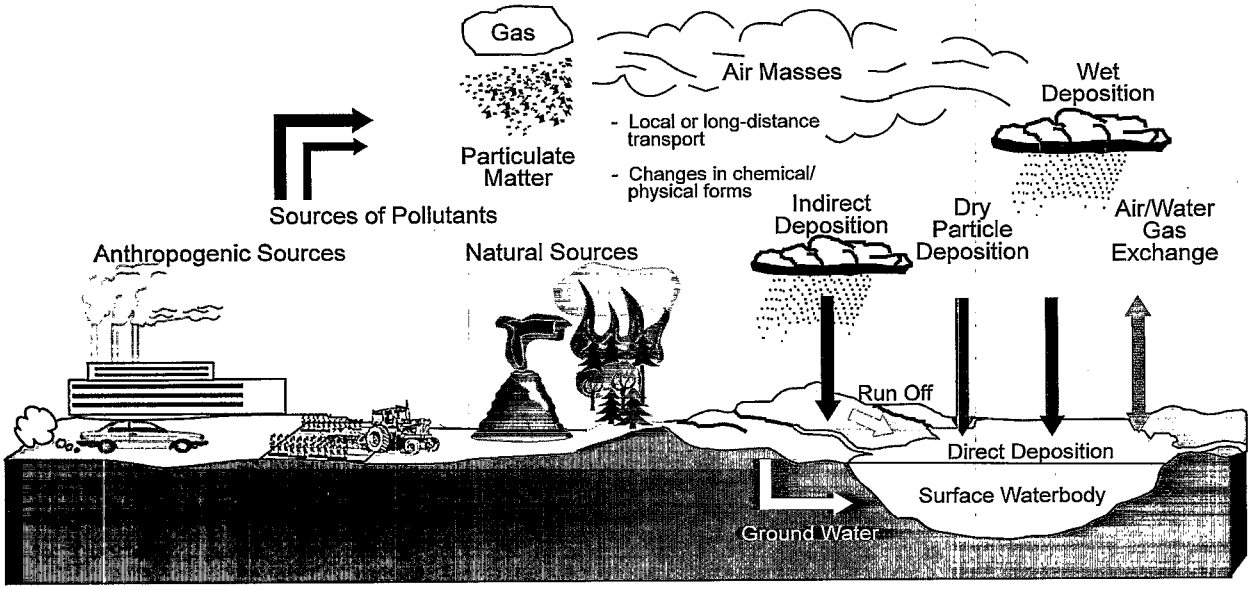
This chapter summarizes recent information on the changes in sources and emissions (including transport and transformation), loadings, and exposure and effects of pollutants of concern in the Great Waters, and is organized into two sections. The first section, organized by pollutant groups (see Chapter I), provides information for each pollutant of concern. The second section discusses trends from assessments that address multiple pollutants. These discussions reflect results of new studies published since the *Second Report to Congress*, some of which address time periods prior to the *Second Report to Congress*. They focus on the observed changes over time, including temporal and spatial changes in sources, emissions, loadings, exposure, and effects reported in new studies combined (where appropriate) with information presented in the *First* and *Second Reports to Congress*.

As background for discussing these questions, it is helpful to review the components of the atmospheric release, transport, and deposition processes (Figure II-1), as well as environmental and public health effects.

Pollutant emissions to the atmosphere can be characterized as "human-made" or "anthropogenic" (i.e., released by human activities), "natural" (i.e., releases of geologically-bound pollutants by natural processes), or "re-emitted" (i.e., the mass transfer to the atmosphere by

biologic/geologic processes for previously deposited pollutants). Examples of human-made emissions are those associated with industrial stacks, municipal waste incinerators, agricultural activities (e.g., pesticide applications, manure management), and vehicle exhaust. Examples of natural emissions include those associated with volcanic eruptions, windblown gases and particles from forest fires, windblown dust and soil particles, and sea spray. In many cases, however, it is difficult to determine if the pollutant inputs are from natural or anthropogenic sources.

Figure II-1
Atmospheric Release, Transport, and Deposition Processes



Airborne releases or emissions of pollutants can be transported away from the source to other locations. Depending on the weather conditions and the chemical and physical properties of the pollutant, air pollutants can be transported varying distances and may undergo physical, chemical, and/or biological transformations during transport (U.S. EPA 1997b). As mentioned in Chapter I, all of the Great Waters pollutants of concern are known to be subject to long-range transport.

Pollutant loadings to waterbodies can occur through many different pathways, including connecting streams and rivers, groundwater inflow, land surface runoff, rerelease of pollutants from sediments, and atmospheric deposition processes. Atmospheric inputs to waterbodies also occur through several mechanisms (Figure II-1). Pollutants that are released to the air can be deposited to land areas, tributaries, or directly to the waterbody by wet or dry deposition. The process of wet deposition refers to the removal of air pollutants from the air by a precipitation event, such as rain or snow. The process of dry deposition refers to the removal of aerosol pollutants through eddy diffusion and impaction, large particles through gravitational settling, and gaseous pollutants through direct transfer from the air to the water (i.e., gas exchange). Air pollutants can also enter the waterbody through indirect deposition which occurs when an air pollutant is deposited to a land area or tributary and is then carried into a waterbody by other routes, such as storm water runoff or inflow from tributaries. Therefore, in these mechanisms, loadings to a waterbody are both directly and indirectly affected by air pollutant emission levels.

Loadings can be expressed as the total amount of a pollutant entering a waterbody over a specified time period (e.g., kg/year) or as a loading rate (e.g., $\mu g/m^2$/year). The tendency of a specific pollutant to enter a waterbody through wet deposition, dry deposition, or gas exchange is related to the

physical and chemical properties of the pollutant as well as to current and local meteorology. Determining the relative atmospheric loading – how the loading from atmospheric deposition compares to that from other pathways, such as groundwater seepage and inflow from connecting surface waterbodies, where portions of the pollutant load may not be of atmospheric origin – is as important as determining total loading. Uncertainties exist, however, in loadings estimates because of errors inherent in sampling methodologies and the assumptions about a specific chemical's behavior that are used to develop deposition estimates. Furthermore, monitoring networks have spatial and temporal limitations, making it difficult to accurately and reliably quantify atmospheric deposition inputs.

Air pollutant loadings to the waterbody may contribute to **environmental and public health effects**, such as when **water quality and drinking water standards** are exceeded (where such standards exist). These standards provide a measure of degradation and possible exposures of plants, animals, and humans to potentially harmful levels of pollutants. The number and magnitude of water quality and drinking water standard exceedances in a waterbody may be affected, in part, by changes in pollutant loadings from the atmosphere, which in turn, are affected by changes in atmospheric emissions of those pollutants. Potential ecological and human health effects from exposures to the Great Waters pollutants of concern are not discussed comprehensively in this report because the *Second Report to Congress* focused on effects from exposures. This report focuses more on recent literature on trends, such as contaminant levels found in biota as well as fish consumption advisories. In many cases, however, information on trends in exposure levels or observed effects was not available. Therefore, surrogate measures of exposure, such as trends in concentration levels in environmental media or biota, are presented. Note that the presence of a pollutant in the environment only translates into an exposure for humans and/or biota if an exposure pathway exists.

The understanding of how the pieces of this complex puzzle fit together continues to increase, although uncertainty and gaps in information remain. This chapter reports on new information that helps to establish the links between emissions, transport and transformation, loadings, and effects. It also includes information on the various elements that contribute to establishing these links. For example, it includes inventories of emission sources, which are essential inputs to models that predict pollutant loadings to waterbodies.

POLLUTANT-SPECIFIC CHANGES

For each pollutant or pollutant group, information on sources and emissions is presented first, followed by new loadings information, and then by new information on exposure and effects. If no new information is available on one of these topics for a particular pollutant or pollutant group, that topic simply is not addressed. The second part of this section, *Cross-Pollutant Changes*, summarizes results from several EPA and other agency programs and projects that are not limited to individual pollutants, but rather provide information on many pollutants in a manner making it more appropriate to present the information as a cohesive piece as opposed to dividing the information among the pollutant groups.

Although a large amount of research has been and continues to be conducted related to the release, transport, loadings, exposure, and effects of Great Waters pollutants of concern, information gaps and uncertainties remain. In general, the information presented below consists of the highlights of the studies, and details on assumptions and uncertainties are not always presented. However, most if not all of the studies discussed below are associated with some type of uncertainty (e.g., related to the number of measurements, the time period of the study, or the lack of standard measurement methods). The original study should be consulted for these details for a better understanding of the assumptions used and resultant uncertainties. While uncertainties and information gaps continue to exist, it should be noted that research is ongoing to attempt to reduce them.

MERCURY AND COMPOUNDS

Mercury is a cationic metal – it has characteristics of both a metal and an organic compound (e.g., it is persistent in the environment, bioaccumulates in the food chain, and vaporizes at low temperatures). Mercury is an excellent conductor of electricity and is widely used in products such as batteries, electric switches, thermostats, thermometers, and barometers as well as a number of industrial processes including pharmaceutical preparations and the production of caustic soda and chlorine.

Mercury can exist in three oxidation states: Hg^0 (metallic), Hg_2^{2+} (mercurous), and Hg^{2+} (mercuric). The oxidation state strongly influences the properties and chemical behavior of mercury. For example, mercurous and mercuric mercury can form numerous inorganic and organic chemical compounds; however, mercurous mercury is rarely stable under ordinary environmental conditions. Most of the mercury encountered in water, soil, sediment, and biota is in the form of inorganic mercuric salts and organomercurics (i.e., chemical compounds with a covalent Hg-C bond, rather than inorganic mercury compounds that simply associate with organic material in the environment). The mercury compounds most likely to be found under environmental conditions include the following: the mercuric salts $HgCl_2$, $Hg(OH)_2$, and HgS; the methylmercury compounds, methylmercuric chloride (CH_3HgCl) and methylmercuric hydroxide (CH_3HgOH); and, in small fractions, other organomercurics (i.e., dimethylmercury and phenylmercury) (U.S. EPA 1997e). The mercury compounds that are of highest concern in terms of exposure and toxicity to humans and biota are the methylated mercury compounds. The *Mercury Study Report to Congress* (U.S. EPA 1997e) provides additional background information on mercury and mercury compounds.

HIGHLIGHTS
Mercury

➤ **Sources.** The largest human-made sources of mercury emissions in the U.S. are coal-fired utility boilers, medical waste incinerators, and municipal waste combustors. Mercury can also be emitted from natural sources, such as volcanic activities. Mercury releases from human activities today are adding to the global reservoir of mercury cycling between land, water, and air that is the result of natural releases and past human activities. Human-made emissions of mercury have declined since 1990, due chiefly to the phase-out of mercury-containing products.

➤ **Loadings.** Atmospheric deposition is a principal source of mercury to several Great Waters, followed by riverine inputs. Urban areas in proximity to the Great Waters can also contribute significantly to loadings via the atmosphere and urban runoff. Deposited mercury that is buried or immobilized in soils and sediments can be transported to waterbodies by high streamflow events like storms and snowmelt. Forested watersheds retard mercury transport to the waterbody compared to agricultural or urban watersheds.

➤ **Human and Ecological Exposure and Effects.** Consumption of contaminated fish represents the dominant pathway for both human and wildlife exposures to methylmercury. The typical U.S. consumer of fish from restaurants and grocery stores is not believed to be at risk. Rather, consumers who eat more fish than is typical or fish that are more contaminated than typical fish may be at risk of adverse effects. Because the developing fetus is regarded as the most sensitive to the effects of methylmercury, women of childbearing age are the population of greatest concern. State and tribal fish consumption advisories have been implemented to warn people about the related risks. Measurements of mercury in loons indicate up to 30 percent of male loons in the northeastern U.S. have mercury levels sufficiently high to cause adverse effects.

Sources of mercury emissions to the air in the U.S. are ubiquitous and can be broadly classified as belonging to one of three types:

1. *Re-emitted mercury* – the re-emission of mercury to the atmosphere by biologic and geologic processes from mercury that was previously deposited to the earth's surface following either anthropogenic or natural releases. For example, a large portion of the deposited mercury is the result of past human-made releases as well as releases from natural sources that previously were sequestered (e.g., arctic tundra, ice sheets, oceans and wetlands);

2. *Natural mercury emissions* – the mobilization or release of geologically-bound mercury by natural processes; or

3. *Anthropogenic mercury emissions* – the mobilization or releases of mercury by human activities.

Atmospheric deposition of mercury occurs primarily through two processes: scavenging of particulate matter by precipitation and conversion of elemental mercury (Hg^0) to ionic mercury during rain formation. Dry deposition of reactive gaseous (Hg^{2+}) and particulate mercury is also thought to be an important process. The three dominant mercury forms act differently in the environment:

- Hg^0 can be transported in the atmosphere for thousands of miles around the globe and deposits very inefficiently because it is insoluble in water and is less chemically reactive;

- Hg^{2+} in the gas-phase may be removed from the atmosphere within a few ten to a few hundred miles of the source because it is highly water soluble and reactive; and,

- Particulate-phase mercury may be deposited at intermediate distances from the source, depending on the size of the aerosol (Schroeder and Munthe 1998).

Current anthropogenic mercury emissions are only one component of the global mercury cycle. The amount of mercury in the land, water, and air at any one location is comprised of mercury from the natural global cycle, the global cycle perturbed by human activities, as well as regional sources and local anthropogenic sources. In addition to air emissions, other sources of mercury include direct water discharges or past uses of mercury, such as fungicide application to crops. Current research continues to indicate that natural sources, industrial sources, and recycled anthropogenic mercury each contribute to about one-third of the current mercury burden in the global atmosphere (Pirrone et al. 1996). However, as discussed further in Chapter IV, new measurement methods suggest that natural mercury emissions rates from mercury-rich soils and bedrocks may be larger than past estimates. It is important to understand the source of mercury and the amount of mercury contributed by each source type so that the most efficient control strategies can be devised.

Given the complexities of the global mercury cycle, much of the research in recent years focused on the environmental fate of mercury and the extent of current contamination in the environment. The state of the science at this time does not provide a complete answer to the question of whether the problem is improving. There is a general downward trend in anthropogenic mercury emissions in the U.S., although emissions from many CAA source categories are not included in this assessment. Atmospheric deposition of mercury to an adjacent waterbody is primarily a function of the amount of particulate-phase and reactive gaseous mercury emitted, the proximity of the source to the waterbody, and meteorological conditions. The National Atmospheric Deposition Program - Mercury Deposition Network (NADP-MDN) has been used to collect weekly wet deposition data for the past 4 years in some locations; however, these early data are not robust enough to discern trends. Additional monitoring data (which includes both wet and dry deposition measurements) from high quality stations over a longer period are necessary for effective trends analyses. These measurements must be performed to allow for meteorological analysis and identification of source contributions to the measured mercury. In general, the research described in this section represents an important step forward in understanding the fate of mercury in the environment and will continue to form the scientific basis for measures taken to address the problem.

Mercury Emissions

Estimates of the global contribution of mercury emissions to the atmosphere from anthropogenic sources are 2,000 to 4,000 tons per year (tpy) and from natural sources are 2,200 to 4,000 tpy, resulting in total mercury air emissions of 4,200 to 8,000 tpy (Pirrone et al. 1998). In comparison, U.S. mercury air emissions for the 1994-1995 timeframe are 158 tpy, as reported in EPA's *Mercury Study Report to Congress* (Table II-1) (U.S. EPA 1997e). Roughly 87 percent of national mercury emissions in the U.S. are from combustion sources, including waste and fossil fuel combustion.

A recently identified source of mercury to the atmosphere is emissions of elemental mercury gas (Hg^0) from soils that have been amended with municipal sewage sludge. Carpi and Lindberg (1997), using a field chamber and soil amended with sewage sludge, found that sludge application to soil increased soil Hg^0 releases by up to two orders of magnitude. The researchers estimated that land application of sewage sludge in the U.S. and European Union may account for approximately 5×10^6 g/year (5 metric tons/year) of Hg^0 released to the atmosphere based on the area of land amended each year and measured Hg^0 emission rates.

While there is a growing body of data on the total amount of mercury emitted from different source types, data on the form (or species) of mercury emitted remain a critical research need. The *Mercury Study Report to Congress* (U.S. EPA 1997e) estimated that anywhere from 0 to 73 percent of the mercury emitted from different source categories is in the form of divalent mercury (Hg^{2+}), the form that deposits and contributes the most to methylmercury concentrations in soil, water, sediment, and biota. There is, however, considerable uncertainty about the amounts of different mercury species emitted from some industrial sources.

The emission inventories provide a snapshot of recent emissions and indicate a downward trend in U.S. emissions between 1990 and 1995. This downward trend follows a general decline in domestic mercury uses and reduced emissions from waste combustion sources (including municipal waste combustors and medical waste incinerators). Industrial demand for mercury peaked in 1964 and fell 75 percent between 1988 and 1996. This decline largely reflects the phaseout of mercury in household batteries and paint. The rate of decline, however, has slowed since 1990. Future trends in mercury emissions from fossil fuel combustion sources, which currently represent a large portion of national mercury emissions, are dependent on future energy needs and fuel use. Mercury emissions from municipal waste combustors and medical waste incinerators, which also are large national sources of mercury emissions, are expected to continue to decline from 1995 through 2000 due to new regulations (see Chapter III) (U.S. EPA 1997e).

It should be noted that several uncertainties exist with the mercury emissions inventory. For example, not all source categories are included (e.g., landfills), and recent data submitted to EPA by the Chlorine Institute indicate that current emissions may be underestimated. Furthermore, mercury emissions from coal-fired electric utilities are expected to increase in future years, based on estimated changes in emissions from implementation of the acid rain program and projected trends in fuel choices and electric power demands in the year 2010 (U.S. EPA 1998p). As noted earlier in this section, the speciation of mercury emitted is important in determining its transport and fate, yet remains a key uncertainty. The EPA has worked with the Department of Energy and the American Society for Testing and Materials to develop a standard method for measuring speciated mercury from utility boilers. The EPA is also working to refine this method for other source categories, such as incinerators. The electric utility industry is currently conducting speciated mercury emission testing on a subset of coal-fired facilities, as discussed in the hazardous air pollutant section of Chapter III. Finally, in order to better understand the relative contribution of mercury to the environment from anthropogenic sources, further

research is needed into the level of natural emissions as well as the amount and original source of mercury that is re-emitted to the atmosphere after being deposited to soils, watersheds, and ocean waters. Although considerable uncertainty still exists, it has become increasingly evident that anthropogenic emissions of mercury to the air rival or exceed natural inputs (U.S. EPA 1997e).

Table II-1
National Anthropogenic Mercury Air Emissions
(Based on 1994-1995 Inventory; U.S. EPA 1997e)

Source Category	1994-1995 Anthropogenic Air Emissions (tons/year)	Percent Contribution to Total U.S. Anthropogenic Air Emissions
Utility boilers: coal combustion, oil, and natural gas	52	33
Municipal waste combustion	30	19
Commercial/industrial boilers: coal and oil	28	18
Medical waste incineration	16	10
Chlor-alkali production	7	4
Hazardous waste combustors	7	4
Chlor-alkali	7	4
Portland cement, excluding hazardous waste-fired	5	3
Residential boilers: oil and coal	4	2
Pulp and paper manufacturing	2	1
Others (<1percent each) [a]	3	2
Total U.S. Anthropogenic Mercury Air Emissions	158[b,c]	100[b,c]

[a] A list of the source categories that contribute less than 1 percent to total U.S. emissions is provided in Appendix B.
[b] This value represents anthropogenic mercury air emissions in the U.S. only.
[c] Values do not add exactly due to rounding.

Mercury Deposition in the U.S.

Once in the air, mercury can be deposited locally or widely dispersed and transported long distances from emission sources. How far it travels and where it is deposited depends in part on the chemical and physical form of the mercury emitted. Elemental mercury may be transported in the atmosphere for relatively long periods (approximately 1 year), allowing its distribution over long distances, both regionally and globally, before being deposited to the earth. In contrast, the residence time of mercuric (Hg^{2+}) compounds in the atmosphere is generally believed to be a few days or less, resulting in deposition closer to sources. Mercury also differs from other metals because it is readily re-emitted to the atmosphere following deposition and chemical transformation. In summary, principal factors that contribute to the modeled and observed patterns of mercury deposition are (1) the emission source locations and characteristics (e.g., stack height); (2) the amount of Hg^{2+} and particulate mercury

emitted; (3) atmospheric chemical and physical properties; (4) uncertainties associated with the emissions inventories, both in the magnitudes and the chemical forms emitted; (5) climate and meteorology, and (6) treatment of natural sources and re-emission within the modeling domain.

Because of the influence of the above six factors on mercury deposition, modeling of mercury transport and deposition at different spatial scales (e.g., national, regional, and local) results in slightly different information appropriate to each scale. Modeling at the local scale (i.e., approximately 100 km), can provide more specific deposition estimates for a specific waterbody, but requires meteorological modeling at the appropriate scale and up-to-date information on the amount of divalent and particulate mercury emitted from the local sources. Recent studies in the Great Lakes region and in Florida show that mercury emissions on local scales can greatly influence loadings in some locations when local sources have significant emissions of divalent and particulate forms of mercury. For example, the South Florida Atmospheric Mercury Monitoring Study (SoFAMMS) collected daily event precipitation samples at 17 sites for 1 month and, using a suite of chemical and meteorological data, was able to demonstrate that local anthropogenic sources strongly influence mercury wet deposition levels (see Section III.C for additional information on SoFAMMS) (Dvonch et al. 1998). National modeling of emissions and meteorology, on the other hand, can assist in answering some questions, but should not be applied to regional or local analyses without critical review. The various forms of mercury in emissions are all important in predicting atmospheric transport and deposition patterns, and care must be taken in estimating local deposition or longer range impacts.

For the *Mercury Study Report to Congress*, EPA used the Regional Lagrangian Model of Air Pollution (RELMAP) to model mercury emissions from the continental U.S. to estimate the amount of mercury being deposited to the U.S. The model inputs included meteorological data from 1989, the U.S. emissions inventory of 158 tons/year (U.S. EPA 1997e), and assumptions about the chemical species of mercury emitted from various source types. The results showed total deposition (i.e., wet and dry deposition of all forms of mercury) to be over 10 Fg/m^2 throughout most of the continental U.S. east of the Mississippi River, with values over 30 Fg/m^2 for the northeast corridor and other urban areas. The highest mercury deposition rates were predicted to occur in the southern Great Lakes and Ohio River valley, the Northeast, and scattered areas in the South (with highest deposition rates in the Miami and Tampa areas). Deposition of mercury is strongly influenced by the magnitude of emissions, the chemical species of the emitted mercury (ionic and particulate-bound forms of mercury being the most readily deposited), ozone and soot concentrations, and total annual precipitation.

On a national basis, the computer simulation suggested that about one-third (~ 52 tpy) of U.S. anthropogenic mercury emissions is deposited within the lower 48 States. The remaining two-thirds (~ 107 tpy) of mercury emissions are transported beyond U.S. borders. In addition, the simulation suggests that another 35 tpy of mercury from the global reservoir (which includes mercury from U.S. emissions) are deposited in the U.S., resulting in a total deposition of roughly 87 tpy. Note, however, that the results of the simulation include some uncertainty, as discussed in more detail in the following paragraph. According to the simulation, 98 percent of the deposited mercury of anthropogenic origin was emitted in the form of ionic mercury or particulate mercury. The emissions inventory and estimated chemical/speciation profiles indicate that of all combined ionic and particulate mercury emissions, 29 percent is from coal-fired electric utility boilers, 25 percent is from municipal waste combustion, 18 percent is from medical waste incineration, 16 percent is from coal-fired commercial and industrial boilers, and 12 percent is from all other modeled sources. The methods for developing these mercury emission estimates and speciation profiles are described in detail in volumes I and II of EPA's *Mercury Study Report to Congress* (U.S. EPA 1997e).

As the *Mercury Study Report to Congress* explains, there are a number of uncertainties in the RELMAP analysis, with a large degree related to the state-of-the-science on chemical and physical forms of mercury air emissions and their chemical and physical transformations in the atmosphere. The model seems most sensitive to the speciation of mercury emissions. Speciated emissions data remain a critical research need, as discussed above in the section on mercury emissions. In addition, atmospheric chemistry data are incomplete in the model. Furthermore, there is inadequate information on the atmospheric processes that affect wet and dry deposition (U.S. EPA 1997e). (Note that other sections of this report describe additional research and model development activities in these areas.)

To test the validity of the model, the wet deposition results from the RELMAP simulation were compared to limited measured data from the Mercury Deposition Network (MDN). The RELMAP estimates agreed with actual measurements within a factor of two in most cases and tended to underestimate measured wet deposition. Overall, the RELMAP model seemed to produce reasonable spatial patterns of annual wet deposition. Better spatial coverage by a long-term deposition network is needed to better evaluate the model. In addition, the high deposition rates estimated for some urban areas could not be verified because measurement data in urban areas are lacking (U.S. EPA 1997e).

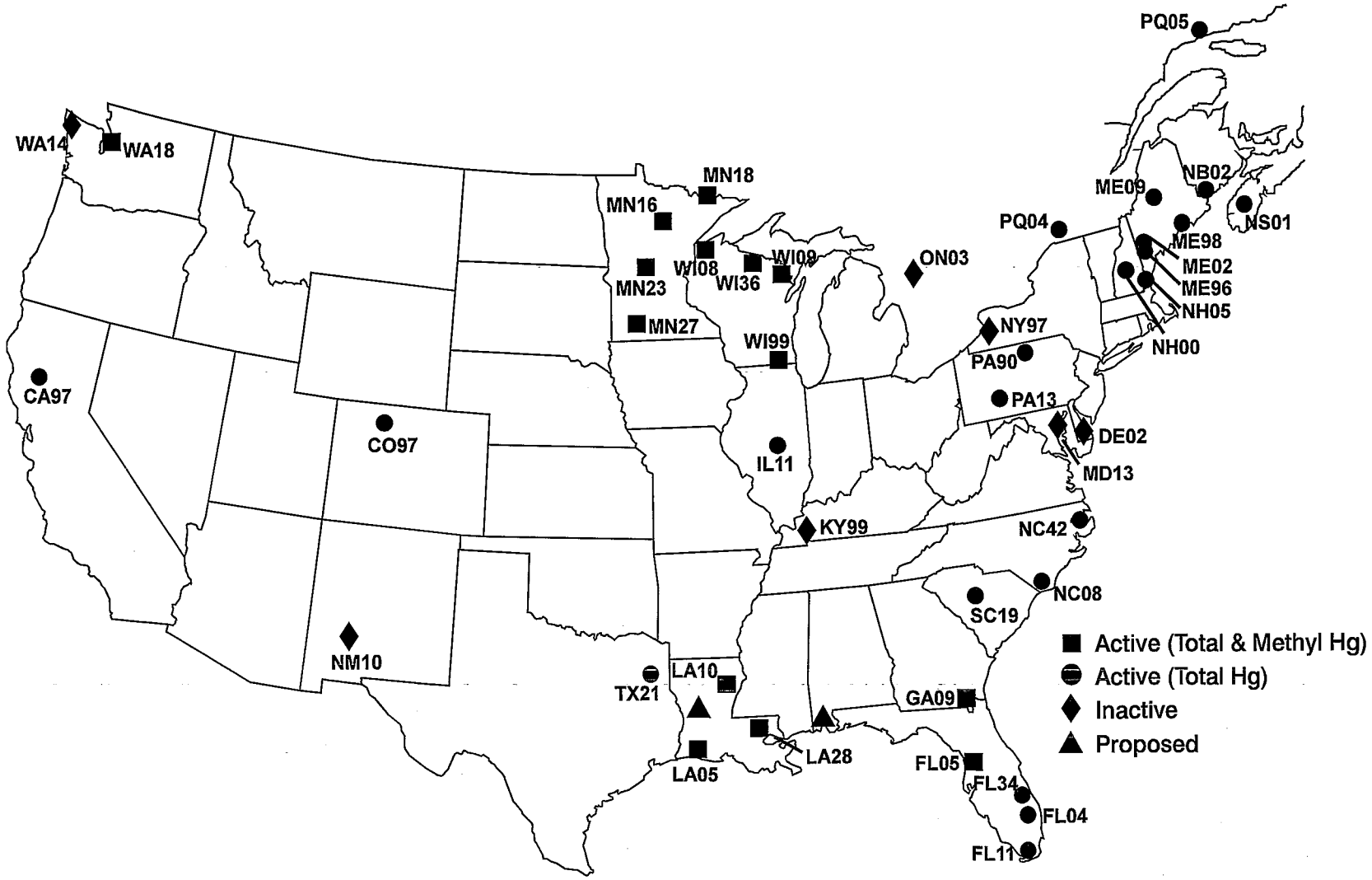
Mercury Emission Inventory by Percent Contribution by Source Category for Eight Northeast States

- Municipal waste combustion -- 45 percent
- Non-utility boilers -- 18 percent
- Electric utility boilers -- 13 percent
- Manufacturing sources -- 7 percent
- Area sources -- 6 percent
- Sewage sludge incineration -- 6 percent
- Medical waste incineration -- 5 percent

In addition to the *Mercury Study Report to Congress*, there have been a number of other recent modeling and monitoring studies of mercury deposition. For example, a regional-scale analysis, performed by the Northeast States and Eastern Canadian Provinces in collaboration with EPA, also illustrates the importance of local, regional, and global sources to mercury deposition levels (NESCAUM 1998). The purpose of this study was to determine whether deposition in the Northeast could be attributable to sources within the region or to long-range transport of mercury from upwind States. In this analysis, a RELMAP simulation suggested that approximately 47 percent of the modeled deposition in the Northeast is attributable to sources in the region, while approximately 30 percent is attributable to sources located outside the region. The remaining 23 percent was estimated to be from the global atmospheric reservoir, which includes mercury from U.S. emissions. The accompanying text box lists the source categories contributing to mercury emissions in the Northeast. As with the national EPA study, combustion sources account for more than 85 percent of the mercury emissions. Note, however, that the results of the simulation include considerable uncertainty.

The modeling predictions described above are based on average emissions over a year and as such do not provide an indication of trends over time. Two ways to assess trends in emissions and deposition are to consistently measure deposition over a wide geographic area over a long period of time, or to analyze sediment cores. Mercury monitoring of wet deposition at a number of sites began recently, and there are not enough data at present to evaluate trends over a widespread area. The Mercury Deposition Network (MDN), which began as a transition network of 17 sites in 1995, became an official subnetwork of NADP in 1996. Currently, nearly 40 sites are in operation throughout the U.S. and Canada, with sites concentrated on the east coast and in the Great Lakes basin (Figure II-2). However, the network is relatively sparse in its overall coverage, introducing uncertainty into trends analyses. The objective of the MDN is to develop a long-term database of weekly total mercury concentrations in precipitation and the annual and seasonal flux of total mercury in wet deposition. The present sites were not selected to be representative of the region and, in some cases, are highly influenced by local emission

Figure II-2
Monitoring Sites of the Mercury Deposition Network – 1999



sources. Site sponsors consist of U.S. and Canadian government agencies, State government agencies, universities, associations, and consulting firms. Currently, MDN data are available for 1996 and 1997.

Available MDN monitoring data from 1996 and 1997 indicate that the volume-weighted mean concentration of total mercury in precipitation from 22 sites ranged from 6.0 to 18.9 ng/L and annual deposition of mercury ranged from 2.1 to 25.3 $\mu g/m^2$ (or 2,100 to 25,300 ng/m^2). In 1997, average mercury concentrations in rain ranged from 6.2 to 18.3 ng/L at the 21 sites that had a full year of monitoring data and the average concentration for all sites was 10.6 ng/L. In 1996, average mercury concentrations at nine sites with a full year of data ranged from 6.0 to 14.1 ng/L with an average for all sites of 10.2 ng/L. In 1997, the annual average wet deposition of mercury for 21 sites ranged from 4.3 to 25.3 $\mu g/m^2$ (or 4,300 to 25,300 ng/m^2), whereas in 1996, the annual average wet deposition of mercury for nine sites ranged from 6.3 to 19.7 $\mu g/m^2$ (or 6,300 to 19,700 ng/m^2). In the eastern U.S., average summer mercury concentrations are more than double winter concentrations and average summer deposition values are more than three times winter values. This can be explained by higher concentrations of mercury in the rain and higher rainfall amounts during the summer. One interesting finding from the 1996 and 1997 MDN data is that several individual weeks were associated with unusually high mercury deposition amounts as a result of heavy rainfall and above average mercury concentrations. Based on National Weather Service information, the storms that contributed the precipitation in these weeks tracked along the east coast, including New Jersey, New York City, Providence, and Boston. Therefore, it is likely that emissions of mercury from these urban centers entered the air mass of the storm and were deposited at these MDN sites (Sweet et al. 1999).

While the MDN monitoring sites have not been in operation long enough to discern any temporal trends in mercury levels, one monitoring station has been in operation in the Lake Champlain basin at Underhill, Vermont since 1992. This is now the longest continuous monitoring program in the world for total mercury wet deposition, ambient particulate phase mercury, and ambient total gas phase mercury. Results do not show long-term trends in atmospheric mercury concentrations except perhaps for a downward trend for vapor phase mercury. Based on monitoring of wet deposition and modeling of dry deposition, total annual deposition of mercury in 1993 was estimated at 15.1 $\mu g/m^2$ (or 15,100 ng/m^2). However, research suggests that the dry deposition flux of mercury may be much greater and re-emission of mercury from forest ecosystems may be significant, introducing considerable uncertainty in the total wet plus dry deposition estimates (Hanson et al. 1995, Rea et al. 1998, Scherbatskoy et al. 1999, 1998, 1997).

The Underhill monitoring data collected between December 1992 and December 1997 also show seasonal variations. Deposition peaked in summer months when mercury concentrations in precipitation are highest, while the concentration of mercury vapor in the air showed only slight seasonal variation. The seasonal variations observed in mercury deposition at Underhill were similar to those observed in the Great Lakes basin (Hoyer et al. 1995, Burke et al. 1995). Concentration of particulate-associated mercury in the air in 1993-94 tended to be greatest in winter months at this site. Concurrent monitoring of mercury concentrations in stream water in a nearby forested catchment in the Lake Champlain basin showed dissolved concentrations to be fairly constant, but found that total mercury (dissolved plus particulate) was strongly correlated with flow rates. Thus, high flows may re-mobilize mercury adsorbed to sediment and organic matter. Also, maximum stream flows were often associated with spring snowmelt, which appears to be an important regulator of annual transport patterns of mercury within the Lake Champlain basin (Scherbatskoy et al. 1999, 1998, 1997, Shanley et al. 1999).

In another recent analysis, Pirrone et al. (1998) estimated atmospheric emissions and deposition of mercury in North America and compared these estimates to vertical profiles of mercury accumulation rates in sediment cores from four Great Lakes sites . The results of this analysis illustrate that

atmospheric deposition has been significant since the 1900s and still is the major contributor of mercury to the Great Lakes, although a variety of other sources (including direct discharges) also contribute to mercury inputs in the Great Lakes. Based on sediment core data and emissions estimates, Pirrone et al. calculated the atmospheric deposition flux of mercury to North America as a whole to be between 14.3 and 19.8 $\mu g/m^2/yr$ (1.43 to 1.98 $ng/cm^2/yr$), whereas in the Great Lakes region, the atmospheric deposition flux of mercury was calculated to be higher, at 135 $\mu g/m^2/yr$ (13.5 $ng/cm^2/yr$). This difference is likely due to local anthropogenic emissions and subsequent deposition of mercury in the Great Lakes region. Furthermore, mercury accumulation rates in sediment cores from the Great Lakes from pre-industrial to modern times increased from 0.7 to 235 $ng/cm^2/yr$ in Lake Ontario, from 0.8 to 65 $ng/cm^2/yr$ in Lake Michigan, and from 3 to 175 $ng/cm^2/yr$ in Lake Erie. All of these values are larger than those reported in sediment cores from small remote lakes in the northeastern U.S., indicating local and regional sources of mercury are deposited in the Great Lakes region rather than simply from the regional background in the northeastern U.S.

Engstrom and Swain (1997) analyzed sediment core data to detect recent trends in mercury emissions in the upper Midwest and found that, for a number of Minnesota lakes, mercury deposition peaked in the 1960s and 1970s and then declined. These declines were not seen in remote lakes in southeastern Alaska, indicating that deposition from the global pool had not declined. The decline in deposition inputs to Midwestern lakes can be attributed to reduced emissions from regional and local sources, which are thought to be largely due to pollution controls (particularly on waste incinerators) and a shift from coal to natural gas for residential and commercial heating.

Mass Balance Studies of Mercury

Preliminary results of the Lake Michigan Mass Balance Study (LMMBS; described in the *Second Great Waters Report to Congress*) are becoming available. Results from two studies (Hurley et al. 1998b, Landis 1998), which are discussed below, show varying results. The study by Hurley et al. (1998b) involved sampling eleven tributaries that are representative of a variety of watersheds as part of the LMMBS. For example, land-use patterns included agricultural, forest, wetland, and urban. Ten of the 11 tributaries sampled are associated with areas of concern due to past contamination. Sediment core and sediment trap analysis is still being conducted by EPA for the LMMBS samples; however, tributary sample analysis has been completed. The mean mercury concentration in the 11 tributaries was 6.9 ng/L, with the highest concentrations in the rivers running through the urban areas. The highest mean mercury concentration (36.0 ng/L) was observed on the Fox River running through Green Bay, while the lowest mean concentration (1.0 ng/L) was observed on the Muskegon River. In general, the northern rivers had lower tributary inputs than the more industrialized southern rivers.

Hurley et al. (1998b) calculated tributary loading to Lake Michigan from each river as the product of the daily average water discharge and the daily mercury concentration. The estimated annual tributary mercury flux was 230 kg/year. The total annual mercury input into Lake Michigan from the atmosphere and the tributaries during the LMMBS was estimated to be 1,419 kg/year. Atmospheric deposition was determined to be the dominant source of mercury inputs to Lake Michigan, contributing approximately 84 percent (1,189 kg/year) of the annual total.

The study by Landis (1998) involved a hybrid-modeling framework based on monitoring data in the Great Lakes region. Annual deposition estimates from Landis (1998) are provided in Table II-2. In addition, the Landis modeling effort indicated that the Chicago/Gary urban area was responsible for at least 19 percent of the total atmospheric deposition to Lake Michigan. It should be noted that reactive mercury deposition is not included in this estimate of the urban influence and, thus, likely represents an underestimate of the true impact of the Chicago/Gary urban area.

Table II-2
Preliminary Estimates of Total Atmospheric Mercury Deposition to Lake Michigan

Deposition	Annual Total (kg)	Annual Mean (µg/m^2)
Wet	614 ± 186	10.6 ± 3.2
Aerosol Dry	69 ± 38	1.2 ± 0.7
Reactive Gaseous Mercury[a]	506	8.8
Dissolved Gaseous Mercury[b]	-460	-8.0
Total	729	12.6

[a] Reactive gaseous mercury (RGM) deposition values do not include error bars because they reflect a sensitivity analysis performed on a single measurement in place of direct measurements since no measurement method was available for RGM at the time of the study.
[b] Dissolved gaseous mercury (DGM) deposition values do not include error bars because the values reflect a single measurement taken to establish modeling parameters.
Source: Landis 1998

In another effort, Hurley et al. (1998a) collected and analyzed field samples from the Fox River in Wisconsin, which is located in a highly industrialized area and feeds into Green Bay, and ultimately, Lake Michigan. While total mercury in this river is high relative to other tributaries feeding into Lake Michigan, most of the mercury is bound to sediments. In the Fox River, resuspended sediments appear to be the predominant source of mercury into Green Bay. However, in the Lower Fox River and Green Bay, methylmercury concentrations are relatively low, suggesting that particulate-bound mercury has limited bioavailability (Hurley et al. 1998a).

As part of the Chesapeake Bay Atmospheric Deposition Study (CBADS), mercury wet deposition, dry deposition, and gas exchange were measured at sampling sites in the Chesapeake Bay representative of regional background levels. Total atmospheric deposition of mercury to the Chesapeake Bay was estimated to be 80 kg/year. Wet deposition of mercury to the Chesapeake Bay surface waters was estimated to be 162.1 kg/year (± 10-20 percent). Dry deposition was estimated to be 32.4 kg/year (accurate within a factor of two or three). Gas exchange was a net output that was estimated to be -115 kg/year (± 40 percent). Note that to develop these estimates, it was assumed that 10 percent of the surface waters below the fall line (see Figure II-5) are impacted by urban deposition and that the loading estimates are sensitive to this assumed percentage value (Chesapeake Bay Program 1999a).

Several additional mass balance studies are ongoing in Lake Michigan, Long Island Sound, and the Chesapeake Bay to characterize mercury inputs to as well as losses from these systems (Table II-3). In Long Island Sound, Fitzgerald (1998) collected and analyzed precipitation, air, surface water, and biota samples to develop a mass balance for mercury. In the Chesapeake Bay, Mason et al. (1997) developed a mercury mass balance based on precipitation, throughfall, and stream water samples. The mass balance study of Lake Michigan by Mason and Sullivan (1997) is a smaller effort than the LMMBS and is primarily a mass-balance modeling exercise based on data extracted through a literature review.

Similarities among the various mass balance studies include the finding that atmospheric deposition is a pathway with enough relative contribution in these waters to be of consideration (e.g., from 10 to greater than 80 percent). Furthermore, several of these studies indicate that air-water exchange (through net volatilization) and sediment burial are two major loss pathways.

Table II-3
Mercury Sources Identified in Mass Balance Studies
(by percent contribution)

Location	Atmospheric Deposition (%)	Urban Area (%)	Riverine	Ground-water	Direct Discharge	Comments
Lake Michigan [a]	~ 80	~ 30 (Chicago)	~17	< 1	Not reported	Air-water exchange and sediment burial account for the major loss pathways from the lake, but atmospheric deposition dominates inputs
Chesapeake Bay [b]	> 50 (Wet 47-53) (Dry 9-11)	Baltimore (percent not known)	~33-49	Not reported	Not reported	Over 90 percent of the mercury entering the watershed is retained in the terrestrial system and does not reach the aquatic system
Long Island Sound [c]	~10	New York New Jersey (percent not known)	~ 52	Not reported	~ 36	Tidal exchange, sediment burial, and air-water exchange are major loss pathways. 45 percent of the mercury entering the Sound is re-emitted

[a] Mason and Sullivan (1997).
[b] Mason et al. (1997).
[c] Fitzgerald (1998).

Factors Affecting Mercury Fate and Transport in Watersheds and Tributaries

Recent research illustrates that a number of factors influence whether mercury that is deposited to watersheds or tributaries will be transported to the lake itself. The principal factors that affect mercury loading to the aquatic ecosystem appear to be (1) the amount of annual precipitation; (2) the influence of the urban air plume (in terms of local deposition of the contaminated plume); (3) storms and other events, like snowmelt, which influence stream flow and the resuspension of particle-bound mercury in sediments; and, (4) prevailing land use (e.g., forestry, agriculture, urban). Some of these factors appear to influence the amount of mercury in throughfall (i.e., precipitation that has washed through the forest canopy) and litterfall (i.e., fallen leaves) as well as the amount of mercury that is sequestered in organic soils which prevent its transport through the watershed.

Rea et al. (1996) found that throughfall and litterfall are the dominant components of mercury deposition in forested areas of the Lake Champlain basin. Monitoring during a 6-week period in August and September 1994 produced estimates of annual throughfall and litterfall deposition of 11.7 $\mu g/m^2/yr$ and 13 $\mu g/m^2/yr$, respectively. Thus, the total below-canopy deposition rate was estimated to be 24.7 $\mu g/m^2/yr$, considerably greater than the 15.1 $\mu g/m^2/yr$ wet plus dry deposition estimated by Scherbatskoy

et al. (1997). In the former study, precipitation deposition accounted for just 32 percent (or 7.9 $\mu g/m^2/yr$) of this estimated below-canopy deposition.

Several recent studies related to mercury cycling in the Lake Champlain basin, which focused primarily on forested systems, suggest that urban and agricultural systems may retain less atmospheric mercury than forested systems. For example, an analysis of mercury deposition and stream export (i.e., transport of mercury out of the basin through streamflow) in an 11 hectare, deciduous forested catchment (i.e., a portion of the larger watershed) in the Lake Champlain basin based on field sampling showed net annual stream export of mercury was less than 10 percent of wet deposition. Consistent with an earlier study in the basin, stream export was strongly related to flow and dominated by mercury in particulate form (i.e., mercury complexed with organic material). In 1 year of the study, fully one-half of the total annual export occurred on the single day of peak snowmelt. A more recent stream water sampling and deposition monitoring study also showed that most of the mercury transported in stream water is in the particulate phase, with the dissolved phase contributing only about 30 percent of the annual export. Analysis of soil water mercury reported in this study suggested that the transport of dissolved mercury in shallow soil water may be the primary mechanism for mercury movement from the forest floor to streams. These studies showed that the forested catchment is a net sink for atmospheric mercury and indicated that forests in the Lake Champlain basin help reduce the movement of mercury to the lake because the mercury is bound to soil organic matter. Some portion of the mercury is mobilized, however, and undergoes three possible fates: transport to streams through groundwater and overland (flood) flow, incorporation into vegetation by root uptake, or volatilization back to the atmospheric pool (Scherbatskoy et al. 1998, 1997, Shanley et al. 1999) .

Mercury Exposure and Effects

Once deposited, mercury may enter terrestrial and aquatic food chains. Mercury concentrations increase at successively higher levels in the food chain such that predators at the top of these food chains are potentially at risk from consumption of mercury in contaminated prey. Of the various forms of mercury in the environment, methylmercury has the highest potential for bioaccumulation in the food chain. In the *Mercury Study Report to Congress*, the Agency concluded that fish-eating birds and mammals are particularly at risk from mercury emissions and exposures. Ecosystems most at risk from airborne releases of mercury exhibit one or more of the following characteristics:

- Located in areas where atmospheric deposition of mercury is high;
- Include surface waters that are already impacted by acid deposition;
- Possess characteristics other than low pH (e.g., high organic content, long aquatic food chains) that result in high levels of bioaccumulation; and/or,
- Include sensitive species (U.S. EPA 1997e).

Although a large body of research exists on mercury exposure and effects, as discussed below and in previous *Great Waters Reports to Congress*, areas of uncertainty remain, particularly with respect to mercury methylation processes and their implications for exposure levels. The EPA and others are currently planning and conducting research projects to address the fate and transport of mercury compounds and subsequent exposures to methylmercury. Some examples of research under way related to methylmercury include investigations of the following:

- The role of a variety of biogeochemical parameters in watersheds, wetlands, and marine environments on mercury bioavailability, bioconcentration, and methylation;
- The effects of anthropogenic ecosystem changes on methylation rates, such as increased sulfate deposition, increased nutrient loads, and increased loadings of mercury due to soil disturbance;
- The role of sulfur (especially sulfide) in mercury methylation in watersheds as related to microbial activity; and
- The effects of agricultural sulfur on methylmercury in soils and wetland areas.

Effects of mercury can be two-fold. First, mercury in lower-level organisms can impact the health of those organisms. Next, these organisms become prey for higher-level organisms, resulting in higher-level organisms having higher concentrations of mercury than the lower-level organisms. Concentrations of methylmercury in fish tissue have been measured at levels one million times greater than the mercury concentration in the surrounding water column, illustrating the extent to which methylmercury can be concentrated in organisms.

Principal factors affecting the bioavailability of mercury in the waterbody include (1) whether it is bound to organic matter (which would inhibit bioavailability); and, (2) characteristics of the lake itself, such as surface area to volume ratio (with more shallow lakes having more bioavailable mercury). When conditions in a waterbody change (i.e., pH), the bioavailability of a compound can change without a change in the concentration of the compound.

Analyzing sediments for mercury concentrations can reveal the extent to which bottom dwelling organisms are exposed. Eskin et al. (1996) compiled and analyzed sediment contamination data from the Chesapeake Bay and its tributaries from 1984 to 1991. The potential for sediment contamination to impact aquatic life was evaluated by comparing median and maximum contaminant concentrations to no observed effects levels (NOELs) for aquatic biota, probable effects levels (PELs) for aquatic biota, or other aquatic health benchmarks. The NOELs are concentrations above which impacts on aquatic life are judged to be *possible*. Aquatic life impacts are considered *probable* at concentrations above the PELs. For toxic effects to occur, metals must not only be present at concentrations above aquatic life benchmarks, but must also be bioavailable.

Where data were available for comparison, sediment mercury concentrations in the Chesapeake Bay, in general, were lower in 1991 than in previous years. The median and maximum concentrations of mercury at all locations in the mainstem of Chesapeake Bay were 0.08 and 0.8 ppm, respectively, which are below the mercury PEL of 1.4 ppm for aquatic biota. However, because maximum concentrations of mercury in all segments of the bay exceeded the mercury NOEL of 0.1 ppm for aquatic biota, adverse impacts are still possible. The median and maximum mercury concentrations in tributaries were 0.1 and 4.66 ppm, respectively. Mercury concentrations were highest in the Middle River, other northwestern tributaries, and near the Sewells Point Naval Complex on the James River. Median mercury concentrations exceeded the mercury NOEL for aquatic biota in several tributaries, including the James and Potomac Rivers. The PEL concentration for mercury was exceeded only in the James River at stations near the Sewells Point Naval Complex.

Sources with episodic emissions or releases of mercury and other pollutants to the Delaware Bay influence concentrations in biota. Horseshoe crab eggs are of particular importance for monitoring efforts because they are a major food source for many species of migrating shore birds during the spring migration. While measurements of mercury levels in the eggs of horseshoe crabs from Delaware Bay,

New Jersey in 1993, 1994, and 1995 do not indicate any temporal trend, levels of mercury varied markedly among years, with the highest values in 1994 (Burger 1997). The variations in mercury levels and the peak in 1994 highlights the importance of episodic sources of pollutants to the Delaware Bay, which is situated downstream from highly industrialized areas in Trenton, New Jersey; Wilmington, Delaware; and, Philadelphia, Pennsylvania. Burger (1997) also analyzed mercury concentrations in female horseshoe crab leg muscle tissues from the Delaware Bay and found that mercury concentrations were higher in leg muscle tissues than in eggs and were indicative of few adverse effects to developing horseshoe crabs.

In an evaluation of higher level organisms, Brazner and DeVita (1998) detected mercury in yellow perch and spottail shiners from 23 sites in Green Bay, Lake Michigan. The overall distribution of mercury tissue concentrations was fairly uniform within the bay, indicating that mercury contamination originates primarily from non-point sources, including atmospheric deposition. Certain sites exhibited unusually high mercury residues in fish, however, suggesting these sites may be associated with direct discharges of mercury nearby. The mercury residue levels of 9.4 to 31.0 ng/g in yellow perch and 10.5 to 33.5 ng/g in spottail shiners were well below the International Joint Commission Aquatic Life Guideline for total mercury of 500 ng/g. Furthermore, these mercury levels were similar to or below the most recently reported (i.e., 1993, 1985) mercury levels in fish in most of the locations in the Great Lakes.

Mercury levels apparently decreased in yellow perch taken from the St. Lawrence River, which drains the Great Lakes, in 1991-1992 and were compared to results from a 1975 study (Ion et al. 1997). Mercury was detected in all yellow perch samples taken from various sites along the river. Comparison of average mercury concentrations to historical levels in St. Lawrence River fish indicates a two- to three-fold decrease since 1975. Because mercury contamination and bioaccumulation can vary considerably from site to site and species to species, additional studies of this type are needed for trends analysis of mercury levels in fish.

Fish-eating birds are of particular interest for evaluating the effects of mercury because relative to their body weight, exposures can be very high. Hughes et al. (1997) detected significant differences in mercury concentrations in both osprey eggs and chick feathers from nests at four Great Lakes study areas in Ontario (three natural lakes and one reservoir) and two Delaware Bay sites in New Jersey. Overall, eggs from the Ogoki Reservoir and chick feathers from St. Mary's River (Ontario) and the Ogoki Reservoir had significantly higher mercury concentrations than the eggs and feathers sampled from the other locations. Despite these geographic variations, mercury levels in osprey eggs, chick feathers, and adult feathers at all locations were below the levels associated with toxic reproductive effects, such as reduced number of eggs, decreased hatchability, increased hatchling mortality, and altered reproductive behavior. Fish sampled at each site also exhibited significant differences in mercury levels with a clear spatial pattern, similar to that observed for chick feathers. However, the study did not assess the relative impact of point source discharges and atmospheric deposition on spatial variation.

In a recent study that attempted to link spatial variations in mercury levels in biota with variations in atmospheric deposition of mercury, Evers et al. (1998a) found that mercury concentrations in the feathers and blood of adult and juvenile common loons from sites within five regions across North America (Canadian Maritimes, New England, Upper Great Lakes, northwestern U.S., and Alaska) increased significantly from western regions to eastern regions. Adults had average blood mercury levels which were 10 times higher than juveniles. Feather mercury levels increased significantly over a 4-year period, probably due to bioaccumulation of mercury over that time period. At some of the study locations where exposure is high (e.g., some eastern North American locations), the loon's natural mechanisms to excrete excess mercury (e.g., feathers, eggs, demethylation and storage in the liver and kidney) may not be sufficient to balance current exposure levels. The researchers note that the observed

geographical gradient agrees with EPA's Regional Environmental Monitoring and Assessment Program (R-EMAP) modeling of atmospheric deposition of mercury and predicted high impact areas (U.S. EPA 1997e) and the Northeast States and Eastern Canadian Provinces Mercury Study (NESCAUM 1998). Within the upper Great Lakes region (nine sites total), blood mercury levels were influenced the most by lake biogeochemistry and hydrology, rather than atmospheric deposition. Significantly higher concentrations from sites in north-central Wisconsin and central Ontario were detected. The authors conclude that loons breeding on low-pH lakes in the upper Great Lakes and in all lake types in northeastern North America are the most at risk from mercury contamination. A comparison of the measured blood mercury concentrations to health threshold levels determined from laboratory studies indicate that 12 to 31 percent of New England's breeding loon population exceed these levels (Evers 1998b). Based on feather mercury levels, over 30 percent of the male loons exceed threshold levels for health effects.

Like wildlife, fish consumption represents the dominant pathway for human exposure to methylmercury. Given the current scientific understanding of the environmental fate and transport of this element, it is not possible to quantify how much of the methylmercury in fish consumed by the U.S. population is contributed by U.S. emissions relative to other sources of mercury (such as natural sources and re-emissions from the global pool). As a result, it cannot be assumed that a change in total mercury emissions will be linearly related to any resulting change in methylmercury in fish, nor over what time period these changes would occur. This is an area of ongoing study.

The effects of methylmercury on humans and especially the developing fetus are well known and have been described in detail in earlier *Reports to Congress*. The critical target for methylmercury toxicity for humans is the nervous system. The factors that affect whether the mercury exposure is sufficient to cause health effects depend primarily on the species of fish consumed, the concentration of methylmercury in the fish, the quantity of fish consumed, and how frequently fish is eaten. The 1997 EPA *Mercury Study Report to Congress* includes a detailed analysis of potential public health impacts relating to fish consumption. This analysis was based on information on the mercury levels in various types of fish, dietary surveys, and EPA's current reference dose (RfD) for methylmercury. The RfD is an estimated daily ingestion level anticipated to be without adverse effect to persons, including sensitive subpopulations, over a lifetime. Note that the National Academy of Science is currently reviewing more recent mercury health effects studies and will recommend whether the EPA should revise its RfD for methylmercury. This review and recommendation is expected to be completed by mid-2000.

The *Mercury Study* report concluded that the typical U.S. consumer eating fish from restaurants and grocery stores was not in danger of consuming harmful levels of methylmercury from fish and is not advised to limit fish consumption on the basis of mercury content. However, eating more fish than is typical or eating fish that are more contaminated than typical fish can change the advice. Because the developing fetus is regarded as the most sensitive to the effects of methylmercury, women of child-bearing age are the population of greatest concern.

In addition, for cultural or economic reasons, some people in certain ethnic groups (e.g., Asians, Pacific Islanders, Native Americans) and subsistence fishers eat substantially more fish than the average consumer. Because of the higher amounts of fish in their diets, people in these groups, especially the women of child-bearing age, need to be aware of how much mercury is in the fish they consume. The *Mercury Study* report has more detailed information about amounts of mercury found in various types of fish.

The Food and Drug Administration (FDA) and State, local, and tribal agencies issue advisories that suggest limiting the consumption of contaminated fish. Mercury fish advisories in the U.S. have been

issued by 40 States and some tribes. Eleven States have Statewide advisories for mercury in freshwater lakes or rivers, and five States have Statewide advisories for mercury in coastal waters. Based on data from December 1997, 20 of 56 Great Waters are associated with mercury fish consumption advisories (U.S. EPA 1998m). The EPA is sponsoring work with the State of Wisconsin to assess current methods of communicating fish consumption advisories and to recommend improvements.

Despite this advice, a recent survey on awareness and effectiveness of fish advisories found that only half of the Great Lakes sport fish consumers were aware of health advisories related to fish consumption (Tilden et al. 1997). While one study of Lake Ontario anglers concluded that the angler subpopulation's health does not significantly differ from those experienced by the general North American population (Cole et al. 1997), a different study concluded that the consumption of contaminated fish affected time-to-pregnancy (i.e., the amount of time between when a women plans to become pregnant and the date of pregnancy). The study also found that 42 percent of the women surveyed reported consumption of contaminated fish from Lake Ontario, despite highly publicized advisories against the consumption of fish from Lake Ontario by women of reproductive age (Buck et al. 1997).

According to Tilden et al. (1997), the most widely accepted advisory recommendation was cleaning and cooking methods. While cooking methods are effective to reduce PCB exposure, mercury concentrations will actually increase, on a per weight basis, with all cooking methods (including pan frying, deep-frying, baking, boiling, and smoking) because of the moisture and fat lost during cooking (Morgan et al. 1997). Unlike PCBs which accumulate in fatty tissue that can be removed prior to consumption, mercury accumulates in the muscle tissue or filet portion.

Studies of mercury exposure in U.S. populations are limited, particularly for the general population. However, research is being planned as part of EPA's draft Mercury Research Strategy (see Chapter III) to address this need. A number of studies have been conducted involving sport fisherman and Native peoples, which have shown high levels of fish consumption in a number of populations; but, risk of exposure is dependent, in large part, on the level of contamination in the fish (U.S. EPA 1997e). A recent study of Ojibwa tribal members from the Great Lakes region indicates that mercury concentrations in Ojibwa populations are low; however, researchers identified several individuals with elevated levels of mercury (Gerstenberger et al. 1997).

OTHER METALS (LEAD AND CADMIUM)

Sources and Emissions of Lead and Cadmium

Lead and cadmium are naturally-occurring trace metals and, therefore, are expected to be detected at some background level in the environment, depending on the location. Furthermore, because lead and cadmium are metals, they are by nature persistent compounds that may cycle between environmental compartments. Therefore, while it is possible to estimate lead and cadmium emissions from anthropogenic sources, it is difficult to determine which sources (e.g., specific human-made sources, natural sources, cycling) contribute to total and atmospheric deposition inputs to the Great Waters.

Based on a study of lead, cadmium, and other trace metals emissions and air concentrations in the Great Lakes region from 1982 to 1993, (1) sources of lead in 1993 included steel manufacturing, coal combustion, non-ferrous metal production, and waste disposal (Figure II-3), and (2) sources of cadmium

in 1993 included coal combustion, solid waste and sewage sludge incineration, iron-steel manufacturing, non-ferrous metals production, and several smaller sources (Figure II-5) (Pirrone and Keeler 1996).

A review of cadmium and lead emission sources on a national scale, based on EPA's National Toxics Inventory for the time period from 1990-1993 (1990-1993 NTI), provides a somewhat different picture compared to regional sources for the Great Lakes (Tables II-4 and II-5). The larger national-level sources of lead air emissions are mobile sources, including non-road vehicles and equipment (aircraft) and on-road vehicles. The larger national-level sources of cadmium air emissions are secondary lead smelting, primary copper smelting, and primary lead production (U.S. EPA 1999a). The NTI data for lead air emissions also indicate that there are many source categories that are emitting similar amounts of lead compounds, rather than there being a few source categories contributing the majority of the air emissions. The differences observed between the local (Figures II-3 and II-5) and national (Tables II-4 and II-5) source characterizations for these pollutants highlight the importance of examining both local sources and regional sources, as well as the need to consider the influence of urban areas on nearby waterbodies.

HIGHLIGHTS
Other Metals (Lead and Cadmium)

➤ **Sources.** In some urban locations, local sources (versus regional sources) are primarily responsible for atmospheric deposition of lead and cadmium to the Great Lakes, whereas the opposite is true in some more remote locations. The largest national sources of lead air emissions are mobile sources, including non-road vehicles and equipment (aircraft) and on-road vehicles. Emissions and ambient air concentrations of lead have continued to decrease since the 1970s. The largest national sources of cadmium air emissions include secondary lead smelting, primary copper smelting, and primary lead production. Cadmium air emissions increased from 1982 to 1988 in the Great Lakes region and have not shown a trend since 1988.

➤ **Loadings.** In the Great Lakes region, lead deposition decreased from 1988 to 1994, and cadmium deposition may have decreased from 1992 to 1994. In the Chesapeake Bay, atmospheric deposition of cadmium and lead contributes no more than 5 to 7 percent, respectively, to total loadings. Total inputs from all pathways of lead and cadmium increased in the eastern section of the bay from 1966 to 1995 due to the increase in human population and industrial activities. However, trends in the relative contribution of atmospheric deposition of lead and cadmium to the Chesapeake Bay have not been discerned. In the Long Island Sound, lead inputs have been decreasing since 1980 and atmospheric deposition contributes 70-90 percent of total lead inputs.

➤ **Environmental Concentrations.** Sediment contamination studies in the Chesapeake Bay provide conflicting results – one study found increased lead and cadmium levels, possibly due to an increase in population, industrial activities, and agricultural activities, whereas another found decreasing levels. Lead and cadmium levels in Chesapeake Bay sediments, in general, were between the aquatic biota NOEL and PEL; however, bioavailability of these metals is low. In the Delaware Bay, lead levels decreased in horseshoe crab eggs in recent years, but cadmium levels were more variable.

A study of lead emissions and air concentrations indicates that, overall, U.S. lead emissions have decreased since the 1970s, primarily due to the phaseout of leaded gas. Lead emissions in the Great Lakes region decreased steadily from 1982 to 1993 at a rate of about 6.4 percent per year. Lead emissions in the Great Lakes region in 1988 were estimated to be 4,430 tpy. Lead concentrations in ambient air also showed steady declines over the entire region between 1982 and 1993. The regional decrease in lead emissions and air concentrations is correlated to the reduction of lead in gasoline (from 0.28 g/l in 1982 to 0.026 g/l in 1989) and an increase in unleaded gasoline use in both the U.S. and Canada since 1981. Sources of lead emissions also shifted over this time period. As shown in Figures II-3 and II-4, about 80 percent of lead emissions were from leaded gasoline use in 1982 compared to 1993 when most lead emissions were from four major industrial sources in the Great Lakes region (Pirrone and Keeler 1996). Another study of lead deposition to sediments in Central Park Lake in New York City

using sediment cores indicates, however, that municipal solid waste incineration contributed a large portion of the lead deposited during the same time period that lead from leaded gasoline was deposited. Furthermore, lead emissions from municipal solid waste incinerators decreased beginning in 1966 due to progressive facility closures following regulations. Therefore, although it is clear that lead emissions and deposition have decreased, it is possible that solid waste incineration has been underestimated as a source of atmospheric lead in the past, particularly to urban areas (Chillrud et al. 1999).

In contrast to lead emissions and ambient air concentrations, cadmium emissions in the Great Lakes region increased from 1982 to 1988 at a rate of 5.2 percent per year, with a peak in 1988 at 150 tpy. Since 1988, emissions of cadmium have not increased or declined (Pirrone and Keeler 1996).

Deposition of Lead and Cadmium

Data collected under the Integrated Atmospheric Deposition Network (IADN), a joint U.S.-Canada program that monitors pollutant levels in air and precipitation at remote sites in the Great Lakes region, by Eisenreich and Strachan in 1992 and Hoff et al. in 1994 show that total deposition of lead decreased from 1988 to 1994 in each of the Great Lakes. Paode et al. (1998) found that measured average lead fluxes in Chicago, Illinois were 0.07 $mg/m^2/day$, whereas over Lake Michigan, lead fluxes were 0.003 $mg/m^2/day$, demonstrating the urban influence of Chicago on atmospheric deposition of lead onto Lake Michigan. Dry deposition of cadmium in the Great Lakes region decreased from 1992 to 1994; however, this decrease may be due to improved averages in the particulate air concentrations of the metals (Hoff et al. 1996). Sweet et al. (1998) estimated annual wet and dry deposition rates to Lake Michigan, Lake Erie, and Lake Superior using air and rain concentrations of lead and cadmium measured at three U.S. IADN sites and found that lead and cadmium deposition levels were similar at all lakes. Since the IADN sites are situated in remote locations, this finding indicates regional atmospheric deposition, rather than local emissions, is the source of lead and cadmium at the remote sites on these three lakes.

Estimates of total inputs of trace metals to the Chesapeake Bay indicate that inputs of trace metals from the non-tidal watershed are much higher than trace metal inputs to tidal waters from point source discharges, urban storm water inputs, and direct atmospheric deposition to the tidal waters. The boundary for the non-tidal and tidal waters in the Chesapeake Bay region is indicated in Figure II-6.

Inputs from the non-tidal watershed to the Chesapeake Bay consist of all point and non-point sources, including atmospheric deposition. Atmospheric deposition (measured as direct deposition to water surfaces based on monitoring data) of cadmium and lead contributes approximately 4.6 and 5.6 percent, respectively, to total inputs to the Chesapeake Bay (Table II-6). Note that these percentages may be underestimates because the monitoring site locations do not reflect the urban influences of Baltimore, Maryland on the Chesapeake Bay. However, these percentages are similar to those for other metals but are much less than percentages for polycyclic aromatic hydrocarbon (Eskin et al. 1996).

Figure II-3
1993 Lead Emissions in the Great Lakes Region
(Pirrone and Keeler 1996)

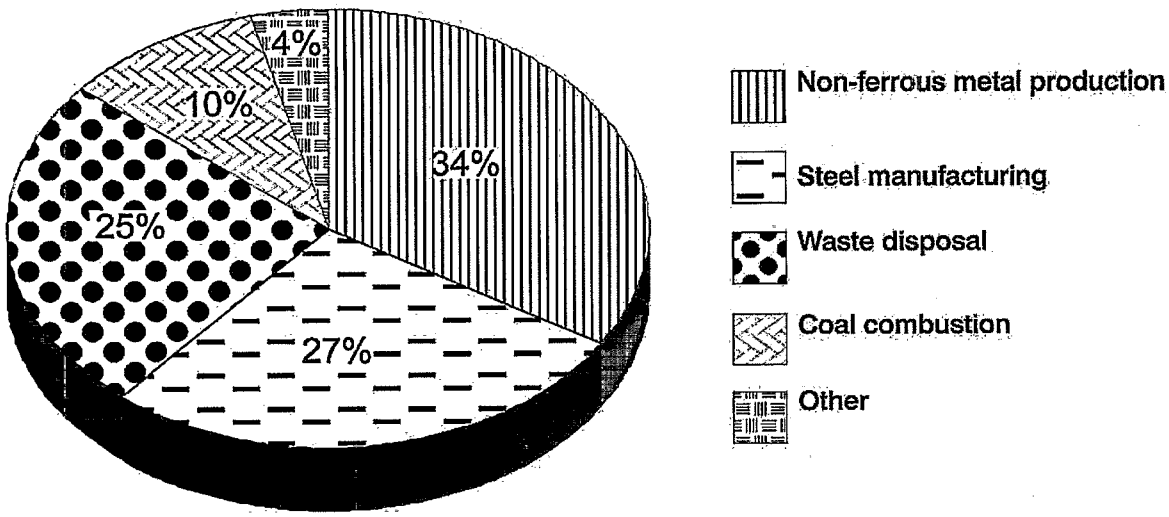


Figure II-4
1982 Lead Emissions in the Great Lakes Region
(Pirrone and Keeler 1996)

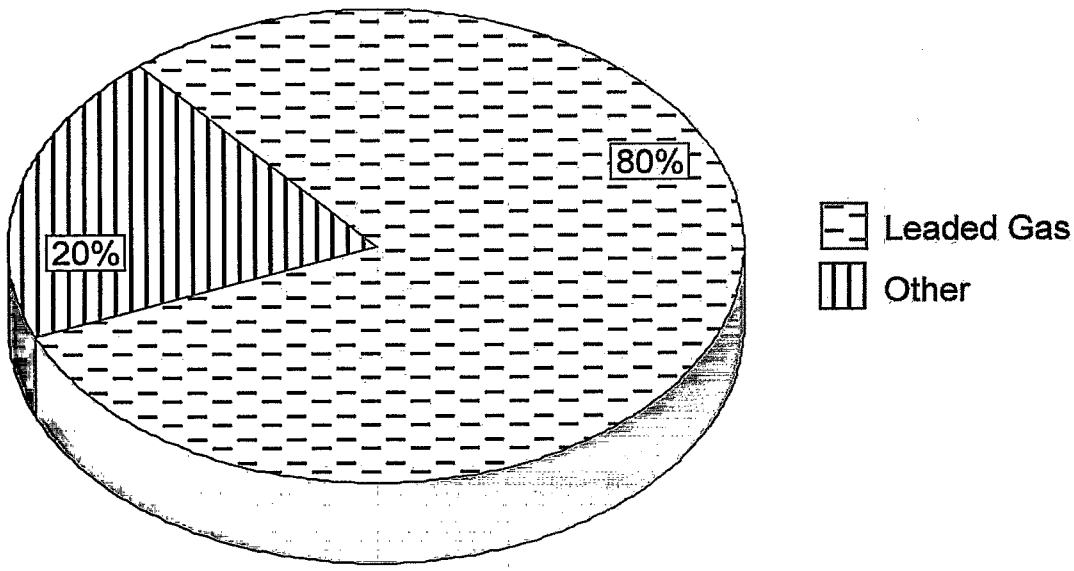


Figure II-5
1993 Cadmium Sources in the Great Lakes Region
(Pirrone and Keeler 1996)

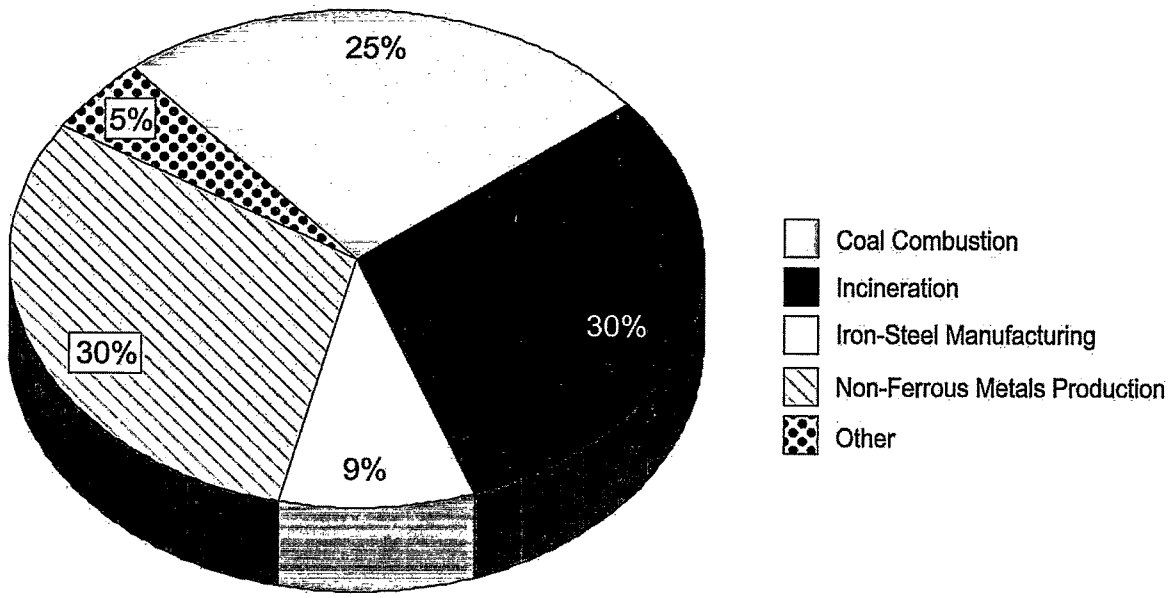


Table II-4
National Anthropogenic Lead Air Emissions
(Based on EPA's 1990-1993 National Toxics Inventory)

Source Category	Anthropogenic Air Emissions (tons/year)	Percent Contribution to Total U.S. Anthropogenic Air Emissions
Mobile sources: non-road vehicles and equipment - aircraft	619	18.7
Mobile Sources: on- road vehicles	418	12.6
Primary lead	259	7.8
Steel wire and related products manufacturing	163	4.9
Mobile sources: non-road vehicles and equipment - other	158	4.8
Primary copper smelting	152	4.6
Pulp and paper: combustion	150	4.5
Lead oxide in pigments	136	4.1
Secondary lead smelting	106	3.2

Source Category	Anthropogenic Air Emissions (tons/year)	Percent Contribution to Total U.S. Anthropogenic Air Emissions
Municipal waste combustion	80	2.4
Non-stainless steel manufacture - EAF	79	2.4
Utility boilers: coal combustion, all types	72	2.2
Secondary copper smelting	71	2.2
Medical waste incineration	63	1.9
Hazardous waste incineration	56	1.7
Secondary nonferrous metals production	55	1.7
Pressed and blown glass and glassware manufacturing	52	1.6
Storage batteries manufacturing	51	1.5
Portland cement manufacture: hazardous waste-fired	41	1.2
Primary nonferrous metals production	40	1.2
Paint application: no spray booth	35	1.1
Others (< 1 percent each) [a]	454	13.7
Total U.S. Anthropogenic Lead Air Emissions	3,310[b]	100[b]

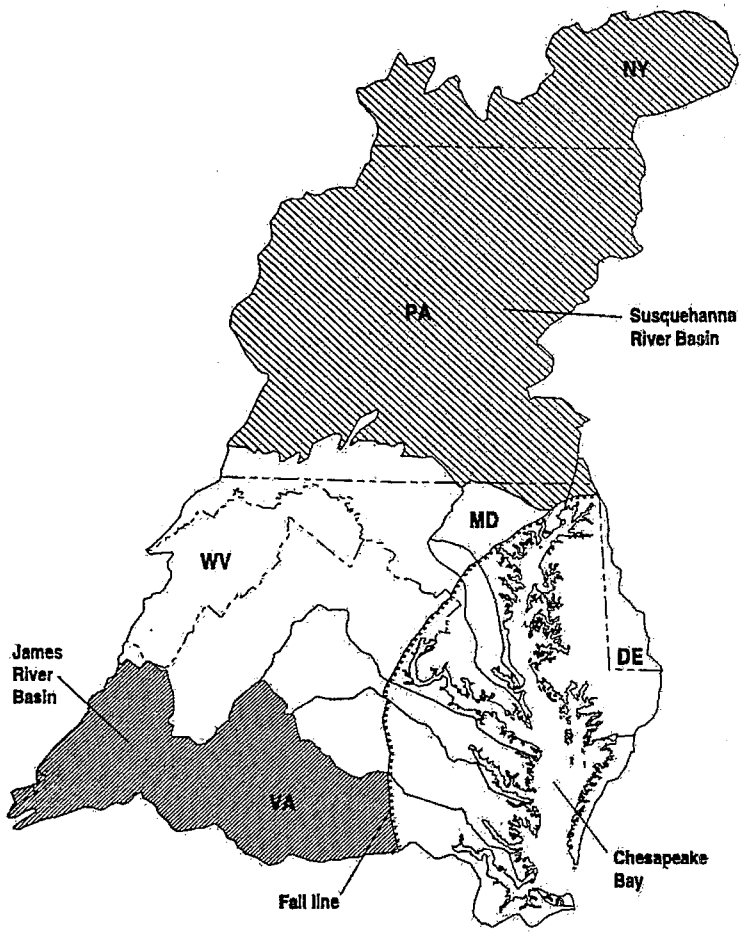
[a] A list of the source categories that contribute less than 1 percent to total U.S. emissions is provided in Appendix B.
[b] This value represents anthropogenic lead air emissions in the U.S. only.
Source: U.S. EPA 1999a

Table II-5
National Anthropogenic Cadmium Air Emissions
(Based on EPA's 1990-1993 National Toxics Inventory)

Source Category	Anthropogenic Air Emissions (tons/year)	Percent Contribution to Total U.S. Anthropogenic Air Emissions
Secondary lead smelting	84	42.2
Primary copper smelting	16	8.1
Primary lead	16	7.9
Hazardous waste incineration	9	4.3
Petroleum refining: catalytic cracking units	7	3.3
Municipal waste combustion	5	2.5
Medical waste incineration	5	2.4
Cadmium refining and cadmium oxide production	5	2.3
Secondary copper smelting	5	2.3
Industrial inorganic chemical manufacturing	4	2.2
Sewage sludge incineration	4	1.9
Cadmium stabilizers production	4	1.8
Pulp and paper: combustion	3	1.7
Portland cement manufacture: hazardous waste-fired	2	1.2
Inorganic pigments: cadmium pigments in plastics	2	1.1
Primary nonferrous metals production	2	1.0
Others (<1 percent each) [a]	28	13.8
Total U.S. Anthropogenic Cadmium Air Emissions	201[b]	100[b]

[a] A list of the source categories that contribute less than 1 percent to total U.S. emissions is provided in Appendix B.
[b] This value represents anthropogenic cadmium air emissions in the U.S. only.
Source: U.S. EPA 1999a

Figure II-6
Chesapeake Bay Tidal and Non-tidal Waters
(Tributary basins are shaded)



Note: Fall line is the upper boundary of the tidal waters of the Chesapeake Bay.
Source: Chesapeake Bay Program 1994

Table II-6
Trace Metal Inputs to the Chesapeake Bay (kg/year)

Trace Metal	Total Inputs to Chesapeake Bay[a]	Direct Atmospheric Deposition to Tidal Waters	Direct Atmospheric Deposition to Tidal Waters as Percent of Total Inputs
Cadmium	27,800	1,200	~4.6
Lead	275,300	14,500	~5.6

[a] Includes inputs from the non-tidal watershed (including atmospheric deposition to the watershed), storm water inputs to the tidal waters, point source inputs to the tidal waters, and direct atmospheric deposition to the tidal waters.
Source: Eskin et al. 1996

Data collected by the Chesapeake Bay Atmospheric Deposition Study (CBADS),[1] as presented in the Chesapeake Bay Program's *1999 Toxics Loading and Release Inventory* (TLRI), result in estimates of total inputs to the Chesapeake Bay for cadmium and lead (42,600 and 254,000 kg/year, respectively) that are similar to the Eskin et al. (1996) values. Inputs of lead and cadmium via atmospheric deposition to the tidal waters of the Chesapeake Bay as a percentage of the total inputs are 7 percent and 3 percent, respectively. Inputs from the non-tidal watershed dominate for lead and cadmium (Chesapeake Bay Program 1999a, 1994).

A study on the eastern shore of the Chesapeake Bay in Maryland using sediment cores found that lead deposition rates range from 0.32 ± 0.20 to 13.60 ± 0.98 mg/m^3/yr, and cadmium deposition rates range from 0.01 ± 0.01 to 0.43 ± 0.04 mg/m^3/yr. Total inputs of both lead and cadmium from all pathways increased from 1966 to 1995 due to the increase in human population and industrial activities (Karuppiah and Gupta 1998).

Direct atmospheric deposition rates of lead and cadmium to the Massachusetts Bay based on monitoring data are presented in Table II-7. In general, deposition of lead and most other metals is higher at the monitoring site closer to Boston due to higher urban area emissions. However, deposition of cadmium is similar at both the urban and rural locations. While there is not a seasonal trend for trace metal deposition in general, higher dry deposition of lead has been observed in the winter (Golomb et al. 1997a).

Table II-7
Direct Atmospheric Deposition of Lead and Cadmium to Massachusetts Bay (μg/m^2/yr)

Trace Metal	Urban	Rural
Cadmium	260	280
Lead	2,300	1,400
Total	2,560	1,680

Source: Golomb et al.1997a

In Long Island Sound, sediment cores from salt marshes indicate that lead inputs reached a maximum between 1970 and 1980 at approximately 62 μg/cm^2/yr and have been decreasing since that time at most sites. Atmospheric deposition to the water surface of Long Island Sound contributes approximately 90 percent of total lead inputs. At sites closer to New York City, cores show that atmospheric deposition accounts for approximately 70 percent of lead inputs, suggesting that non-atmospheric sources (e.g., direct discharges, storm water runoff) play a larger role near the urban areas (Cochran et al. 1998).

[1] CBADS measurements include wet deposition, dry deposition, and gas exchange of pollutants at rural sites, with estimations for urban areas.

Lead and Cadmium Levels in the Environment

Lead and cadmium levels have been measured in a variety of environmental media and in a wide range of geographic locations in the Great Waters. The concentrations that are observed in the environment are a result of atmospheric deposition from both domestic and international sources as well as from non-atmospheric sources, including storm water runoff and direct discharges to waterbodies and tributaries.

A recent study in the Chesapeake Bay found an increase in lead and cadmium concentrations in and toxicity of the sediments and pore water (i.e., interstitial water of sediments) of two tributaries (Wicomico River and Pocomoke River) in comparing sediment cores from 1966-1975 to sediment cores from 1986-1995 (Table II-8). The authors examined the historical land use of areas adjacent to these rivers and concluded that the increases were likely due to a variety of pathways. Specifically, (1) increases in metal concentrations in Wicomico River sediments and pore water are probably due to an increase in population, industrial activities, and the volume of the sewage treatment plant influent; (2) increases in metal concentrations in Pocomoke River sediments and pore water are probably due to the increased runoff from agricultural areas, including poultry farms; and, (3) increases in lead concentrations in Wicomico River sediments and pore water are possibly due to an increase in various industrial activities, commercial facilities that use lead compounds, and atmospheric deposition (Karuppiah and Gupta 1998).

Seasonal Variation in Cadmium Concentrations

A monitoring study of the St. Lawrence River, which connects the Great Lakes with the Atlantic Ocean, emphasized the importance of seasonal variation on the concentration and distribution of metals. Quémerais and Lum (1997) collected water from sites in the river and four of its tributaries from March to November 1991 and April to June 1992 and analyzed the samples for dissolved and particulate cadmium. The mean dissolved cadmium concentration for the basin (10 ± 5 ng/L) was in the same range as that measured for rivers considered pristine environments. The mean particulate cadmium concentration (1.3 ± 1.1 µg/g) was also relatively low in comparison to other waterbodies. Dissolved cadmium concentrations exhibited a seasonal trend; concentrations increased in spring and autumn and decreased in summer. The trend was attributed to increased runoff and leaching from soil during spring snowmelt and rainfall events in autumn as well as uptake by surface water microorganisms during summer. The seasonal variation of particulate cadmium was not as apparent, probably due to the negative influence of suspended particulate matter, which is higher in spring and autumn as a result of snowmelt and rainfall patterns.

Other studies have found that sediment concentrations of lead and cadmium are declining, however. Sediment concentration data from the Chesapeake Bay Program from 1984 to 1991, for example, indicate that lead and cadmium concentrations in sediments are declining in some areas of the Chesapeake Bay (Eskin et al. 1996).

An analysis of spatial trends shows that metals concentrations in sediments of the mainstem of the bay were lowest at the head of the bay and in the extreme lower bay. The highest metals concentrations were detected in sediments of the segment of the bay closest to Baltimore, the largest urban center on the bay. Lead contamination trends were somewhat different when concentrations were normalized by the fine particle content of the sediment. In particular, normalized sediment concentrations of lead gradually declined toward the mouth of the bay, suggesting that the Susquehanna River is the primary source of lead. Lead concentrations were highest in the Middle River, other northwestern tributaries, and near the Sewells Point Naval Complex on the James River. The distribution of cadmium contamination was different from other contaminants, where the highest concentrations were located in the sediments of the James (near the Sewells Point Naval Complex) and Patuxent Rivers and in some of the southeastern tributaries (Eskin et al. 1996).

Table II-8
Temporal Changes in Metal Concentrations in Chesapeake Bay Tributaries Based on Sediment Cores

Sediment Core Depth (cm)	Estimated Period (year)	Lead		Cadmium	
		Sediment (mg/kg)	Pore Water (mg/L)	Sediment (mg/kg)	Pore Water (mg/L)
Wicomico River					
0-7.5	1986-1995	18.01	5.12	0.49	0.026
7.5-15.0	1976-1985	7.60	0.54	0.48	0.008
15.0-23.0	1966-1975	7.43	0.42	0.37	0.007
Pocomoke River					
0-7.5	1986-1995	5.14	0.74	0.28	0.025
7.5-15.0	1976-1985	3.51	0.59	0.24	0.003
15.0-23.0	1966-1975	2.35	0.59	0.20	0.002

Source: Karuppiah and Gupta 1998

The potential for sediment contamination to impact aquatic life was evaluated by comparing median and maximum sediment contaminant concentrations to aquatic health benchmarks in both the mainstem of the Chesapeake Bay and its tributaries (Table II-9). Maximum concentrations of cadmium and lead in the mainstem of Chesapeake Bay were below PEL concentrations for aquatic biota. However, impacts must be considered possible because maximum lead and cadmium concentrations exceeded the aquatic biota NOEL in most of the upper half of the bay and at the mouth of the Potomac River (Eskin et al. 1996).

In the Chesapeake Bay tributaries, median and maximum lead concentrations in sediments were 31 and 343 parts per million (ppm), respectively. Median lead sediment concentrations exceeded the lead NOEL in northwestern tributaries, but the aquatic biota PEL for lead was exceeded only near the Sewells Point Naval Complex. The median and maximum cadmium sediment concentrations were 0.6 and 6.0 ppm, respectively. The distribution of cadmium contamination was different from other contaminants, where the highest concentrations were located in the James (near the Sewells Point Naval Complex) and Patuxent Rivers and in some of the southeastern tributaries. Median cadmium sediment concentrations in several of the western and northwestern tributaries exceeded the aquatic biota NOEL; however, the maximum concentrations did not exceed the PEL for aquatic biota (Eskin et al. 1996).

For toxic effects to be possible, metals must not only be present at concentrations above aquatic life benchmarks but must also be bioavailable (i.e., available for uptake by organisms). For instance, a portion of the lead in sediments must not be bound to the organic matter in the sediment, and the lead levels in the sediment must exceed the aquatic life benchmark in order for toxic effects to occur. Eskin et al. (1996) found low bioavailability through most of the mainstem of the bay. However, bioavailable conditions were found in a northern reach of the bay from Turkey Point to Annapolis. This reach includes the most contaminated segment of the bay, between Baltimore and Annapolis (Eskin et al. 1996).

Table II-9
Cadmium and Lead in Sediments of the Chesapeake Bay Mainstem

Contaminant	Concentration		Aquatic Life Benchmark		Location of Maximum Concentrations	Trends
	Median (ppm)	Maximum (ppm)	NOEL[a] (ppm)	PEL[b] (ppm)		
Cadmium	0.4	2.9	1.0	7.5	Baltimore region south to the Little Choptank River; mouth of Potomac River	Concentrations are declining
Lead	35	86	21	160	Baltimore region	Concentrations are declining in some areas

[a] No Observed Effect Level. Level above which impacts are considered "possible."
[b] Probable Effect Level. Level above which impacts are considered "probable."
Source: Eskin et al. 1996

In the Delaware Bay, one study attributed decreases in lead levels in horseshoe crab eggs to a general lowering of pollutants to the bay. Horseshoe crab eggs provide a bioindicator of pollutant levels in the Delaware Bay because they are laid beneath the sand at the bay's high tide line and are continuously inundated with water. Burger (1997) measured levels of lead and cadmium in the eggs and leg muscles of horseshoe crabs in 1993, 1994, and 1995 in the Delaware Bay and found a decrease in lead levels in horseshoe crabs from 1993 to 1995. Cadmium levels in the eggs were significantly higher in 1994 than in 1993 or 1995 (Table II-10). The variations in cadmium levels may suggest an episodic source of cadmium to the bay, which is probable given the high level of upstream industrialization in Trenton, New Jersey; Wilmington, Delaware; and, Philadelphia, Pennsylvania. Burger (1997) also analyzed lead and cadmium levels in female horseshoe crab leg muscle tissues from the Delaware Bay and found that lead and cadmium levels in leg muscle tissues were lower than in eggs and were indicative of few adverse effects.

Table II-10
Lead and Cadmium in Horseshoe Crab Eggs from Delaware Bay

Year	Lead (ppb)	Cadmium (ppb)
1993	558 (±160)	17 (±5)
1994	206 (±34)	310 (±130)
1995	87 (±8)	24 (±7)

Source: Burger 1997

COMBUSTION EMISSIONS

Dioxins and Furans

Polychlorinated dibenzo-p-dioxins (PCDDs; dioxins) and polychlorinated dibenzofurans (PCDF; furans) are compounds that are similar in structure but differ in the number and position of chlorine atoms. Much of the current research uses toxic equivalent quantities (TEQs) to express quantities of dioxins and furans. The TEQs aggregate all of the dioxin and furan congeners into a single factor that is weighted by toxicity of the individual congeners based on ratios to 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD) and to 2,3,7,8-tetrachlorodibenzofuran (2,3,7,8-TCDF). Therefore, dioxin and furan quantities are commonly expressed as dioxin TEQs and furan TEQs, respectively. This is the convention followed in this report to the extent permitted by the research results reported in the literature.

HIGHLIGHTS
Dioxins and Furans

➤ **Sources.** The largest sources of dioxin and furan emissions in the U.S. are municipal waste combustion and medical waste incineration. By 2002, controls applied to these sources will reduce their dioxin and furan emissions by 95-99 percent (see Chapter III).

➤ **Loadings.** In the Great Lakes region, sediment accumulation rates indicate a decline in dioxin and furan inputs since the 1970s. The relative atmospheric contribution and sediment accumulation rates of dioxins and furans entering the Great Lakes differs both between and within the lakes. In some lakes, the atmospheric contribution of dioxins and furans is 100 percent, whereas in other lakes, the atmospheric contribution of dioxins and furans is less than 5 percent. In general, the pattern of industrialization around the lakes influences relative contributions and accumulation rates.

➤ **Concentrations in Biota.** In general, long-term monitoring data indicate that dioxin and furan concentrations in biota in many Great Waters have declined over time.

Dioxins and furans are byproducts of combustion that enter the environment primarily as a result of waste incineration and, to a lesser extent, various manufacturing processes, but also through natural combustion processes. Different sources emit different dioxin and furan compounds in certain combinations. These combinations, or source profiles, can then be used to identify the source of the emissions. One such study developed a chemical mass balance model to determine the contribution of different types of sources to sediment levels in the Housatonic River in Connecticut, Lake Huron, and the Baltic Sea. The study found that air particulate matter, coal-fired power plants, municipal incinerators, and pentachlorophenols (from manufacturing) were, in general, the largest sources of dioxins and furans in sediments. The same study reported that coal-burning was the major source of dioxins and furans prior to 1955 in the U.S. and 1970 in Europe, with municipal incinerators dominating after that time (Su and Christensen 1997). These results are similar to recent EPA estimates of national air emissions and sources of dioxin and furan TEQs which indicate that of the 0.0027 tpy of dioxin and furan TEQ air emissions, municipal waste combustion accounts for 47 percent and medical waste incineration accounts for 24.6 percent (Table II-11). A large number of smaller sources also emit dioxins and furans (U.S. EPA 1999a).

Emissions data for the U.S. and Canada were used with NOAA's hybrid single particle Lagrangian integrated trajectory (HYSPLIT)/transfer coefficient (TRANSCO) computer program to analyze dioxin and furan sources and their contributions to atmospheric deposition of dioxins and furans in the Great Lakes. For any given lake, the simulation results indicated that approximately half of the atmospheric deposition of dioxins and furans arose from sources in the immediately surrounding States and provinces, and half arose from the remainder of the U.S. and Canada. Furthermore, the relative contribution of sources by class were ranked, showing that municipal waste incineration, iron sintering, cement kilns burning hazardous waste, and medical waste incineration are responsible for at least 85 percent of the total atmospheric deposition of dioxins and furans to the Great Lakes. This study also

found that sources of dioxins as far away as central Canada may influence deposition of dioxins to the Great Lakes (Cohen et al. 1995).

Table II-11
National Anthropogenic Dioxin and Furan TEQ Air Emissions
(Based on EPA's 1990-1993 National Toxics Inventory)

Source Category	Anthropogenic Air Emissions (tons/year)	Percent Contribution to Total U.S. Anthropogenic Air Emissions
Municipal waste combustion	0.0012	47.0
Medical waste incineration	0.0007	24.6
Secondary aluminum smelting	0.0002	7.2
Utility boilers: coal combustion, all types	0.0001	4.2
Industrial boilers: wood/wood residue combustion	0.00009	3.4
Mobile sources: on-road vehicles	0.00009	3.4
Open burning: wildfires and prescribed burnings	0.00009	3.4
Portland cement, excluding hazardous waste-fired	0.00004	1.5
Residential wood/wood residue combustion	0.00004	1.5
Wood treatment/wood preserving	0.00003	1.1
Others (< 1 percent each) [a]	0.00007	2.6
Total U.S. Anthropogenic Dioxin and Furan TEQ Air Emissions	0.0027[b]	100[b]

[a] A list of the source categories that contribute less than 1 percent to total U.S. emissions is provided in Appendix B.
[b] This value represents anthropogenic dioxin and furan TEQ air emissions in the U.S. only.
Source: U.S. EPA 1999a

Sediment core studies have also been used to determine the relative atmospheric contribution of dioxins and furans. One sediment core study indicates that the relative atmospheric contribution of dioxins and furans differs by lake based on comparisons of pollutant compositions and accumulation rates among cores to estimates of atmospheric deposition (Table II-12). In the Great Lakes region, accumulation rates of dioxins and furans in sediments began to increase in the early 1940s. Maximum accumulation rates were observed in 1970 ± 10 years, and current accumulation rates are 30 to 70 percent of the maximum levels (Pearson et al. 1997a). Current accumulation rates of dioxins and furans in sediments indicate inter- and intra-lake variations. For example, the current dominant source of dioxins and furans in Lake Superior is believed to be the atmosphere because sediment accumulation rates of dioxins and furans are similar to those of lakes in which the atmosphere is the only possible source of dioxins and furans. In southern Lake Michigan near the Chicago urban area, the atmosphere contributes most of the dioxins in the lake, but 65 to 95 percent of furans entering southern Lake Michigan are estimated to be from non-atmospheric sources. In northern Lake Michigan, the analysis of sediment cores indicates that approximately 50 percent of dioxins and greater than 75 percent of furans are from non-atmospheric sources. From 65 to 95 percent of dioxins and greater than 95 percent of the furans

accumulated in Lake Ontario are estimated to be from non-atmospheric sources based on the analysis of sediment cores (Pearson et al. 1998).

Table II-12
Current Rates of Dioxins and Furans Accumulation in Great Lakes Sediments

Location	Number of Sediment Cores Analyzed	Accumulation Rates in Sediment ($pg/cm^2/yr$)		Percent Contribution from Atmospheric Sources	
		Dioxins [a]	Furans [a]	Dioxins [a]	Furans [a]
Lake Superior	2	7.4 - 8.0	0.8 - 0.9	100	100
Northern Lake Michigan	2	44 - 49	22 - 25	33 - 55	5 - 35
Southern Lake Michigan near Chicago Urban Area	1	17	17	100	5 - 35
Lake Ontario	3	120 - 220	130 - 230	5 - 35	<5

[a] Includes the total of all dioxin homologs. Includes the total of all furan homologs.
Source: Pearson et al. 1998

To help explain and understand the inter- and intra-lake variations in loadings and accumulations of dioxins and furans, the HYSPLIT/TRANSCO computer program was used to estimate the amount of dioxin and furan emitted from identified sources to air and water that enter the five Great Lakes (Cohen et al. 1995). The model was run using 1993 source and emissions data from 1,329 identified sources in the U.S. and Canada. As shown in Table II-13, the modeled deposition flux of dioxins and furans increases from Lake Superior, Lake Huron, Lake Michigan, Lake Erie, to Lake Ontario, following the pattern of industrialization around the lakes. In addition, modeled waterborne inputs (i.e., direct discharges) of dioxins and furans to Lake Superior, Lake Michigan, and Lake Huron play a lesser role than air deposition. For Lake Michigan, these relative source findings are different from those found by Pearson et al. (1998) who used a sediment core methodology to estimate relative atmospheric contributions. In Lake Erie and Lake Ontario, the modeled waterborne inputs are uncertain because of the difficulty in quantifying discharges to and from inter-lake channels (Cohen et al. 1995).

Table II-13
Modeled Air Deposition, Depositional Flux, and Waterborne Inputs of Dioxins and Furans to the Great Lakes

Dioxins and Furans	Superior	Huron	Michigan	Erie	Ontario	Total
Atmospheric Deposition (g TEQ/yr)	5.6	8.6	13.7	7.3	6.4	42
(range)	(2-17)	(3-25)	(5-43)	(2-21)	(2-18)	(13-124)
Depositional Flux ($\mu g/km^2/yr$)	69	145	238	284	337	172
Waterborne Inputs (g TEQ/yr)	1.4	1.4	1.9	11	>3.9	>19.6
Percent Contribution from Atmospheric Sources	80	86	88	40	~62	~68

Source: Cohen et al. 1995

Biota in many Great Waters are contaminated with dioxins and furans (Firestone et al. 1996, Metcalfe et al. 1997a, 1997b, Wade et al. 1997b). Recent studies of contamination of biota in the Great Waters implicate non-atmospheric sources of dioxins (Firestone et al. 1996, Wade et al. 1997b) and furans (Wade et al. 1997b); however, the relative importance of atmospheric sources and non-atmospheric sources remains unclear. In many instances, long-term monitoring indicates that concentrations of dioxins and furans have declined over time. For example, the U.S. Food and Drug Administration monitored the presence of dioxin in the edible portions of fish and shellfish from major U.S. waterways from 1979 to 1994, and the analytical results indicate that levels in aquatic species from most waterways are declining steadily (Firestone et al. 1996). In several locations (e.g., Chesapeake Bay, Delaware Bay, Gulf of Mexico, and Galveston Bay), no dioxin was found in fish samples taken between 1987 and 1994. In addition, Huestis et al. (1997) reported that levels of dioxins and furans in Lake Ontario trout collected from 1977 to 1993 have declined substantially, although most chemical levels appear to have reached a steady state or are declining more slowly now.

Polycyclic Organic Matter

Polycyclic organic matter (POM[2]), like dioxins and furans, is a byproduct of fossil fuel combustion and natural combustion processes (e.g., forest fires), and consists of a group of compounds with similar chemical structures. The POM compounds are emitted in distinct patterns from various source types, allowing the identification of emission sources from emission "fingerprints" (or patterns) in contaminated sediments. Within this class of POM are the polycyclic aromatic hydrocarbons (PAHs). In most cases, human-made sources of PAHs account for the majority of PAHs released to the environment (Simcik et al. 1996). These compounds are usually the POMs of concern as many PAHs are known or suspected human carcinogens. Therefore, the discussion of POMs focuses on PAHs.

HIGHLIGHTS
Polycyclic Organic Matter

➤ **Sources.** Nationally, the largest source of PAHs is consumer products usage, along with many smaller emission sources. Studies in the Great Lakes region also indicate that vehicular emissions, coal-fired power plants, and coke and steel production are large sources of PAH emissions. Note that multiple conventions are used to report POM and PAH quantities because of the differences in health effects of the variety of POM and PAH compounds.

➤ **Loadings.** In general, PAH deposition levels follow the pattern of population density and urbanization. In the Great Lakes region, maximum sediment accumulation levels of PAHs occurred between 1950 and 1975, after which accumulation levels declined. Data from recent years in the region, however, do not indicate a clear trend. In the Chesapeake Bay, data indicate that total loadings for several PAH compounds decreased in recent years, but data collection methodologies were altered over the same time period, making it difficult to assess the true trend.

➤ **Environmental Concentrations.** Concentrations of PAHs in the sediments of tributaries and mainstem of the Chesapeake Bay are below the aquatic biota probable effects level (PEL), but are above the no observed effect level (NOEL) for aquatic biota in the northern bay. In the tributaries, PAH sediment concentrations declined substantially between 1987 and 1991.

There is not one standard convention to measure and express emissions and quantities of POM. Multiple conventions have been developed because of the differences in the health effects of the variety of POM and PAH compounds. Some of the measures that are used include 16-PAH (i.e., the 16 PAH compounds that can be measured using EPA test method 610), 7-PAH (i.e., the seven PAH compounds that are probable carcinogens), or extractable organic matter (EOM) (i.e., the solvent

[2] The abbreviation, POM, is also used by ecologists, limnologists, and oceanographers for Particulate Organic Matter.

extractable portion of organic matter). These conventions, however, are not necessarily followed by independent researchers and thus are not always used in this report.

Based on EPA's 1990-1993 national inventory of air emissions, the dominant 16-PAH emission source is consumer products usage (e.g., metal cleaning, solvents, household cleaning products, and personal care products), along with many other smaller emission sources (Table II-14) (U.S. EPA 1999a).

Table II-14
National Anthropogenic 16-PAH Air Emissions
(Based on EPA's 1990-1993 National Toxics Inventory)

Source Category	Anthropogenic Air Emissions (tons/year)	Percent Contribution to Total U.S. Anthropogenic Air Emissions
Consumer products usage	5,733	33.2
Open burning: wildfires and prescribed burnings	2,540	14.7
Aerospace industry (surface coating)	1,697	9.8
Petroleum refining: other sources not distinctly listed	783	4.5
Primary aluminum production (from ESD)	662	3.8
Pulp and paper: kraft recovery furnaces	649	3.8
Coke ovens: charging, topside, & door leaks	538	3.1
Coke ovens: pushing, quenching, and battery stacks	517	3.0
MON - continuous processes	440	2.5
Gasoline distribution stage II	374	2.2
Gasoline distribution stage I	355	2.1
Petroleum refining: catalytic cracking units	313	1.8
Open burning: scrap tires	294	1.7
Industrial Organic chemicals manufacturing	227	1.3
Pulp and paper: lime kilns	183	1.1
Others (< 1 percent each) [a]	1,967	11.4
Total U.S. Anthropogenic 16-PAH Air Emissions	17,271[b]	100[b]

[a] A list of the source categories that contribute less than 1 percent to total U.S. emissions is provided in Appendix B.
[b] This value represents anthropogenic 16-PAH air emissions in the U.S. only.
Source: U.S. EPA 1999a

In the Great Lakes and other waterbodies, several researchers analyzed sediment cores to identify spatial and temporal trends in PAH deposition and sources, as discussed below. Researchers estimated relative contaminant contributions from specific local human-made sources (e.g., smelters, coke ovens) along with regional sources (e.g., fuel oil combustion, wood burning). In general, recent sediment core

studies show regional deposition from oil and wood burning sources with significant contributions from local sources in industrialized areas.

In Lake Michigan sediments, PAHs originate primarily from vehicular emissions; wood, oil, and natural gas burning for home heating; coal-fired power plants; and, coke and steel production. A study by Simcik et al. (1996) that focused on the urban complex of Chicago, Illinois and Gary, Indiana concluded that the dominant source of PAHs to the entire lake from around 1900 to the present is emissions from coke and steel production. This conclusion differs from that of Karls and Christensen (1998) who found a regional historical pattern in central Lake Michigan, with a significant contribution from wood-burning and an increasing dominance of oil-burning sources (as opposed to coal-burning by coke and steel production), which is consistent with U.S. fuel consumption data. Karls and Christensen also found that PAH loadings at Green Bay, the Fox River, and the Kinnickinnic River were strongly influenced by local industrial activities, primarily coke production at the Milwaukee Solvay Coke Company which operated from 1900 to the 1970s. These differing conclusions support the finding that local urban sources of PAHs are different and often play a larger role in deposition and sediment accumulation than regional sources.

Other studies also indicate that urban sources largely influence PAH deposition. For instance, recent sediment layers in the Milwaukee Harbor area indicate that in urban areas, highway dust can be a significant source of PAHs from emissions of gasoline engine exhaust tar (Christensen et al. 1997). In a carbon particle sediment core study, the relative abundance of oil particles in sediments increased in recent years and is explained by an increase in automobile traffic (Karls and Christensen 1998). Sharma et al. (1997) found that source contributions of PAHs to the St. Mary's River (which connects Lake Superior and Lake Huron) were strongly influenced by traffic density and urban storm water runoff from impervious street surfaces.

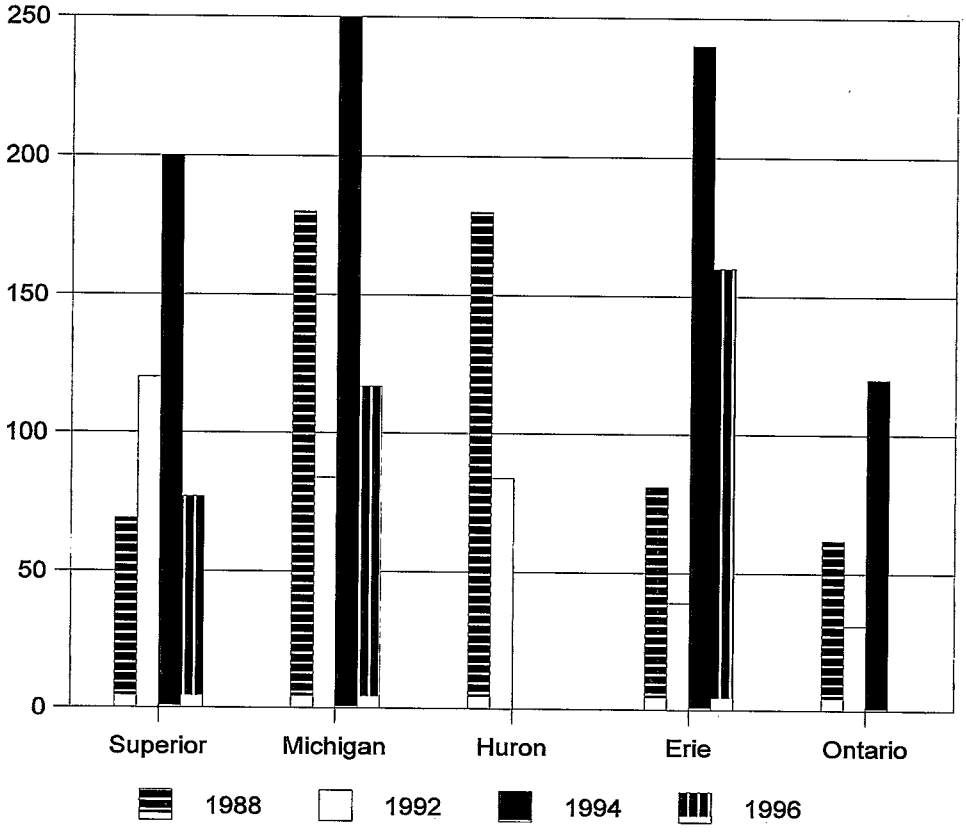
These recent findings for PAHs follow the pattern that was illustrated for several other pollutants of concern:

- Local point sources and regional sources both play a role in the deposition of pollutants to waterbodies;
- Urban areas play a strong role whether through the proximity of industrialized areas to waterbodies or through urban runoff; and,
- The relative contribution to pollutant loadings by different types of emission sources shifts over time depending on industrial activities, pollution controls, and other factors such as trends in fuel use.

Lake Michigan sediment cores show that PAH accumulation began between 1880-1900, consistent with increased PAH emissions from coal combustion during industrialization. Maximum PAH accumulation occurred between 1950-1975 (Simcik et al. 1996). Karls and Christensen (1998) found similar results. Maximum accumulation rates were lower in the southern basin near the Chicago urban area (70 $ng/cm^2/yr$) than in the northern basin (100-150 $ng/cm^2/yr$) because of the south to north transport of sediment-bound PAHs. One study observed a slight decrease in PAH sediment accumulation rates in recent years in some cases due to a switch from coal to oil and natural gas and because of industrial emissions controls (Simcik et al. 1996). In contrast, recent deposition monitoring data from the IADN program do not indicate any clear trend in total atmospheric loadings of PAHs, as indicated by data for benzo[a]pyrene (B[a]P), to the Great Lakes from 1988 to 1996 (Figure II-7). Trends in PAH deposition

levels, in general, are difficult to discern because of (1) the degree of uncertainty in loading calculations and (2) the fluctuations in ambient air concentrations (U.S. EPA 1998l).

Figure II-7
IADN Loading Estimates of B(a)P to the Great Lakes (kg/year)
(U.S. EPA 1998i)



As part of the Lake Michigan Mass Balance Study (LMMBS) and the Atmospheric Exchange Over Lakes and Ocean Surfaces (AEOLOS) study, which examine the influence of urban air emissions on deposition to the Great Waters, Franz et al. (1998) found that dry deposition of PAHs to Lake Michigan contributes approximately 5,000 kg/year. Wet deposition estimates from previous studies are lower at approximately 1,600 kg/year. Franz et al. (1998) also found that dry deposition of PAHs is higher in Chicago than 15 km offshore and at rural sites because of (1) the greater degree of anthropogenic activity within the Chicago area, and (2) the greater atmospheric burden and subsequent deposition of large particles generated within that area. These findings are consistent with the hypothesis that larger particles are generated in urban areas and are deposited closer to the sources than smaller particles.

In the Massachusetts Bay, as in other regions, the sum of wet and dry deposition of PAHs is greater at monitoring sites in urban areas (i.e., closer to Boston and Logan Airport) than at monitoring sites in rural areas (i.e., Truro), with the exception of biphenyl and naphthalenes (Table II-15). Biphenyl and naphthalenes may be more likely to deposit at the rural site because these pollutants are more water soluble and, therefore, may be deposited as wet deposition from clouds carrying biphenyl and naphthalenes from other geographic areas. Deposition of PAHs in this region is highest in the winter, possibly due to increased use of fossil fuels during winter for residential and commercial heating and

wood burning in stoves and fireplaces, or due to the lower mixing heights in the atmosphere which occur during winter in North America (Golomb et al. 1997b).

Table II-15
Sum of Wet and Dry Deposition of PAHs onto Massachusetts Bay (µg/m²/year)

PAH	Urban	Rural
Acenaphthene	5.0	1.8
Acenaphthylene	5.2	3.3
Anthracene	9.5	3.7
Benzo[ghi]perylene	29.2	4.1
Biphenyl	4.3	7.7
Chrysenes	98.8	9.9
Dibenzothiphenes	34.8	13.2
Fluoranthene	129.0	23.9
Fluorenes	50.0	23.0
Naphthalenes	206.6	240.0
Perylene	11.5	3.2
Phenanthrenes	215.1	41.9
Pyrenes	97.8	16.9

Source: Golomb et al. 1997b

In the Chesapeake Bay, estimates from one study of total inputs and atmospheric inputs of PAHs indicate that direct atmospheric deposition to the tidal waters of the Chesapeake Bay range from 30 percent to 56 percent (Table II-16). Estimates of the relative atmospheric contribution for benzo[a]anthracene and chrysene may be slightly high because estimates were not provided for point sources of benzo[a]anthracene and non-tidal watershed inputs for chrysene. Highest loadings occur in the West Chesapeake and Potomac regions. Intermediate loadings occur in the James and Patuxent regions, and lowest loadings occur in the Rappahannock, York, and eastern shore regions (Eskin et al. 1996). Annual net inputs (in terms of atmospheric deposition processes and the largest source of riverine inputs) of nine PAH compounds to the Chesapeake Bay are provided in Table II-17 based on sampling and analysis of surface water and air. In general, gas phase absorption (i.e., absorption of pollutants in the gas phase in the atmosphere by the water surface) of PAHs is higher than estimates of wet and dry deposition loadings; however, wet and dry deposition become more important loading factors for higher molecular weight PAHs (Nelson et al. 1998).

In contrast, the analysis of CBADS monitoring data in the Chesapeake Bay Program's *1999 Toxics Loading Release and Inventory* (TLRI) provides additional and more recent information on total inputs and relative atmospheric deposition inputs of PAHs (including benzo[a]pyrene, chrysene, phenanthrene, and pyrene) to the tidal and non-tidal portions of the Chesapeake Bay (see Figure II-6 for the delineation of the tidal and non-tidal waters in the Chesapeake Bay watershed). Total inputs to the tidal waters of the Chesapeake Bay of benzo[a]pyrene, chrysene, phenanthrene, and pyrene are estimated to be 29,000 kg/year, 29,000 kg/year, 59,000 kg/year, and 40,000 kg/year, respectively. Total atmospheric deposition (wet, dry, and gas exchange) ranges from < 0.5 percent (35 kg/year) for

benzo[a]pyrene to 5 percent (2,900 kg/year) for phenanthrene. Inputs of PAHs from the non-tidal watershed account for less than 3 percent of the total PAH inputs to the Chesapeake Bay (Chesapeake Bay Program 1999a).

Table II-16
Estimates of PAH Inputs to the Chesapeake Bay (kg/year)

PAH	Total Inputs to Chesapeake Bay	Direct Atmospheric Deposition to Tidal Waters	Direct Atmospheric Deposition to Tidal Waters as Percent of Total Inputs
Benzo[a]anthracene	380	130	34
Benzo[a]pyrene	430	130	30
Chrysene	570	320	56
Fluoranthene	1,300	640	49
Naphthalene	1,500	NE	NE

NE = no estimate
Source: Eskin et al. 1996

Table II-17
Annual PAH Inputs to and Losses from the Chesapeake Bay

PAH	Annual Inputs and Losses (kg/year)			
	Susquehanna River[a]	Wet deposition	Dry aerosol deposition[b]	Net gas deposition (or volatilization)[c]
Fluorene	122	16	12	379
Phenanthrene	450	63	92	2875
Anthracene	NA	6	6	130
2-Methylphenanthrene	NA	NA	NA	702
1-Methylphenanthrene	NA	NA	NA	280
Fluoranthene	1130	70	120	679
Pyrenes	1030	75	109	361
Benz[a]anthracene	376	9	34	(4.6)
Chrysene	330	29	85	29

[a] The Susquehanna River is the largest source of freshwater to the Chesapeake Bay.
[b] Calculated from measured ambient aerosol particle contaminant concentrations and annual average dry aerosol deposition velocity of 0.49 cm/s.
[c] Values in parentheses represent net volatilization from rather than net deposition to the waterbody.
Source: Nelson et al. 1998

In a study to quantify the gaseous exchange fluxes across the air-water interface at rural, semi-urban, urban, and industrialized sites of the southern Chesapeake Bay, researchers found that aerosol particle-associated PAH concentrations measured in the atmosphere were similar at the four sites and PAH gas-phase concentrations measured in the atmosphere were as much as 50 times greater at the urban site than at the rural site. Furthermore, at the rural site (Haven Beach, VA), PAH gas-phase

concentrations in the atmosphere are decreasing with time. Specifically, in 1991, the PAH gas-phase concentration was 6,203 pg/m^3 whereas in 1994-1995, PAH gas-phase concentrations were only 4,550 pg/m^3. This study also analyzed seasonal variations and did not find any seasonal variations in PAH gas-phase concentrations at this site. At the non-rural sites, an exponential increase in PAH gas-phase concentrations was observed with temperature increases, indicating that PAHs volatilize from contaminated surfaces near these sites in warmer weather (Gustafson and Dickhut 1997).

In a sediment contamination study of the Chesapeake Bay and its tributaries from 1984 to 1991 by Eskin et al. (1996), PAHs were most concentrated in sediments of the northern bay between Turkey Point and the mouth of the Middle River, just north of Baltimore. Maximum PAH concentrations in sediments (14,854 ppm) at all locations in the bay were below the aquatic biota PEL for total PAHs of 28,000 ppm. However, maximum concentrations exceeded the NOEL of 2,900 ppm for aquatic biota over much of the northern bay north of Annapolis. In the Chesapeake Bay tributaries, PAHs were most abundant in sediments of the western, northwestern, and northeastern tributaries. The highest concentrations were located in the Sassafras River. However, concentrations in sediments in tributaries declined dramatically between 1987 and 1991. Median PAH sediment concentrations in tributaries rarely exceeded aquatic biota NOELs and never exceeded aquatic biota PELs. Thus, toxic effects from PAH sediment contamination are possible in the northern part of the mainstem bay, but are not likely in the tributaries.

NITROGEN AND COMPOUNDS

Nitrogen is an ubiquitous element in the natural environment and is essential to all forms of life. It is a component of protein and genetic material and plays a key role in photosynthesis. Nitrogen cycles through all terrestrial and aquatic ecosystems and in many systems is a primary factor determining the nature, diversity, and productivity of the ecosystems. Diatomic nitrogen gas (N_2), an inert, largely biologically inactive gas, comprises almost 78 percent of the earth's atmosphere. Despite the presence and essential nature of nitrogen throughout the natural environment, excessive levels of certain nitrogen compounds in the atmosphere, deposited to surface media or introduced to aquatic systems, can be detrimental to terrestrial and aquatic habitats and potentially to human health.

Nitrogen compounds are released to the atmosphere from a variety of sources, both natural and anthropogenic. Nitrogen compounds can be present in the atmosphere in a variety of inorganic and organic forms, in gaseous, liquid, or solid (particulate) states. Table II-18 identifies the most common forms of atmospheric nitrogen and summarizes their key sources and properties. As this table indicates, the most important forms of atmospheric nitrogen from the standpoint of atmospheric deposition and loadings to surface waters include the following.

- ***Nitric Oxide and Nitrogen Dioxide (NO and NO_2, or collectively NO_x).*** Nitrogen oxide compounds are the key forms of anthropogenic nitrogen released to the atmosphere and are precursors to the formation of nitrogen compounds that are ultimately deposited to the Great Waters. Most of the nitrogen emitted to the air from anthropogenic sources is in the form of NO_x, primarily NO. Once released to the atmosphere, NO_x is transformed through a variety of complex photochemical reactions to oxidized nitrogen compounds and, ultimately, aerosol nitrate and nitric acid. These end products are deposited to the surface and impact surface waters. Nitrogen oxide also is a catalyst in the formation of ozone.

- ***Nitric acid (HNO_3), Aerosol Nitrate (NO_3), Ammonia and Ammonium (NH_3 and NH_4^+, or collectively NH_x), and Organic Nitrogen Compounds.*** These are the nitrogen compounds that are deposited from the atmosphere to surface media and impact surface waters. Reduced nitrogen compounds (NH_x) compounds (e.g., ammonia, ammonium) also account for a significant portion of nitrogen emissions from anthropogenic (as well as biogenic) sources.

Discussions of atmospheric nitrogen emissions, deposition, and effects in this report focus on these compounds. In particular, the term "atmospheric nitrogen" hereafter refers to these compounds and excludes N_2, the inert, natural component of the earth's atmosphere.

HIGHLIGHTS
Nitrogen Compounds

➤ **Sources.** Sources of anthropogenic nitrogen emissions to air include on-road vehicles, fuel combustion for electricity generation, farm animal wastes, non-road vehicles and engines, fuel combustion by industry, fertilizer application, biomass burning, and miscellaneous other minor sources. Anthropogenic nitrogen oxide (NO_x) emissions rose rapidly through the late 1970s. Nitrogen oxide emissions have leveled off since around 1980, mainly due to controls under the CAA. Nationwide trends in emissions of other compounds cannot be determined with available data.

➤ **Loadings.** The rate of anthropogenic nitrogen compound deposition from the air to the Great Waters rose through the late 1970s, but has leveled off since around 1980. Trends in atmospheric deposition nitrogen (ADN) loading to the Great Waters are uncertain, but have probably been static for the past two decades. ADN loadings to various Great Waters along the east coast account for roughly 10 percent to 40 percent of total nitrogen loadings.

➤ **Human and Ecological Effects.** The effects of ADN on the Great Waters is indistinguishable from the effects of nitrogen from other sources. Anthropogenic nitrogen loads to the Great Waters are currently many times natural rates. Mechanisms for eliminating or assimilating nitrogen are overwhelmed in many waterbodies, leading to nitrogen buildup. Effects of excess nitrogen include algal blooms, eutrophication, anoxia, loss of species diversity, and fundamental changes to ecosystem structure. These effects are observed with increasing frequency and extent in many of the Great Waters of the east and Gulf coasts. In sufficient concentrations, some nitrogen compounds have toxic effects on humans. However, nitrogen compounds generally do not cause exceedances of drinking water standards in the Great Waters.

Nitrogen Emissions and Atmospheric Transport

In the natural environment, diatomic nitrogen gas (N_2) is converted to biologically available forms of nitrogen (e.g., NO_x, ammonium compounds) through nitrogen fixation processes such as lightning and biological fixation. Nitrogen emissions from lightning have been estimated at less than 10 million metric tons per year. Releases of fixed nitrogen from marine ecosystems are less certain and range from less than 30 to greater than 300 million metric tons per year. Nitrogen fixation in terrestrial ecosystems, prior to human influences, ranged from 90 to 140 million metric tons per year; however, these emission levels have increased due to human alterations to the nitrogen cycle (Vitousek et al. 1997).

Of the various forms of nitrogen emitted to the atmosphere through anthropogenic activities, only NO_x emissions have been quantified adequately to allow evaluation of nationwide emissions trends. Nationwide emissions of NO_x generally reflect trends in population, economic activity, and industrialization for most of this century. Nitrogen oxide emissions steadily increased in the first three decades of the century, but declined in the 1930s due to the lower economic activity during the Great Depression. With the start of World War II, NO_x emissions nationwide began a rapid upward trend that continued well into the 1970s. Between 1940 and 1970, total nationwide NO_x emissions increased by a factor of three, from 6.5 million to 19 million metric tons per year (U.S. EPA 1997f, U.S. EPA 1997h).

Table II-18
Common Forms of Atmospheric Nitrogen

Compound	Notation	Sources	State in the Atmosphere	Effects
Inert Compounds				
Diatomic nitrogen gas	N_2	Comprises approximately 78 percent of the earth's atmosphere	Gaseous	Inert; no direct effects. Converted to other compounds through certain biological and physical processes
Nitrous oxide	N_2O	Wide variety of natural sources, fertilizer production and use, fossil fuel combustion, industrial acid production, microbial denitrification in soil and water	Gaseous	Not reactive in lower atmosphere; potent greenhouse gas, 320 times as effective as CO_2 in trapping heat in the atmosphere
Oxidized Compounds				
Nitric oxide	NO	Natural soil emissions, lightning, combustion processes with temperatures greater than 2200 °C	Gaseous	Precursor in the formation of NO_2, nitrates, nitric acid, and ozone in the atmosphere, and to NO_2, nitrate, and nitric acid deposition
Nitrogen dioxide	NO_2	Combustion processes with temperatures greater than 2200 °C, atmospheric oxidation of NO	Gaseous	Highly reactive; a variety of acute and chronic human health effects; precursor or catalyst in the formation of nitrates, nitric acid, and ozone in the atmosphere; deposits slowly to surface media; precursor to nitrate and nitric acid deposition
Aerosol nitrate	NO_3	Photochemical oxidation of NO_x (i.e., NO and NO_2) in the atmosphere; combustion processes. Partitioning of HNO_3 to the aerosol phase by ammonia gas in the atmosphere; microbial oxidation of ammonium in soil and water	Fine particles, dissolved in precipitation	Possible mutagen; variety of adverse health effects associated with fine particles; deposits to surface media. Excess NO_3 in drinking water has adverse health effects, including methylglobinemia in infants. Potent nutrient stimulant of plant growth, contributing to eutrophication in waterbodies
Nitric acid	HNO_3	Photochemical oxidation of NO_x in the atmosphere.	Vapor, liquid aerosol, dissolved in precipitation	Deposits quickly to surface; is the source of most oxidized nitrogen deposited to the surface; causes surface water acidification, property damage; detrimental to aquatic biota and ecological systems. Contributes to NO_3 concentrations and related health effects in drinking water. Potent nutrient stimulant of plant growth, contributing to eutrophication in waterbodies

Compound	Notation	Sources	State in the Atmosphere	Effects
Other oxides of nitrogen	PAN, N_2O_5, others	Intermediate reservoir products in atmospheric chemical reactions forming HNO_3 from NO_x	Gaseous	Eye irritant; possible mutagen; precursor or catalysts in formation of ozone; PAN is an important oxidant in the atmosphere and a component of photochemical smog
Reduced Compounds (NH_x)				
Ammonia	NH_3	Volatilization from animal wastes; natural microbial decomposition of organic matter in oceans and soils; manufacture and application of fertilizers; hydrolysis of urea in fertilized soils; biomass and fossil fuel combustion	Gaseous, vapor, dissolved in precipitation	Highly reactive; reacts to form NH_4^+, establishing an NH_4^+/NH_3 equilibrium in the atmosphere. Deposits to the surface; potent nutrient stimulant of plant growth, contributing to eutrophication in waterbodies
Ammonium	NH_4^+	Formed in the atmosphere from reaction of NH_3 with atmospheric acids such as H_2SO_4 and SO_4, microbial transformation of organic nitrogen, microbial fixation in soil and water	Particulate, dissolved in precipitation	Can be transported long distances; ultimately deposits to the surface; with NH_3 contributes as much as 20-40 percent of atmospheric nitrogen deposition to coastal waters. Potent nutrient stimulant of plant growth, contributing to eutrophication in waterbodies.
Organic Compounds				
A wide variety of compounds of unknown composition, believed to include urea, pollen and, amino acids	Various, mostly unknown	Primarily released from biological processes; also released from anthropogenic processes and formed in the atmosphere from chemical and photochemical reactions of NO_x, hydrocarbons, and O_3	Various	Comprises10-30 percent of atmospheric nitrogen wet deposition to coastal waters; contributes to eutrophication in surface waters

The 1970 CAA introduced significant regulation of NO_x emissions. Various programs and rules under the CAA regulate specific sources of NO_x air emissions (a more detailed discussion of programs affecting NO_x emissions is presented in Chapter III). Nationwide NO_x emissions continued to increase and reached a peak in 1978, but declined slightly in 1979 and 1980 as more and more CAA NO_x programs began to take effect. Since 1980, the effects of increasing economic activity and the effects of increasingly comprehensive regulation of NO_x have roughly balanced out, and NO_x emissions have been relatively constant.

Figure II-8 shows the trend in national NO_x emission rates from 1985 to 1996. As this figure indicates, nationwide NO_x emissions have fluctuated around 21 to 23 million metric tons per year throughout this period. Nationwide NO_x emissions in 1996 were 21.2 million metric tons (U.S. EPA 1997g). Figure II-9 presents 1996 NO_x emissions by State (U.S. EPA 1997f).

Figure II-10 presents relative NO_x emissions from principal anthropogenic sources in 1996 on a nationwide basis (U.S. EPA 1997f). The "all other" category includes miscellaneous minor sources such as chemical and allied product manufacturing, storage, and transport; metals processing; petroleum and related industrial processing; solvent utilization; other industrial processes; and, waste disposal and recycling.

Reduced nitrogen (ammonium) compounds are also released from anthropogenic sources at rates approaching those for NO_x in some areas. Relative contributions of various anthropogenic nitrogen sources to atmospheric nitrogen and nitrogen deposition vary from region to region depending on the predominant economic activities of each region. Both North Carolina and the Chesapeake Bay region, for example, have relatively high concentrations of animal farming operations, a key source of ammonium compound emissions. For example, Aneja et al. (1998) determined that anthropogenic emissions of ammonium compounds account for over 40 percent of anthropogenic nitrogen releases to air in North Carolina (Figure II-11). Similarly, Fisher and Oppenheimer (1991) found that ammonia and ammonium primarily released from crop and animal farming operations contributes 20 to 40 percent of total atmospheric deposition nitrogen (ADN) to the Chesapeake Bay. However, additional research is needed to better quantify ammonia and ammonium emissions to the environment. The EPA is collaborating with the U.S. Department of Agriculture (USDA) and academia to develop emission factors for various practices and to better model transport and deposition.

A recent monitoring study of wet deposition of nitrogen in the Chesapeake Bay used stable isotope analysis to identify sources. Results of the study indicate some seasonal trends. For example, in the spring, a peak in ammonium was observed as a result of agricultural emissions, including fertilizers, soil, animal excreta from the southwest and west regions of the bay. An observed peak in nitrate in the spring indicates greater soil emission rates during the spring; however, the dominant source of nitrate in the region is probably fossil fuel combustion primarily from the northwest and west regions of the bay. No seasonal trend was observed for dissolved organic nitrogen, but it is likely that it originates from sources similar to nitrate sources (Russell et al. 1998).

Once emitted to the atmosphere, nitrogen compounds (particularly NO_x) can travel great distances – 600 to 800 kilometers or farther from the point of emission to the point of deposition (Dennis 1997). The airsheds of the Great Waters for nitrogen compounds (defined as the geographic region from which atmospheric nitrogen deposited to the Great Waters originates) are, therefore, extensive and collectively encompass much of the U.S. as well as parts of neighboring countries. Using the Regional Atmospheric Deposition Model (RADM), Dennis (1997) determined that the NO_x airshed for the Chesapeake Bay watershed encompasses roughly 900,000 square kilometers, including all or part of 15 States and two Canadian provinces, extending into Indiana to the west, South Carolina to the south, and Quebec to the north. Using the same methodology, Dennis recently conducted a more refined analysis and revised the estimate of the Chesapeake Bay airshed upward to 1,081,600 square kilometers (see Figure IV-2). Thus, nitrogen emitted to the atmosphere several States away can be deposited to the watersheds of the Great Waters and to the waterbodies themselves.

Figure II-8
National NO_x Emission Trends, 1985-1996
(U.S. EPA 1997f)

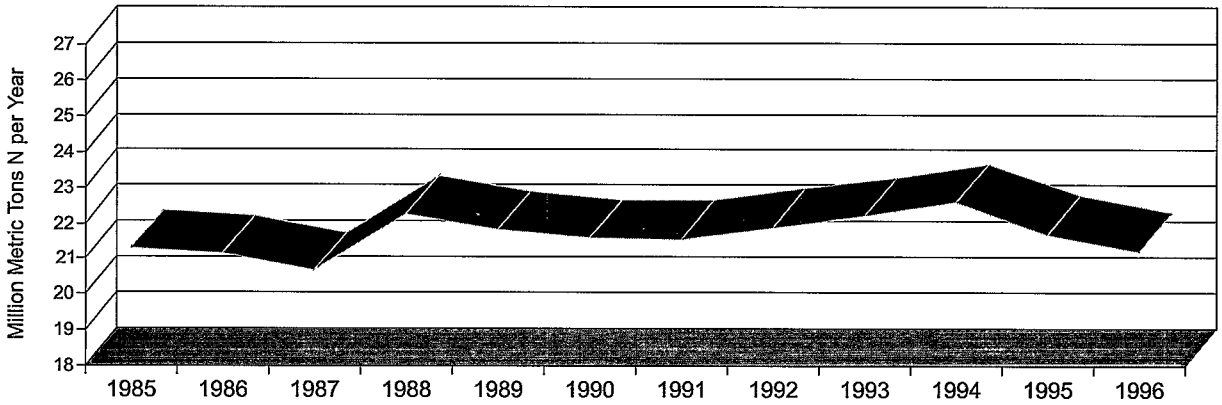


Figure II-10
1996 National Anthropogenic NO_x Emissions by Principal Source Category
(U.S. EPA 1997f)

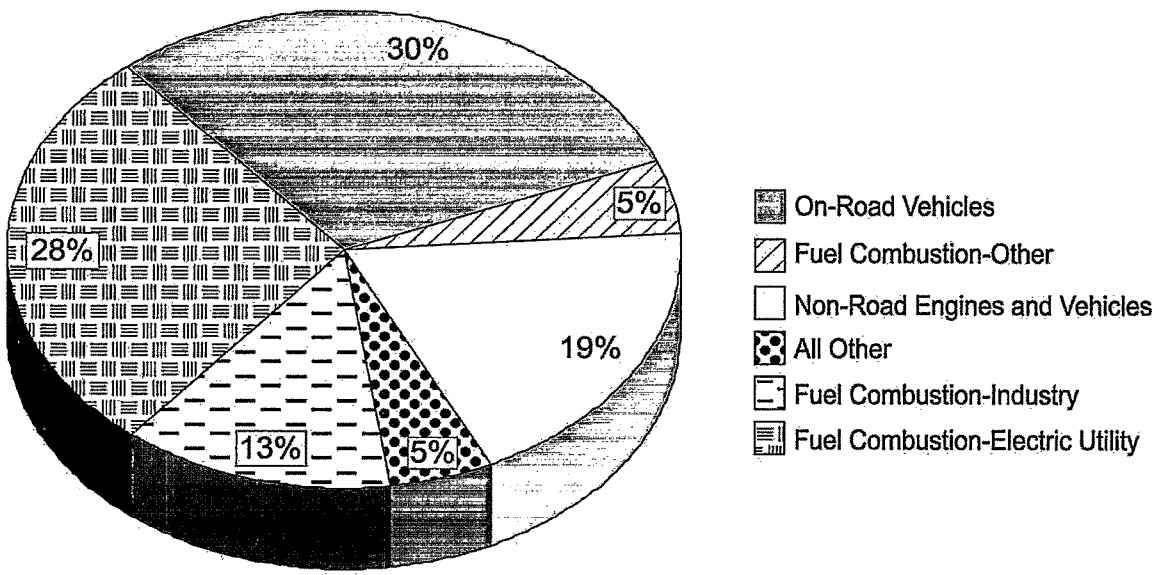
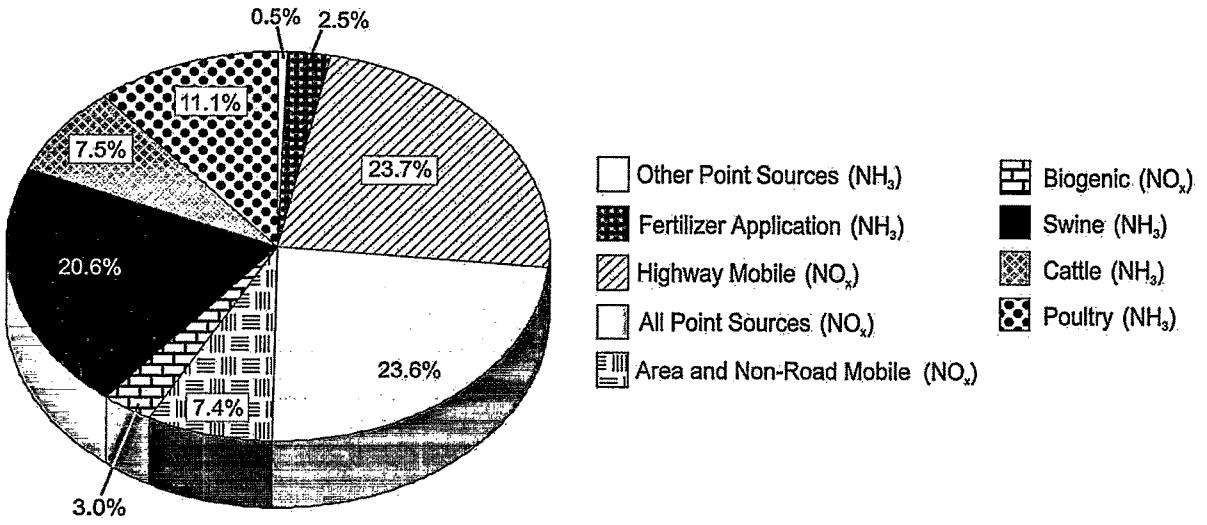


Figure II-9
1996 NO_x Emissions by State (Thousand Metric Tons/Rank)
(U.S. EPA 1997f)

Figure II-11
Estimated Nitrogen Emissions to Air by Principal Source Category in North Carolina – Anthropogenic and Biogenic (Aneja et al. 1998)



Nitrogen Deposition in the U.S.

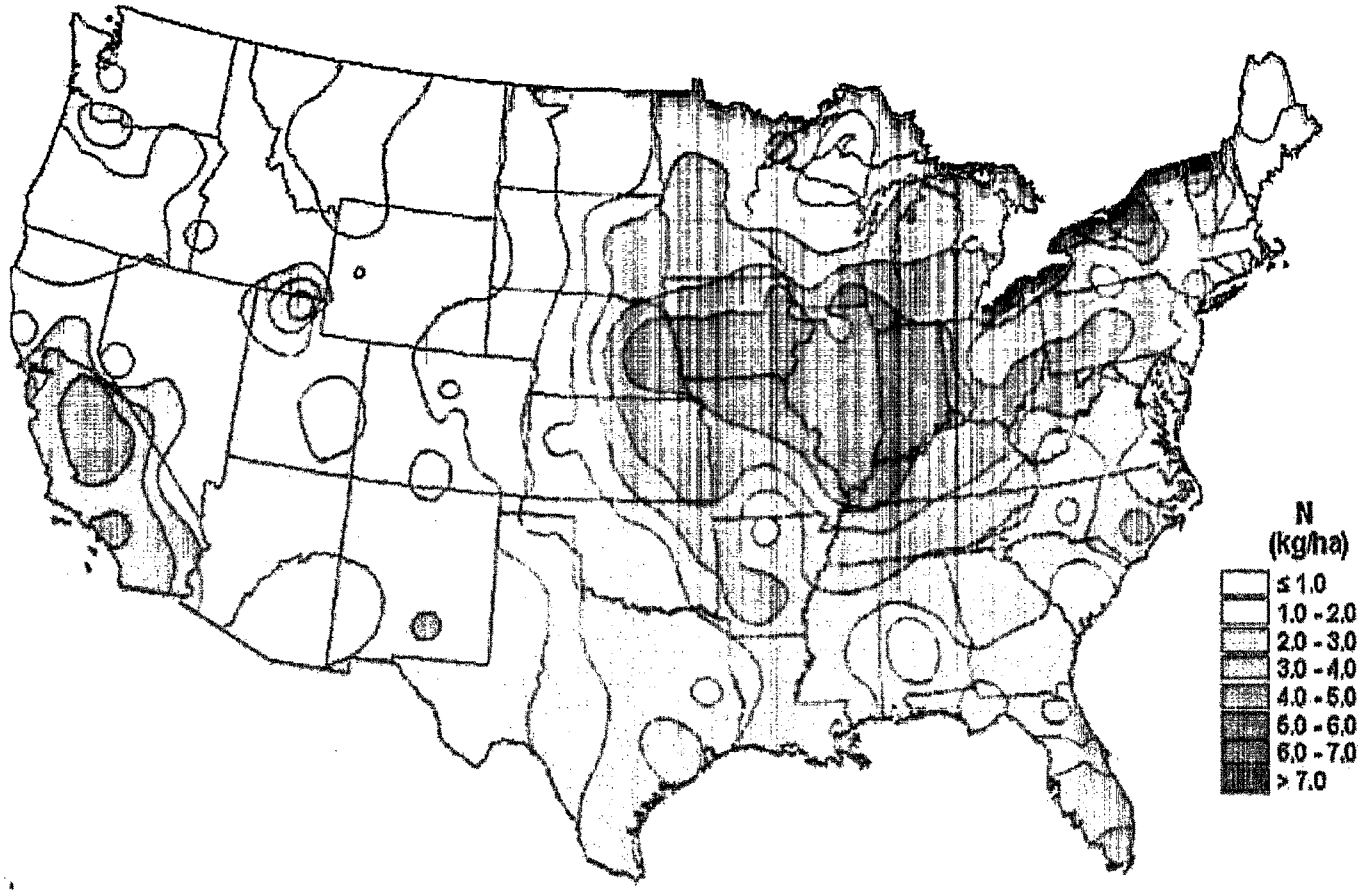
Nitrogen compounds are deposited to surface media by dry and wet deposition. Rates and trends of inorganic nitrogen wet deposition to the Great Waters can be evaluated based on data developed by the National Atmospheric Deposition Program (NADP) National Trends Network (NADP/NTN), which monitors wet deposition rates for ammonium (NH_4^+) and nitrate (NO_3) at approximately 200 monitoring stations across the country. The network provides weekly and annual deposition rates for these two nitrogen compounds (in terms of kg/ha as the ions) and for total inorganic nitrogen (in terms of kg/ha as N).[3]

The NADP/NTN has monitoring sites within the watersheds of most of the Great Waters, providing measurements of nitrogen wet deposition to at least portions of the watersheds. Data from monitoring points nearest the Great Waters can be extrapolated to estimate regional nitrogen wet deposition to the Great Waters and their watersheds. While this approach involves increasing uncertainty with increasing distance between monitoring sites and waterbodies, it nevertheless is currently the best available approach for evaluating the trends in inorganic nitrogen wet deposition to the Great Waters.

Figure II-12 presents rates of inorganic nitrogen wet deposition from the atmosphere as measured at the NADP monitoring sites nationwide for 1997. Data from individual monitoring points have been extrapolated in this figure to obtain isopleths for inorganic nitrogen wet deposition rates nationwide (including the Great Waters). These data indicate that inorganic nitrogen wet deposition ranged from lows of 0.1 kg/ha/yr in Alaska and 0.2 kg/ha/yr in Oregon to a high of 9.6 kg/ha/yr in upstate New York.

[3] Total inorganic N is calculated based on the sum of ammonium and nitrate nitrogen (Personal Communication with Van C. Bowersox, NADP Coordinator, 12/9/98).

Figure II-12
Estimated Inorganic Nitrogen Wet Deposition, 1997
(NADP/NTN 1998a)



Adapted from http://nadp.sws.uiuc.edu.

The NADP monitoring data suggest that the rates of inorganic nitrogen wet deposition to many of the Great Waters watersheds have been relatively constant for the past two decades. Figures II-13, II-14, and II-15 present the average rates of nitrogen wet deposition within the watersheds of three of the Great Waters, from 1980 through 1997.[3] Year-to-year patterns vary among watersheds, but all three watersheds exhibit a similar trend – relatively constant rates of nitrogen wet deposition over this period. Data from other sources suggest that deposition trends vary locally, however. Deposition of NH_x has increased since 1980 in areas downwind of intensive livestock operations (Paerl 1999), and annual atmospheric deposition of NO_3, NH_x, and total inorganic nitrogen measured at the NADP site nearest to Tampa and Sarasota Bays has increased significantly in the past decade. The increase in nitrogen atmospheric deposition near Tampa and Sarasota bays is correlated with increases in population in the region over the same time period (Dixon et al. 1996).

As with all dry deposition processes, nitrogen dry deposition is extremely difficult and expensive to monitor directly, and no programs currently do this. The lack of a reliable approach for quantifying dry deposition remains a significant gap in the understanding of nitrogen deposition processes and effects. Dry deposition can be inferred based on the nitrogen content of atmospheric gases and particles and computed deposition rates. Two networks are using this approach: NOAA's Atmospheric Integrated Research Monitoring Network (AIRMoN-dry) and the EPA-sponsored Clean Air Status and Trends Network (CASTNet). However, extrapolation of inferred nitrogen dry deposition values to surrounding areas is unreliable (Hicks 1998, Dennis 1999) and is still under investigation. In the absence of measured nitrogen dry deposition rates, many investigators have attempted to estimate dry deposition based on a ratio of dry deposition to wet deposition. A 1:1 ratio is most commonly derived (Hinga et al. 1991, Valiela et al. 1997). However, this approach also introduces considerable uncertainty, and the 1:1 ratio is applicable only for oxidized inorganic nitrogen, not ammonium compounds (Dennis 1999, Chimka et al. 1997).

Organic nitrogen is also deposited from the atmosphere to the ground and surface waters. However, measurement of dry organic nitrogen deposition is unreliable and, to date, only a limited number of measurements of organic nitrogen dry deposition rates have been made (Scudlark et al. 1998). Measurement of wet organic nitrogen is more reliable, and more and more data on wet deposition are becoming available. Based on a number of measurements of the organic nitrogen content in precipitation on the mid-Atlantic coast, Scudlark et al. (1998) found that organic nitrogen averages at least 20 percent of total dissolved nitrogen in precipitation, and can comprise as much as 64 percent of total dissolved nitrogen for single precipitation events. Based on measurements in North Carolina, Peierls and Paerl (1997) determined that roughly 30 percent of dissolved nitrogen in precipitation and rainwater deposition was organic nitrogen. Based on these results, it appears that organic nitrogen is a significant component of atmospheric deposition nitrogen (ADN) loads to surface waters, although the current understanding of the contribution of organic nitrogen to overall ADN is limited.

[3] NADP data were analyzed by selecting the monitoring sites within the watersheds of the Great Waters and averaging the reported deposition rates for all monitoring sites within each watershed to obtain an average deposition rate for the watershed. Data from 9 monitoring sites were averaged for the Chesapeake Bay watershed; data from 18 monitoring sites were averaged for the Great Lakes watersheds, and data from 4 monitoring sites were averaged for the Lake Champlain watershed.

Figure II-13
Nitrogen Wet Deposition in the Chesapeake Bay Watershed
(NADP/NTN 1998b)

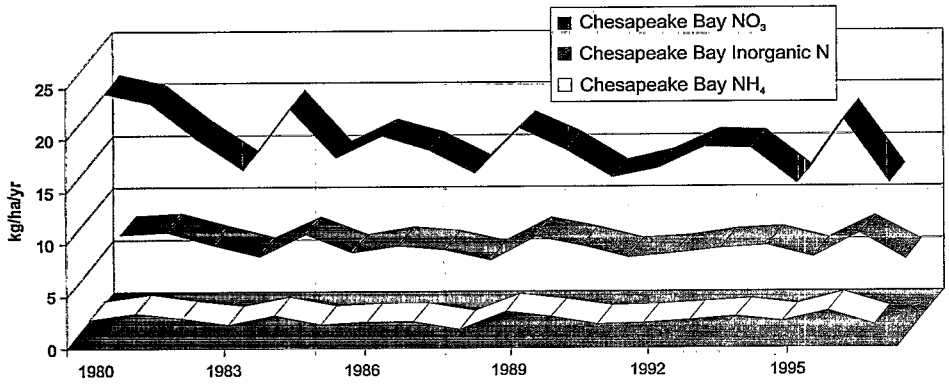


Figure II-14
Nitrogen Wet Deposition in the Great Lakes Watersheds
(NADP/NTN 1998b)

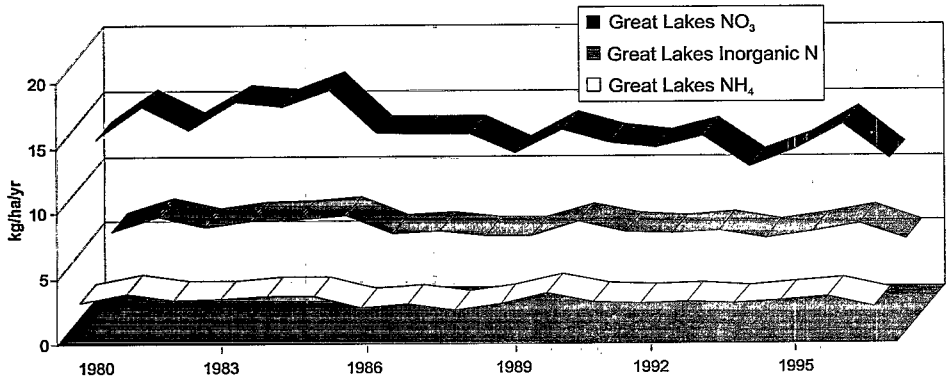
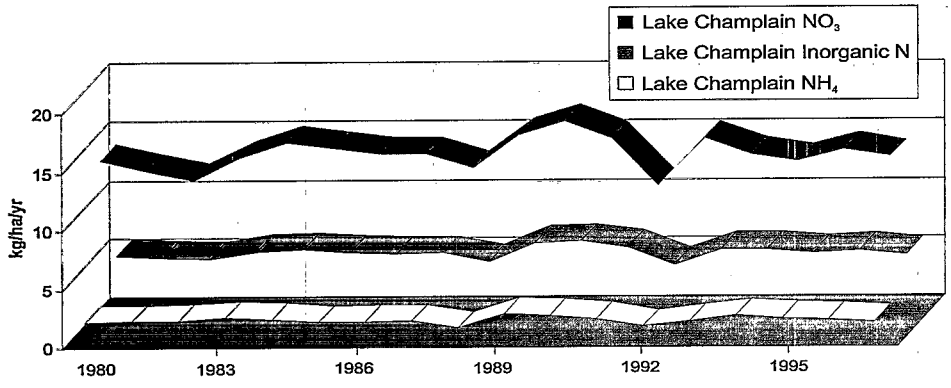


Figure II-15
Nitrogen Wet Deposition in the Lake Champlain Watershed
(NADP/NTN 1998b)



Note: Total inorganic N expressed as N; NO_3 and NH_4^+ expressed as the ions.

Atmospheric Nitrogen Loadings to the Great Waters

The total ADN load delivered to each of the Great Waters is a sum of the nitrogen deposited directly to the waters surface, plus a portion of the nitrogen deposited to the watershed. Although only a fraction of the atmospheric nitrogen deposited to watersheds ultimately reaches downstream Great Waters waterbodies, ADN transfer from watersheds is nevertheless the more important pathway for introduction of ADN to these waterbodies. This is due to the much larger areal extent of watersheds relative to the surface areas of the receiving waterbodies. The portion of ADN transferred from watersheds to downstream waterbodies is still relatively uncertain, and although several ongoing projects are attempting to quantify this, the rate of ADN transfer from watersheds to downstream waterbodies remains one of the greatest sources of uncertainty in estimating nitrogen and ADN loadings to surface waters. Transfer rates appear to vary greatly among watersheds and among areas within watersheds. For example, estimates of ADN transfer rates from various watershed areas to Great Bay Estuary, New Hampshire and to Waquoit Bay, Massachusetts are presented in Table II-19. Table II-20 presents estimated overall ADN transfer estimates for the watersheds of various coastal waterbodies (as developed by a number of researchers using a variety of techniques). The ADN transfer rates in Tables II-19 and II-20 were generally estimated based on estimated steady-state flux of nitrogen.

The ADN loads to surface waters join nitrogen loadings from a variety of other sources. Key sources of nitrogen reaching surface waters via routes other than the atmosphere (i.e., point source discharges, runoff, ground water) include (1) fertilizer from agricultural operations, recreation areas, suburban lawns; (2) manure from animal production facilities; (3) municipal and industrial treatment plant sludge and effluent and residential septic tanks; (4) crop residues (especially nitrogen-fixing crops such as legumes); and, (5) industrial wastes.

Table II-21 presents a number of estimates of the total nitrogen load, the nitrogen load attributable to ADN, and the portion of total nitrogen load represented by ADN for a number of Atlantic and Gulf coast Great Waters (developed using a variety of techniques). Based on these data, ADN contributes roughly 10 to 40 percent of total nitrogen loads reaching the bays and estuaries studied. If these values are assumed to be representative of the remaining Great Waters in the east and Gulf coast region, it is clear that ADN contributes very significantly to the total load of nitrogen reaching the Great Waters of these regions. It should be noted, however, that ADN accounts for roughly only 1 percent of nitrogen loadings in the Mississippi River basin, which has highly diverse sources of anthropogenic nitrogen (Goolsby et al. 1998, as cited by Paerl 1999). The ADN is also likely to be a relatively less important contributor to nitrogen loadings in Pacific coast Great Waters than is the case in the east and Gulf coast areas, due to the prevailing westerly winds over Pacific coastal waters and their watersheds, originating from unpolluted ocean areas.

Table II-19
Transfer of Atmospheric Deposition Nitrogen (ADN) from Various Watershed Areas

Great Bay Estuary, New Hampshire[a]		Waquoit Bay, Massachusetts[b]	
Type of Area Receiving ADN	Percent ADN Transferred to Estuary	Type of Area Receiving ADN	Percent ADN Transferred to Bay
Urban	38	Roads, runways, commercial areas	25
Active agriculture	10	Agricultural land (other than turf and cranberry)	10
Forest	5	Natural vegetation	9
Surface water in the watershed	90	Ponds in the watershed	44
Wetlands	3	Cranberry bogs	10
Disturbed/open land	6	Turf	10

[a] Talbot and Mosher 1998
[b] Valiela et al. 1997

Table II-20
Transfer of Atmospheric Deposition Nitrogen (ADN) from Watersheds to Several Bays and Estuaries

Watershed	Percent of ADN Transferred to Bay/Estuary	Reference
Albemarle-Pamlico Sounds	15	Paerl and Fogel 1994, as cited in Valigura et al. 1997
Chesapeake Bay	15	Linker 1998
Delaware Bay	9.4	Scudlark and Church, 1993, as cited in Valigura et al. 1997
Long Island Sound	15	Long Island Sound Study, as cited in Valigura et al. 1997
Narragansett Bay	7	Hinga et al. 1991, as cited in Valigura et al. 1997
New York Bight	12	Hinga et al. 1991, as cited in Valigura et al. 1997
Waquoit Bay	11	Valiela et al. 1997

Table II-21
Atmospheric Nitrogen Loads Relative to Total Nitrogen Loads in Selected Great Waters*

Waterbody	Total Nitrogen Load (million kg/yr)	Atmospheric Nitrogen Load (million kg/yr)	Percent Load From the Atmosphere
Albemarle-Pamlico Sounds[a]	23	9	38
Chesapeake Bay[b]	170	36	21
Delaware Bay[c]	54	8	15
Long Island Sound[d]	60	12	20
Narragansett Bay[e]	5	0.6	12
New York Bight[e]	164	62	38
Based on ADN loads from the watershed only (excluding direct nitrogen deposition to the bay surface):			
Waquoit Bay, MA[f]	.022	.0065	29
Based on atmospheric N deposition directly to the Waterbody (excluding ADN loads from the watershed):			
Delaware inland bays[g]	1.3	.28	21
Flanders Bay, NY[h]	.36	.027	7
Guadalupe Estuary, TX[i]	4.2 - 15.9	.31	2 - 8
Massachusetts Bays[j]	22 - 30	1.6 - 6	5 - 27
Narragansett Bay[k]	9	.4	4
Newport River Coastal Waters, NC[a]	.27 - .85	.095 - .68	>35
Potomac River MD[l]	35.5	1.9	5
Sarasota Bay, FL[m]	.6	.16	26
Tampa Bay, FL[n]	3.8	1.1	28

*As cited in Valigura et al. 1997: [a]Paerl and Fogel 1994, [b]Linker 1998, [c]Scudlark and Church 1993, [d]Long Island Sound Study 1996, [e]Hinga et al. 1991, [f]Valiela et al. 1996, [g]Delaware Bays NEP, [h]Peconic Bay NEP, [i]Brock et al. 1995, [j]Massachusetts Bays NEP 1996, [k]Nixon et al. 1995, [l]Boynton et al. 1995, [m]Sarasota Bay NEP 1995, [n]Tampa Bay NEP, Zarbock et al. 1994

Effects of Nitrogen Loadings on the Great Waters

While ADN contributes significantly to the overall adverse effects associated with excessive nitrogen loadings to the Great Waters, the effects of atmospherically-derived nitrogen on the Great Waters is very difficult to distinguish from the effects of nitrogen loadings from other sources. The effects of atmospheric nitrogen deposition to the Great Waters and associated trends can only be reviewed in terms of the effects of overall nitrogen loadings to these waters and the associated trends.

Although historical data for total aquatic nitrogen concentrations in the Great Waters are not available, nitrate concentrations have been monitored in many rivers and drinking water supplies in the U.S. for decades. Nitrate (NO_3) concentrations in surface water are directly related to anthropogenic nitrogen loadings to watersheds and surface waters (Vitousek et al. 1997). Aerosol nitrate is the most mobile form of nitrogen in watersheds and aquatic systems, and other forms of anthropogenic nitrogen, notably NH_x and organic nitrogen, are partly oxidized to NO_3 in the environment (Hessen et al. 1997). Surface water NO_3 concentration trends, although only part of the picture, provide some insight on the trends in anthropogenic nitrogen loadings and their effects on the Great Waters.

Available data indicate a significant increasing trend in the fluxes of nitrates in major surface waters in the U.S., including the Great Waters and their tributaries.

- Nitrate has more than doubled in the Mississippi River since 1965 (Vitousek et al. 1997). Note, however, that ADN accounts for roughly only 1 percent of nitrogen loadings in the Mississippi River basin (Goolsby et al. 1998, as cited by Paerl 1999).

- Nitrate fluxes in 10 major rivers feeding coastal waters in the northeastern U.S. have increased by factors of three to eight since 1900 (Jaworski et al. 1997). This increase is correlated with a three- to five-fold increase in NO_x emissions in the watersheds of these same rivers and more than a four-fold increase in atmospheric nitrogen deposition rates to northeastern coastal watersheds over the same time period (Vitousek et al. 1997).

- Increases in surface water nitrogen concentrations of 10 to 25 percent have been observed in Long Island Sound, portions of the Potomac River, and the Chesapeake Bay, the Neuse River, and portions of Biscayne Bay over the past 2 to 15 years (NOAA 1996, NOAA 1997a, as cited in U.S. EPA 1997f).

The long-term upward trend in nitrogen fluxes and nitrogen concentrations in many surface waters reflects similar upward trends in atmospheric emissions of nitrogen, in fertilizer use, and in municipal wastewater discharges in the initial seven decades of this century. Some evidence suggests, however, that nitrogen fluxes attributable to atmospheric deposition may have leveled off in more recent years in at least some waterbodies.

- Based on historical watershed data, Jaworski et al. (1997) found that nitrogen fluxes (total and attributable to ADN) for a number of northeastern U.S. coastal waters rose rapidly until about 1970, but have been more or less constant since then.

- Stoddard et al. (1998) found that the nitrate concentrations in higher elevation lakes and streams in the northeastern U.S. have either remained relatively constant or shown no discernable trend since the early 1980s. Atmospheric deposition nitrogen is the principal and, in some cases, the only source of anthropogenic nitrogen loadings to these lakes, which feed tributaries of the Great Waters.

Fluxes of ADN and total nitrogen to some of the Great Waters may, therefore, have leveled off in recent years. For many of the Great Waters, however, these fluxes remain many times greater than natural levels that occurred prior to human activity in the watersheds and airsheds (Vitousek et al. 1997). Human loadings of nitrogen to the land can overwhelm the capacities of watersheds to assimilate nitrogen, meaning that greater and greater percentages of the nitrogen deposited to watersheds are transferred directly to downstream waterbodies. Thus, constant rates of nitrogen deposition to watersheds can result in increasing nitrogen loadings to downstream waterbodies. Loadings of nitrogen to waterbodies can similarly overwhelm the capacity of waterbodies to assimilate or eliminate nitrogen, leading to a significant buildup of nitrogen concentrations and, in some cases, nitrogen saturation and related ecosystem degradation (Vitousek et al. 1997, Moffat 1998). These factors are believed to contribute to significant increases in concentrations of nitrogen in some of the Great Waters and rivers feeding the Great Waters, specifically the Mississippi River and a number of major rivers in the northeastern U.S. (Vitousek et al. 1997).

The "natural" or ecologically ideal concentration of nitrogen in the water column varies considerably among waterbodies. As one example, the Chesapeake Bay Program (1992, as cited in U.S. EPA 1998b) has determined that the most desirable concentration of dissolved inorganic nitrogen in the Chesapeake Bay (i.e., the concentration necessary to promote continued survival of submerged aquatic vegetation) is <0.15 mg/L.

Recent NOAA Estuarine Eutrophication Surveys (NOAA 1996, 1997a, b, c, as summarized in U.S. EPA 1998t) indicate elevated nitrogen concentrations in many of the Great Waters coastal estuaries on the Atlantic and Gulf coasts. These surveys found the following:

- High aquatic nitrogen concentrations (> 1 mg/L) in two of 18 estuaries on the U.S. north Atlantic coast, 14 of 22 mid-Atlantic estuaries, 11 of 21 south Atlantic estuaries, and 18 of 37 Gulf of Mexico estuaries; and,

- Moderate to moderately elevated aquatic nitrogen concentrations (0.1 - 0.9 mg/L) in 13 of 18 north Atlantic estuaries, five of 22 mid-Atlantic estuaries, seven of 21 south Atlantic estuaries, and 19 of 37 Gulf of Mexico estuaries.

Excess nitrogen loadings in the Great Waters can have a range of effects on the ecological structure and processes of these waters and, ultimately, on human use of the waters. Effects of excess nitrogen loadings can be considered in three broad categories (U.S. EPA 1997a).

1. Dramatic increases in plant productivity and algal blooms in systems where algal growth was previously nitrogen-limited. This can in turn cause fish kills, loss of submerged aquatic vegetation due to reduced sunlight penetration, loss of recreational use of waterbodies, and ultimately hypoxic (low oxygen) or anoxic (no oxygen) conditions as algal blooms die and decomposition processes consume the available oxygen – a condition referred to as eutrophication. Hypoxic and anoxic conditions can lead to loss of aquatic biodiversity as species requiring higher oxygen concentrations are lost, can affect the biogeochemical cycling of nutrients and toxic compounds, and can affect the bioavailability of toxic compounds. In addition to these consequences associated with normal algal blooms, excess nitrogen loadings are also linked to harmful or toxic algal blooms, often referred to as red or brown tides, involving algae species that are toxic to other aquatic biota or humans, often causing fish kills and human illness or mortality (Paerl et al. 1999, 1998, Paerl 1997a, Pelley 1998, Tibbets 1998, Rabalais et al. 1996).

2. Changes in the ecological balance brought on by fundamental changes at the base of the food web (i.e., the loss of aquatic grasses and other vegetation and the dominance of a relatively few algal species) and loss of biological diversity throughout the system as fundamental aquatic ecological parameters such as oxygen level, sunlight penetration, diversity and abundance of food species, and habitat availability change.

3. Direct toxicity of some nitrogen compounds to aquatic organisms. Toxicity of nitrogen compounds to aquatic organisms is not well understood, but mounting evidence suggests that these compounds can affect the ability of some species to adapt to other stresses (Linton et al. 1998). Dissolved ammonia at concentrations above 0.2 mg/L can be toxic to aquatic organisms (Paerl 1997a, as cited by U.S. EPA 1997a).

Many of these adverse effects have been observed by researchers in the Great Waters. Examples are provided below.

- McClelland and Valiela (1998) and Short and Burdick (1996) found that increased loads of anthropogenic nitrogen to Waquoit Bay, Massachusetts have a significant effect on the pathways by which nitrogen enters the food web. The diets of primary consumers in the bay are influenced by the dominant forms of food material available. Eelgrass is an important dietary component of primary consumers in areas of the bay receiving lower anthropogenic nitrogen loadings.

Laboratory studies by Short and Burdick (1996) indicated that excessive nitrogen loadings stimulated growth of algal competitors that shade and stress eelgrass. These laboratory results were borne out in field observations in Waquoit Bay, where domination by algal competitors was observed in areas of significant eelgrass decline. McClelland and Valiela (1998) found that eelgrass is lost and is largely replaced by algae in areas of the bay receiving higher anthropogenic nitrogen loadings. Thus, an important pathway for nitrogen entry to the food web may be altered. McClelland and Valiela conclude that this change in the nitrogen pathway probably affects the rate and manner in which nitrogen is cycled within the bay ecosystem and may reduce the stability of shallow estuarine ecosystems in the bay. Loss of eelgrass also results in loss of habitat for shellfish and other small fish, further affecting the ecological structure and function of the bay.

- Allen and Hershey (1996) demonstrated that the immediate effects of atmospheric deposition of nitrogen can vary seasonally for some waterbodies. In a north shore river tributary to Lake Superior, availability of nitrogen was found to alternate with availability of phosphorus and other factors as the limiting factor controlling algal biomass growth. Introduction of additional nitrogen resulted in increased algal biomass during early summer and late summer, but not during other seasons. Introduction of additional phosphorus caused increased algal growth in spring, but not in other seasons. Algal growth was not affected by additional nitrogen or additional phosphorus during the fall season, suggesting limitation by factors other than nitrogen or phosphorus availability. The dissolved inorganic nitrogen to soluble reactive phosphorus (DIN/SRP) ratio was not a reliable predictor of nutrient limitation in the studied river.

- Linton et al. (1998) found that the combination of global warming and increased ammonia loadings from atmospheric and other sources could have a greater impact on aquatic species in Lake Superior than either factor alone. These researchers studied rainbow trout reactions to elevated aquatic ammonia concentrations in conditions simulating Lake Ontario under a global warming scenario (2 °C above natural water fluctuating water temperature cycle). Their results indicate that elevated ammonia concentrations could reduce the ability of rainbow trout to adapt to long-term, small temperature increases such as could be caused by global warming.

- The disappearance of the economically important scallop from Tampa Bay waters is attributed to algal blooms and associated changes in ecological conditions in the bay, which in turn is attributed to excess nitrogen loadings (U.S. EPA 1997a).

- Hypoxia attributed to nitrogen-induced eutrophication is now a common and recurring condition in many bays along the Atlantic and Gulf coasts, notably Long Island Sound, New York-New Jersey Harbor, Chesapeake Bay, Albemarle-Pamlico Sound, and the Mississippi Delta region (U.S. EPA 1997a).

- Major algal blooms attributed to increases in aquatic nitrogen concentrations have occurred in several Atlantic and Gulf coast waters in recent years. These blooms have been accompanied by deleterious visual and odor effects and fish kills (U.S. EPA 1997a).

- The number of U.S. coastal areas experiencing major, recurring harmful or toxic algal blooms, linked to increases in aquatic nitrogen availability, have doubled since 1972 and include areas on the Atlantic, Gulf of Mexico, and Pacific coasts (Pelley 1998, Tibbetts 1998).

The NOAA Estuarine Eutrophication Surveys (NOAA 1996, 1997a, b, c, as summarized in U.S. EPA 1998t) indicate that the adverse effects of excess nitrogen concentrations are currently evident to

greater or lesser degrees in approximately 89 percent of the U.S. coastal estuary Great Waters. These surveys report the following:

- Anoxic or hypoxic conditions (oxygen content 0 - 2 mg/L) in portions of four of 18 north Atlantic estuaries, 13 of 22 mid-Atlantic estuaries, 13 of 21 south Atlantic estuaries, and 26 of 37 Gulf of Mexico estuaries; and,

- Biologically stressful levels of oxygen (2 - 5 mg/L) in at least portions of nine of 18 north Atlantic estuaries, 21 of 22 mid-Atlantic estuaries, seven of 21 south Atlantic estuaries, and 35 of 37 Gulf of Mexico estuaries.

Human Health Effects of Nitrogen in the Great Waters

Under the Safe Drinking Water Act, EPA set maximum contaminant levels (MCLs) for nitrate and nitrite in drinking water to protect human health. These levels are established at 10 mg/L of nitrate and 1 mg/L of nitrite. The primary adverse human health effect associated with exposure to nitrate or nitrite is methemoglobinemia, a dangerous condition in which nitrite in the blood stream prevents oxygen transport, potentially leading to brain damage or death. This condition is rare in adults, but is a significant danger for infants because microbes present in the stomachs of infants can convert large amounts of nitrate to nitrite. The National Research Council concluded, in 1995, that results from epidemiological studies are inadequate to support an association between nitrate or nitrite exposure from drinking water in the U.S. and increased cancer rates in humans. However, an epidemiological study led by a researcher from the National Research Council, published in 1996, of rural populations using community water supplies in Nebraska concluded that long-term exposure to elevated nitrate levels in drinking water may contribute to the risk for non-Hodgkin's Lymphoma. Research is continuing in this area (U.S. EPA 1997h, Ward et al. 1996).

Data indicate that most of the Great Lakes nearshore waters can be used as a source of drinking water with normal treatment, meeting the nitrate and nitrite levels (U.S. EPA 1998n). Similarly, Lake Champlain was not associated with any violations of standards in place for drinking water supplies due to Great Waters pollutants of concern from 1986 to 1995 (Lake Champlain Basin Program 1999). Further reductions in pollutant concentrations may reduce the cost of drinking water treatment in some areas.

Excess nutrients in coastal waters have also been implicated in the increased incidence of harmful or toxic algal blooms in the coastal waters of nearly every U.S. coastal State. These blooms have been linked to health effects among exposed human populations ranging from shortness of breath to dizziness, diarrhea, disorientation, permanent memory loss, and occasion death (Pelley 1998, Tibbetts 1998). To the extent that ADN contributes to elevated nitrogen content in coastal waters, ADN also contributes to human health risks associated with harmful and toxic algal blooms. No evidence exists to date suggesting negative human health effects attributable directly to atmospherically-derived nitrogen in the Great Waters.

BANNED AND RESTRICTED USE SUBSTANCES

Polychlorinated Biphenyls (PCBs)

HIGHLIGHTS
Polychlorinated Biphenyls

➤ **Sources.** In general, the number and magnitude of PCB sources have decreased 20-fold in the past 20 years. PCB sources generally are limited to atmospheric deposition and localized stationary sources. The largest national-level stationary air emission source of PCBs is hazardous waste incineration, along with several smaller sources. Sources to surface waters include atmospheric deposition, urban runoff, resuspension of contaminated sediments, and recycling through volatilization followed by deposition.

➤ **Loadings.** In the Great Lakes, studies show that deposition of PCBs has decreased and that PCBs in surface waters may be in equilibrium with the atmosphere (i.e., no net loadings). Deposition rates are higher near the Chicago urban area when compared to offshore and rural locations. In the Chesapeake Bay, a net loss of PCBs from the water surface has been observed.

➤ **Environmental Concentrations and Exposure.** PCB contamination levels in biota from the Great Lakes have declined; however, certain human subpopulations continue to be exposed to PCBs through fish consumption. In the Chesapeake Bay, maximum PCB sediment concentrations are below the PEL and NOEL for aquatic biota, with the exception of one location on the James River.

Manufacture of PCBs in the U.S. was voluntarily stopped in the late 1970s, and the use of PCBs was restricted under the Toxic Substances Control Act (TSCA) in 1979. Therefore, sources of PCBs in the U.S. are limited. However, PCBs are still present in older commercial and industrial equipment (e.g., electrical transformers and capacitors) and are released from non-point and non-regulated sources including Superfund sites. In addition, despite reductions in use, PCBs are released during some combustion processes, such as the incomplete burning of PCB-containing wastes during incineration. New research is also indicating that PCBs are actually being formed during certain combustion processes, similar to the way in which dioxins and furans are formed (Lemieux et al. 1999).

The number and magnitude of sources of PCBs have decreased 20-fold in the last 20 years, and sources of PCBs generally are limited to atmospheric deposition and localized point source discharges (Froese et al. 1997). Some studies detected PCBs in smallmouth bass from Fumee Lake, a remote lake in Michigan (Henry et al. 1998), and in the blubber of Beluga whales from Alaska's north coast (Wade et al. 1997a), indicating long range transport and atmospheric deposition as a major source in these areas. Many recent studies in the Great Waters, however, implicate non-atmospheric sources of PCBs (e.g., Brazner and DeVita 1998, Metcalfe et al. 1997a). The relative importance of atmospheric inputs and non-atmospheric sources remains unclear. Nevertheless, the largest stationary source of PCB air emissions on a national basis, as indicated by EPA's 1990-1993 NTI, is hazardous waste incineration at dedicated hazardous waste incinerators, along with several other smaller sources (Table II-22) (U.S. EPA 1999a). The EPA is pursuing additional research to determine the extent of PCB releases during incomplete combustion in these and other facilities as well as the effects on emissions and loadings of PCBs.

In Lake Superior, specifically, approximately 52 kg/year of PCBs are from point source inputs, 15 kg/year are from runoff (including pollutants in rainfall plus pollutants that are transported from land areas), and 85-156 kg/year are from wet and dry deposition (Hoff et al. 1996). Similarly, water samples collected during an investigation of the source of PCBs in the Detroit River (which feeds Lake Erie) indicate that the dominant sources of PCBs in this location are surface runoff from urban areas during rainfall and resuspension of in-place deposits of PCBs in the sediments of the river and bays due to high flow events in the rivers and due to severe wind conditions in the lakes (Froese et al. 1997).

Table II-22
National Anthropogenic PCB Air Emissions
(Based on EPA's 1990-1993 National Toxics Inventory)

Source Category	Anthropogenic Air Emissions (tons/year)	Percent Contribution to Total U.S. Anthropogenic Air Emissions
Hazardous waste incineration	0.03	56.0
Sewage sludge incineration	0.005	10.3
Fabricated metal products, nec	0.005	10.1
Industrial boilers: natural gas combustion	0.004	7.7
Portland cement manufacture: all fuels	0.004	7.3
Scrap and waste materials	0.001	2.7
Scrap tire combustion	0.001	2.1
Refuse systems	0.0006	1.2
Fabricated rubber products, nec	0.0005	1.0
Others (< 1 percent) [a]	0.0008	1.7
Total U.S. Anthropogenic PCB Air Emissions	0.052[b]	100[b]

[a] A list of the source categories that contribute less than 1 percent to total U.S. emissions is provided in Appendix B.
[b] This value represents anthropogenic PCB air emissions in the U.S. only.
Source: U.S. EPA 1999a

Research based on deposition monitoring under the Atmospheric Exchange Over Lakes and Ocean Surfaces (AEOLOS) project found that the "urban plume" of Chicago increases the deposition of PCBs to Lake Michigan and is a major source of atmospheric PCBs to coastal Lake Michigan. Total ambient air PCB concentrations on the lakeshore near Chicago are two to three times higher than background levels and the values collected under IADN, which monitors remote locations on the Great Lakes (Offenberg and Baker 1997). Franz et al. (1998) found similar results using dry deposition monitoring data from AEOLOS and the Lake Michigan Mass Balance Study where fluxes of particulate PCBs were higher in Chicago than less than 15 km offshore and at rural sites. The higher urban fluxes are due to increased anthropogenic activity and the greater atmospheric burden of large particles that are generated in the urban area and deposited closer to their sources. Franz et al. (1998) estimated dry deposition of PCBs to Lake Michigan to be approximately 1,100 kg/year, which is at least an order of magnitude greater than some previously reported values. Other studies report inputs of PCBs through air/water exchange at approximately 880 kg/year and wet deposition at 50 - 250 kg/year (Franz et al. 1998). In contrast, Offenberg and Baker (1997) found that the amount of yearly wet deposition plays a major role in determining the annual deposition of PCBs to Lake Michigan, and IADN monitoring results indicate that gas absorption is a dominant deposition process for delivering PCBs to lake surfaces (U.S./Canada IADN Scientific Steering Committee 1998).. The observed differences from these studies illustrate the importance of considering local meteorology and the location of monitoring stations (rural versus urban) when interpreting deposition data.

In addition to depositing PCBs released to the atmosphere from emission sources, atmospheric deposition processes play a strong role in PCB recycling in Lakes Superior, Michigan, and Huron, but

less of a role in Lakes Erie and Ontario. Volatilization of PCBs from the water, however, is the dominant mechanism for exchange of PCBs between the lakes and the atmosphere. According to IADN monitoring data from 1988 to 1996, wet and dry deposition of PCBs to each of the Great Lakes decreased (Hoff et al. 1996) (Table II-23). In the Detroit River, which is a heavily industrialized and urbanized area and is a primary transport route of PCBs to Lake Erie and Lake Ontario, the mass of PCBs transported through the Trenton Channel to the western basin of Lake Erie in 1995 was estimated at 600 kg/yr, constituting a potential decrease from 1986 estimates of approximately 1,500 kg/year. (Note, however, that different sampling methodologies were employed in the different years and may be responsible for a portion of the change (Froese et al. 1997)). Other monitoring studies show a net loss of PCBs to the atmosphere, particularly from Lake Superior and Lake Michigan (Offenberg and Baker 1997, U.S. EPA 1998m). For example, PCBs are being purged from Lake Superior at 21 percent per year of its annual burden (Hoff et al. 1996). Another monitoring data analysis indicates that gas transfer of PCBs out of the lakes is counteracted by net deposition, meaning PCBs are in equilibrium with the water surface and the atmosphere (Hillery et al. 1997).

Table II-23
IADN Loading Estimates of PCBs (wet and dry) for the Great Lakes (kg/year)

Year	Superior	Michigan	Huron	Erie	Ontario
1988	550	400	400	180	140
1992	160	110	110	53	42
1994	85	69	180	37	64
1996	50	42	N/A	34	N/A

Source: U.S. EPA 1998l

Studies using surface water and air sampling indicate that Chesapeake Bay waters have higher relative concentrations than the overlying atmosphere, and volatilization is the dominant loss process for PCBs from the bay. Polychlorinated biphenyls are volatilizing to the atmosphere at an average rate of 35 $\mu g/m^2/yr$, with a range of 31 to 112 $\mu g/m^2/yr$. Annual loss of PCBs from the bay from net volatilization (403 kg/yr) is 10 times greater than inputs from wet and dry deposition of 37 kg/yr and is more than two times greater than the loadings from the Susquehanna River of 165 kg/yr (Nelson et al. 1998). The Chesapeake Bay Program's *1999 Toxics Loading and Release Inventory* (TLRI) provides information on the total inputs and relative atmospheric inputs of PCBs to the tidal and non-tidal waters of the Chesapeake Bay based on monitoring data. Total PCB inputs to the Chesapeake Bay, without point source estimates, are similar for both inputs from the non-tidal watershed (254 kg/year) and inputs from wet and dry atmospheric deposition to the tidal waters (245 kg/year) (Chesapeake Bay Program 1999a).

The comparison of current contamination levels to historical values provides evidence that concentrations of PCBs in biota have declined in the Great Lakes. For example, Dykstra et al. (1998) measured PCB concentrations in nestling blood (1989-1994) and addled eggs (1986-1993) of bald eagles in Lake Superior and inland Wisconsin sites. A comparison to published and unpublished historical data of PCBs in addled bald eagle eggs on Lake Superior indicates that PCB levels declined significantly between 1969 and 1993; however, PCBs may still be at levels high enough to affect reproductive success. Similarly, Ion et al. (1997) measured the concentration of PCBs in yellow perch from the St. Lawrence River between 1975 and 1991-1992. Comparison of average PCB concentrations detected to historical levels in St. Lawrence River fish indicates that levels have decreased by a factor of 30 since 1975. The concentration of PCBs in all fish sampled by Ion et al. (1997) were below the International Joint

Commission guideline of 100 ng/g in whole fish, which is protective of aquatic life and consumers of fish.

A study by Eby et al. (1997) shed doubt on findings reported in the *Second Great Waters Report to Congress* that concluded that PCB concentrations in Great Lakes have leveled off or even increased slightly in the last 10 years (U.S. EPA 1997b). In those earlier studies, scientists hypothesized that the trend in PCB contamination could be due to changes in the composition of the food web. Eby et al. (1997) demonstrate, however, that sampling methodology may have influenced the apparent PCB concentration trends rather than changes in fish diet. As part of a long-term PCB monitoring survey, the USGS collects and analyzes bloaters (*Coregonus hoyi*) from the same site in Lake Michigan every year. Because fish size is used to predict fish age, similar size fish are collected each year. However, since the early 1980s, bloater populations experienced a decline in growth rate due to a shift in diet, which placed older, more contaminated bloaters in the size range sampled. This resulted in an apparent trend of steady state or increasing PCB levels. In contrast, Eby et al. (1997) modeled an increase in the amount of the most contaminated prey in the bloater diet and found that this shift in diet had little effect on PCB contamination levels in bloaters.

A recent study by Haffner et al. (1997) found that total PCB concentrations in herring gull eggs decreased significantly between 1981 and 1992 in Lake Ontario and Lake Erie, although the decrease in lake Erie was not of the same magnitude. The PCB concentrations detected during this study are not expected to affect reproduction in herring gull populations. In addition, Haffner et al. (1997) found that total PCB concentrations in double-crested cormorant eggs did not decrease in Lake Ontario but decreased slightly in Lake Erie between 1981 and 1992. The PCB concentrations detected in this study may be high enough to cause toxicological stress on the cormorant population (e.g., egg mortality, infertility, deformity).

Although some studies indicate that PCB levels in fish have decreased or reached a steady state in recent years, consumption of contaminated fish from the Great Lakes remains a concern. Several recent studies examined the relationship between increased health risks in certain subpopulations and the consumption of fish from the Great Lakes contaminated with PCBs, concluding that certain subpopulations continue to be exposed to PCBs through fish consumption despite declines in contamination levels and despite continued fish consumption advisories. Study results include the following.

- Pellettieri et al. (1996) determined that the estimated PCB intake for sport fisherman in the Chicago area exceeded the level recommended by the Great Lakes Advisory Task Force of no more than one meal of fish per week (assuming the average meal of fish is composed of 227 g of uncooked fish and that the intake of PCBs is reduced by half due to skinning, trimming, and cooking).

- A study of Ojibwa tribal members from the Great Lakes region indicated that PCB concentrations in Ojibwa populations were low with a maximum value of 9.6 ppb in blood serum; however, a significant association was found between serum PCB concentration and age (Gerstenberger et al. 1997). This is important because PCBs are persistent and bioaccumulate in the food chain.

- According to a 1997 survey (Tilden et al. 1997) of Great Lakes States' residents, the most widely accepted fish consumption advisory recommendation was cleaning and cooking methods. A study of six cooking methods determined that choice of cooking method greatly impacted the loss of PCBs during the cooking process, with smoking and microwave baking as the most

effective methods. Smoking reduced PCB levels by 65 percent, and microwave baking reduced PCB levels by 60 percent (Salama et al. 1998).

In a sediment contamination study of the Chesapeake Bay and its tributaries from 1984 to 1991 by Eskin et al. (1996) under the Chesapeake Bay Program, PCBs were detected at low levels in sediments throughout the bay and its tributaries, indicating that aquatic life impacts (based on the aquatic biota NOEL and PEL) from PCBs in the bay are not widespread. The highest concentrations were detected in sediments in the northern bay near the Baltimore area and on the James River. Maximum PCB concentrations were below aquatic biota NOEL and PEL concentrations in sediments at all locations, with the exception of one location on the James River.

Despite observed declines in some organisms, a study of PCB levels (as serum concentrations) in male and female juvenile American alligators in Lake Apopka, Florida indicated that 23 of 28 PCB congeners were present in juvenile alligators. The mean total PCB concentration in the serum was 1.54 ± 0.12 (SE) ng/mL. Alligators in Lake Apopka have been reported to have a number reproductive and endocrine system abnormalities which may be due to embryonic exposure to these and other compounds (Guillette et al. 1999).

Pesticides

Table I-1 presents the uses and sources of Great Waters pesticides and indicates that many of these pesticides are banned or use restricted; however, many of these pesticides may still be used in other countries. Therefore, long-range transport of emissions from other countries may contribute to current levels in Great Waters along with other current sources, such as use of existing stocks of pesticides, and releases from contaminated sites (e.g., Superfund sites) and from soils upon which pesticides were applied in the past. Because most of these pesticides are persistent and bioaccumulative, pesticides that were used or unintentionally released previously can cycle between environmental compartments (e.g., air, water, soil, sediment, biota). Therefore, some of these pesticides can be re-released to the atmosphere, transported short or long distances, and redeposited to surface waters.

HIGHLIGHTS
Pesticides

➤ **Sources.** Because most of the Great Waters pesticides of concern are banned or restricted use substances, the major sources include long range transport from other countries, use of existing pesticide stocks, and releases from contaminated sites and soils.

➤ **Loadings.** Since many of these pesticides are persistent and bioaccumulative, they are cycling between environmental compartments. In general, deposition of these substances remained constant or declined in recent years.

➤ **Environmental Concentrations.** DDT and its metabolites have, in general, declined to levels below which adverse environmental effects of concern are expected. In Chesapeake Bay sediments, chlordane and dieldrin concentrations were between the level expected to show widespread effects and the level expected to show limited effects.

Given these possible sources of banned and restricted use pesticides, efforts to reduce releases focus primarily on promoting (1) international agreements to reduce or eliminate the manufacture and use of these pesticides, and (2) pesticide collection programs (i.e., Clean Sweeps as described on page III-56). Additional future reductions may result from the clean up of existing stockpiles and contaminated sites. Research continues at EPA and other institutions on soil remediation methods that will reduce emissions of these pollutants from contaminated sites. In addition, efforts to limit the exposure of human and ecological populations to these pesticides include EPA and other agency monitoring programs and continued outreach activities to make the public aware of contaminated fish and waterbodies. Some

recent studies of these pesticides focus on the role of atmospheric transport, both historic and current, in the contamination of waterbodies and biota of the Great Waters.

Atmospheric transport and deposition of these pesticides remains a principal source in the Great Waters, as shown by studies of both regional and global transport. For most pesticides, however, the relative contribution of atmospheric deposition versus other sources (e.g., runoff from contaminated soils, groundwater contamination) to total loadings is not yet known. It is expected that with the development of mass balance models and better sources and emissions estimates, it will be possible to determine relative contributions of different sources of pesticides and other pollutants to the Great Waters waterbodies. Furthermore, the contamination of biota in remote waterbodies that receive no point source inputs emphasizes the role of atmospheric transport and deposition of pesticides. For example, DDT/DDE, toxaphene, chlordane, dieldrin, and hexachlorobenzene were detected in smallmouth bass from Fumee Lake, which is in the Upper Peninsula of Michigan. The remote location of Fumee Lake suggests atmospheric loading is the source of pesticide contamination. Concentrations of these pesticides (except toxaphene which is associated with elevated levels in Lake Superior because of point source inputs) were similar to those found in fish from Lake Superior, where atmospheric inputs are thought to be the primary source of contaminants. All pesticide concentrations were less than those reported in Lake Michigan, which typically receives contaminants by direct input from point sources as well as atmospheric deposition (Henry et al. 1998). The detection of toxaphene, DDT/DDE, and chlordane in the blubber of Beluga whales from Alaska's north coast (Wade et al. 1997a) demonstrates the global scale of atmospheric transport. Toxaphene, chlordane, lindane, α-HCH, hexachlorobenzene, dieldrin, and DDE also were detected in a variety of organisms at multiple trophic levels in Lake Superior (Kucklick and Baker 1998).

Banning and restricting uses of these pesticides under the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA) in the past 25 years have proven to be the most effective methods of removing these compounds from the environment, as would be expected. Because these substances are no longer intentionally released in the U.S. and the goal was to remove them from the environment, a group of researchers investigated how long these pesticides would persist in the atmosphere above the Great Lakes. Cortes et al. (1998) calculated atmospheric half-lives (i.e., the amount of time required for the concentration in the air to decrease by 50 percent) and used measured atmospheric concentrations to estimate minimum virtual elimination dates (i.e., when the contaminant levels are below the detection limits of the measurement equipment) in the Great Lakes regional atmosphere for DDT, DDE, dieldrin, chlordane, lindane (γ-HCH), and hexachlorobenzene (HCB) (Figure II-16). Based on these dates, DDT and DDE will be the first compounds to be reduced to concentrations below current detection limits in the Great Lakes atmosphere, followed by dieldrin and chlordane between 2010 and 2030, γ-HCH by 2030, and HCB by 2060. These estimates assume current rates of long-range transport of these pollutants into the region. Because of their persistence, it should be noted that elimination of these pollutants in the atmosphere does not mean that concentrations would be eliminated in deposited media by these dates. However, these estimates indicate that reduction strategies in the Great Lakes, along with the original bans or restrictions on the use of these substances, are having the intended effect.

DDT/DDD/DDE. In general, several studies indicate declines in atmospheric deposition of DDT and its metabolites as well as declines in concentrations in different environmental media. According to deposition monitoring data collected under IADN, wet and dry deposition of DDT to Lakes Superior, Michigan, and Erie in 1996 was less than values from 1994, 1992, and 1988 (Table II-24) (U.S. EPA 1998l). The IADN data from 1993-1994 indicate that the net flux of total DDT is into Lake Superior and Lake Erie, possibly due to the high levels of wet deposition during 1993-1994. In Lake Huron, there is a net loss of total DDT to the atmosphere, and total DDT is in equilibrium between the surface water and the atmosphere in Lake Ontario. These data indicate a reversal from net volatilization of total DDT from

Figure II-16
Expected Year that Great Waters Pesticides will be Below Current Detection Limits in the Great Lakes Atmosphere
(Cortes et al. 1998)

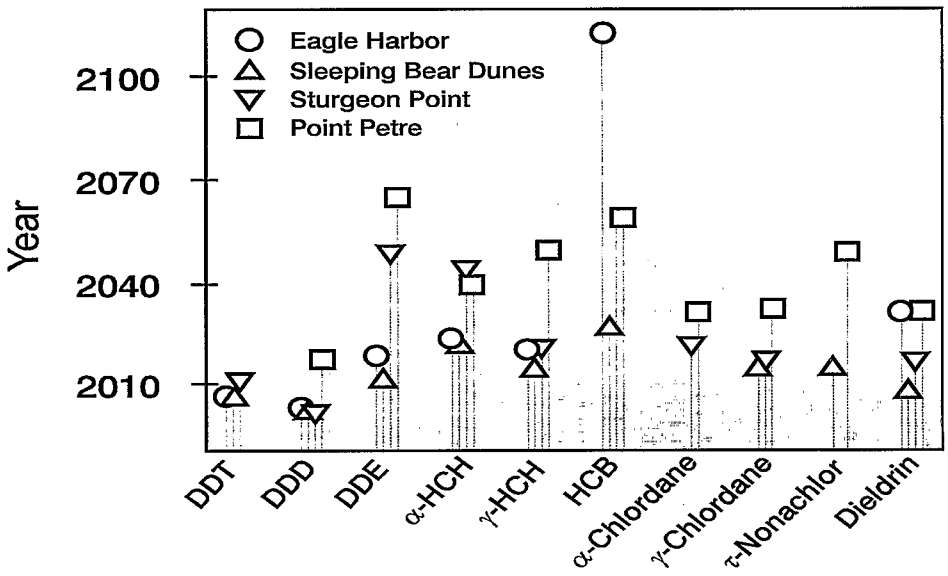


Table II-24
IADN Loading Estimates of DDT (wet and dry) to the Great Lakes (kg/year)

Year	Superior	Michigan	Huron	Erie	Ontario
1988	90	64	65	33	26
1992	34	25	25	12	10
1994	17	32	37	46	16
1996	4	12	N/A	2	N/A

Source: U.S. EPA 1998l

some of the lakes to net deposition to those lakes (U.S./Canada IADN Scientific Steering Committee 1998).

Current data indicate that elevated air concentrations of DDT and its metabolites in the South Haven, Michigan area are due to volatilization from the soil where the pesticide was placed during historical applications (Michigan Department of Environmental Quality 1998). The Great Lakes National Program Office (GLNPO; see page III-35) is funding a study to evaluate the source of the DDT and its metabolites in the air near a large fruit-growing area in South Haven where, historically, a number of researchers detected elevated levels of DDT and its metabolites. These levels are about 20 times greater than concentrations detected elsewhere in the Great Lakes basin. Recent studies that reported on DDT and DDE levels at a Canadian IADN site in Egbert, Ontario found that while increased levels in Egbert were not as pronounced as those in southwest Michigan (i.e., less than half), back trajectory analysis indicated that air parcels with elevated DDT and DDE levels usually had passed through South

Haven, Michigan. South Haven is located in an area which is primarily devoted to fruit and vegetable production where the historic application of DDT could have been extensive. However, another potential source of DDT and its metabolites could be the current use of dicofol (1,1-*bis*(4'-chlorophenyl)2,2,2-trichloroethanol). It has been postulated that agricultural practices and air-soil diffusion affect DDT volatilization rates from soil. Therefore, a study by the Michigan Department of Environmental Quality is under way to determine impacts of tillage practices on ambient DDT levels in southwest Michigan and to assess any possible long-range transport effects (Michigan Department of Environmental Quality 1998).

Brazner and DeVita (1998) detected DDE in yellow perch and spottail shiners from 23 sites in Green Bay, Lake Michigan. Because the overall distribution of DDE tissue concentrations was fairly uniform within the bay, DDE contamination probably originates primarily from non-point sources, including atmospheric deposition. The few sites that exhibited unusually high DDE residues in fish may have point sources of DDE in nearby rivers.

1,1'-(dichloroethenylidene)bis(4-chlorobenzene), or DDE, has been detected in a variety of organisms at multiple trophic levels in Lake Superior (Kucklick and Baker 1998). For example, Dykstra et al. (1998) measured DDE concentrations in nestling blood (1989-1994) and addled eggs (1986-1993) of bald eagles in Lake Superior and inland Wisconsin sites. A comparison to published and unpublished historical data indicated that DDE levels declined significantly between 1969 and 1993, with current levels below those known to result in reproductive impairment.

In a study of juvenile American alligators in Lake Apopka, Florida, p,p'-DDE was detected in female serum at 17.98 ± 5.3 (SEM) ng/mL and in male serum at 7.35 ± 2.4 (SEM) ng/mL. Serum concentrations were significantly higher in females than males. Both male and female alligators in this area have been associated with abnormal reproductive and endocrine functions (Guillette et al. 1999).

In a sediment contamination study of the Chesapeake Bay and its tributaries from 1984 to 1991 by Eskin et al. (1996) under the Chesapeake Bay Program, concentrations of DDT and its metabolites tended to decline from the head to the mouth of the mainstem bay. Highest concentrations (0.0016 ppm DDT and 0.0023 ppm DDE) were detected in sediments in the upper half of the bay and near the mouths of the York and Potomac Rivers. Elsewhere in the lower half of the bay, concentrations were generally below detection limits. All DDT sediment concentrations were below the aquatic biota PEL of 0.270 ppm and the aquatic biota NOEL of 0.0045 ppm. DDE sediment concentrations were below the aquatic biota PEL of 0.130 ppm at all locations, but maximum concentrations exceeded the DDE NOEL of 0.0017 ppm for aquatic biota in some locations in the upper bay. In the tributaries, concentrations were generally below the PELs for aquatic biota.

Dieldrin. Recent monitoring data from IADN indicate that dieldrin is being transferred out of the Great Lakes with volatilization from Lake Superior, Lake Erie, and Lake Ontario at 5.9, 6.7, and 6.6 times the loadings in 1994 (Hoff et al. 1996). In general, concentrations of dieldrin in precipitation decreased from 1986 to 1994, but concentrations were three to four times higher at the Lake Erie site than at the other sites due to higher usage rates in the surrounding agricultural areas. Higher wet deposition levels were also observed over Lake Michigan due to the greater degree of urbanization surrounding that lake (U.S. EPA 1998l). In Chesapeake Bay sediments between 1984 and 1991, dieldrin was detected at one site at levels between the aquatic biota PEL and NOEL (Eskin et al. 1996).

Chlordane. Chlordane, which was restricted in 1973 to use as a termiticide and was banned in 1988, was used primarily as a pesticide for corn crops (Aigner et al. 1998). Gas phase concentrations of chlordane in the air over Lake Superior do not currently exhibit a decreasing trend, as has been seen with

several other banned pesticides. In Chesapeake Bay sediments between 1984 and 1991, maximum chlordane concentrations were detected at levels between the aquatic biota PEL and NOEL (Eskin et al. 1996).

Toxaphene. In general, historical toxaphene concentration profiles in sediment cores across the Great Lakes indicate that the onset of contamination was between 1940 and 1950. Maximum levels occurred in the early 1970s to early 1980s, and accumulation rates in Lake Superior and Lake Michigan largely declined from historical peaks, as shown by comparing maximum historical rates to present rates (Table II-25). In addition, Lake Ontario and Lake Superior may have received toxaphene from non-atmospheric sources in the past (Pearson et al. 1997b). A back trajectory analysis using IADN monitoring data indicates that the Great Lakes receive toxaphene via atmospheric deposition from sources outside of the Great Lakes basin (U.S./Canada IADN Scientific Steering Committee 1998). Pearson et al. (1997b) investigated the history of toxaphene concentrations and accumulation in dated sediment cores from Lakes Superior, Ontario, and Michigan and two control lakes near Lake Superior that receive toxaphene only via atmospheric deposition. Concentrations at the surface of the cores, which represent current contamination levels, were similar among all samples with the exception of two samples from northern Lake Michigan. The similar toxaphene concentrations among the Great Lakes cores and control lakes suggest atmospheric input is the dominant source of toxaphene to the Great Lakes. The higher concentrations in the surface of the two northern Lake Michigan cores indicates that this area may be receiving current inputs (as great as 30-50 percent) from non-atmospheric sources.

Table II-25
Current and Maximum Rates of Toxaphene Accumulation in Great Lakes Sediments [a]

Location	Number of Sediment Cores Analyzed	Accumulation Rates in Sediment ($ng/cm^2/yr$)	
		Present	Maximum Historical
Lake Superior	3	0.097 - 0.14	0.25
Northern Lake Michigan	3	0.52 - 1.01	1.07
Southern Lake Michigan near Chicago Urban Area	1	0.24	0.32
Lake Ontario	3	0.39 - 0.60	1.4

[a] All accumulations are focus-corrected.
Source: Pearson et al. 1997b

Hexachlorocyclohexanes. The α-HCH and γ-HCH isomers are Great Waters pollutants of concern, whereas β-HCH (i.e., another of the eight HCH isomers) is not. Both α-HCH and γ-HCH can be transformed into β-HCH, which is a more toxic and bioaccumulative HCH isomer. Monitoring data indicate that air concentrations of HCH are decreasing, but the atmosphere continues to be a large source of HCHs to the Great Lakes. Since the 1980s, α-HCH and lindane (γ-HCH) precipitation concentrations declined; however, between 1991 and 1994, lindane concentrations increased at Lakes Superior and Ontario, possibly simply due to data variability. Although dry deposition inputs of HCH are relatively small (e.g., ranging from 0.2 to 1.5 kg/yr in all but Lake Ontario), the 1994 estimates of dry deposition are three to 10 times higher than 1992 estimates. Wet deposition, which is generally uniform across the lakes, ranges from 46 to 170 kg/yr and is between 0.25 and 1.2 times 1992 estimates, indicating a general downward trend in wet deposition. A gradient of α-HCH exists, with higher loadings in the lower lakes than in Lakes Superior and Michigan. Lindane has the highest wet deposition inputs over Lake Huron, possibly due to a source in upper Michigan (U.S. EPA 1998l).

The net gas flux of HCH ranges from -140 to +1200 kg/yr (where positive values indicate volatilization out of the lake) based on monitoring studies. Both Lake Michigan and Lake Ontario are saturated with α-HCH (Hoff et al. 1996). For example, the mass of α-HCH lost through air-water gas exchange in Lake Ontario is similar to major input and loss factors such as wet deposition and advection (i.e., transport of the pollutant out of the lake) (Ridal et al. 1997). The IADN monitoring data also indicate that total HCHs are in equilibrium with the surface water and the atmosphere in the Great Lakes (Hillery et al. 1997).

Hexachlorobenzene. In general, levels of atmospheric deposition of HCB has remained constant over the past few years. Cohen et al. (1995) used the HYSPLIT/TRANSCO computer program to determine the amount of HCB emitted from sources to air and water media that is deposited in the five Great Lakes. The model was run using 1993 source and emissions data from 1,329 identified sources in the U.S. and Canada. Table II-26 provides modeled air deposition estimates, average depositional flux, and water effluent inputs for the five Great Lakes. The results indicate the following.

- The depositional flux of HCB to the lakes increases from Lake Superior, Lake Michigan, Lake Huron, Lake Erie, to Lake Ontario, following the trend in industrialization around the lakes.

- Hexachlorobenzene inputs from air deposition are greater than water discharge inputs for Lake Superior, Lake Michigan, and Lake Huron. In Lake Ontario and Lake Erie, the opposite is true; however, the estimates for air deposition and waterborne inputs are uncertain for HCB because sources outside the U.S. and Canada were not considered in the model and because estimates represent the amount of HCB that would be deposited if the lakes were uncontaminated with HCB (Cohen et al. 1995).

Table II-26
Modeled Air Deposition, Depositional Flux, and Waterborne Inputs of HCB to the Great Lakes

HCB	Superior	Michigan	Huron	Erie	Ontario	Total
Total Deposition (kg/yr)	11	15	16	15	23	79
(range)	(4-49)	(5-73)	(6-74)	(6-65)	(9-101)	(30-362)
Depositional Flux (g/km^2/yr)	0.13	0.26	0.27	0.58	1.19	0.32
Waterborne Inputs (kg/yr)	0.1	0.8	0.6	<72	35	<108.5

Source: Cohen et al. 1995

In general, based on IADN monitoring data, wet and dry deposition of hexachlorobenzene (HCB) to the Great Lakes did not change from 1992 to 1994. In Lake Ontario, however, wet deposition of HCB increased six times in 1994 over the 1992 estimate. Hexachlorobenzene is in near equilibrium with the atmosphere and surface water, but is slightly loading Lake Superior and Lake Michigan and slightly volatilizing from Lake Erie and Lake Ontario (Hoff et al. 1996).

CROSS-POLLUTANT TRENDS

The EPA and other groups have initiated programs and projects to collect and compile information to assess current sources, emissions, loadings, exposure, and effects of pollutants in the Great Waters. Because many of these programs and projects address multiple pollutants of concern and

present cross-pollutant results, it is more appropriate to present the pollutant-specific information together with the program background rather than to split the information between pollutant categories. Therefore, background information on these various programs or projects along with information on their results is presented below by program or project. Note that most of these programs and projects are designed to assess contamination levels in certain media or specific waterbodies. While each program or project discussed below addresses a Great Waters waterbody and pollutants of concern, these programs generally do not directly assess atmospheric deposition. Instead, they address general pollution from many sources, one of which may be the atmosphere. Other programs and initiatives that portray how EPA and other agencies are working together to address the remaining issues related to atmospheric deposition of pollutants to the Great Waters are discussed in Chapter III.

FISH CONSUMPTION ADVISORIES

Fish consumption advisories are means of limiting human exposure when fish taken from a particular waterbody contain levels of pollutants that exceed recommended intake levels. National and regional trends in fish advisories can be estimated using EPA's database which contains information such as date of advisory, waterbody name and location, pollutant, fish or other wildlife species and range, advisory type, advisory status (e.g., active), and a contact name and telephone number (U.S. EPA 1998k). Note that different sampling methodologies and different approaches among States and years makes comparisons among States and years difficult. In addition, the amount of monitoring can vary widely from State to State.

Types of Fish Advisories

States, including the four U.S. Territories and tribal governments, issue advisories for waterbodies in an effort to reduce health risks associated with exposure to pollutants in certain freshwater fish and shellfish species:

- *Advisories to limit consumption* – advisories to either the general population or subpopulations potentially at greater risk (e.g., children, pregnant or nursing women) to restrict their consumption of specific species of fish and other wildlife.
- *Commercial fishing bans* – advisories that prohibit the commercial harvest and sale of fish, shellfish, and/or wildlife species from a designated water body (U.S. EPA 1998k).

In 1997, the number of waterbodies under advisory represented 16.5 percent of the Nation's total lake acreage and 8.2 percent of the Nation's river miles. In addition, 100 percent of the Great Lakes surface waters and their connecting waters, and a large portion of the Nation's coastal waters, are under advisory for one or more Great Waters pollutants of concern. According to the Listing of Fish and Wildlife Advisories in December 1997, 39 of 56 Great Waters were associated with fish advisories for pollutants of concern (see Appendix C for details on fish advisories in the Great Waters). The number of advisories in effect for U.S. waterbodies in 1997 increased 5 percent over the number reported in 1996 and 80 percent over the number reported in 1993 (U.S. EPA 1998k). This increase does not necessarily reflect actual increases of these pollutants in the environment. Rather, the increase is attributed to an increase in the number of assessments of contamination levels in fish and wildlife tissues as well as the use of more rigorous EPA risk assessment procedures by some States to set advisories.

Although advisories in the U.S. have been issued for a total of 46 pollutants, five Great Waters pollutants – mercury, PCBs, chlordane, dioxins, and DDT – comprised 95 percent of the advisories in 1997 (U.S. EPA 1998k). The number of advisories in the U.S. increased for three of these major pollutants and decreased for one, specifically:

- Mercury advisories increased 6 percent (1,677 to 1,782) from 1996 to 1997 and 98 percent from 1993 to 1997 (899 to 1,782). This increase is primarily because of new mercury advisories in 11 States, particularly in Minnesota, Ohio, and Louisiana.

- Advisories for DDT and dioxin rose slightly from 1996 to 1997. Advisories for DDT rose 3 percent (32 to 33), and dioxin advisories rose 8 percent (60 to 65), after a decline in the number of dioxin advisories in 1996. Dioxin advisories were rescinded in some States, in part, because many pulp and paper mills changed their processes. However, in Michigan, according to the 1999 Michigan Fish Advisory from the Michigan Department of Community Health, dioxin fish advisories were expanded to include Lake Huron, Lake Michigan, and Lake Superior.

- The number of PCB advisories rose 84 percent from 1993 to 1997. In the last year of this period, however, advisories decreased 5 percent (617 to 588), due primarily to rescinding and readjusting some previously issued advisories. Despite this recent decline, 30 new advisories were issued in 1997 -- mostly by Ohio, Maine, Alabama, Connecticut, Minnesota, Nebraska, and New York – making it difficult to discern trends from these advisories alone.

WATER QUALITY ASSESSMENTS

Surface waters and ground waters throughout the U.S. are monitored regularly to ensure that they are meeting the water quality standards appropriate to the intended uses. Pollutant concentration levels are measured to ensure that freshwater sources, including the Great Lakes and Lake Champlain, that are intended to be used for drinking water do not exceed the maximum contaminant level (MCL) guidelines for drinking water sources. In other surface waters, including all Great Waters, monitoring is conducted to ensure that ambient water quality criteria and other relevant guidelines are not exceeded and that the waterbodies meet the designated uses (e.g., fishing, swimming, boating). The current status of the Great Waters in terms of water quality criteria is described below.

Drinking Water

Of the Great Waters, only the Great Lakes and Lake Champlain are fresh waters that can be used as drinking water sources. Data indicate that both of these water systems provide safe drinking water, particularly as related to the Great Waters pollutants of concern. Based on data from 1994 and 1995 collected in support of section 305(b) of the Clean Water Act (CWA) by States, tribes, and other jurisdictions, most of the Great Lakes nearshore waters can be used as a source of drinking water with normal treatment (U.S. EPA 1998n). Similarly, the Lake Champlain Basin Program reported that from 1986 to 1995, only nine violations of EPA drinking water standards were reported – all of which were for coliform contamination in Vermont (Lake Champlain Basin Program 1999).

Other Water Uses

Data on water quality conditions of the Nation's waters, including the Great Lakes in particular, are collected and reported by States, tribes, and other jurisdictions to EPA every 2 years as required under section 305(b) of the CWA. These assessments are based on water quality data that are compared to water quality standards, such as designated beneficial uses, numeric and narrative criteria for supporting each use, and an antidegradation statement to protect existing uses and to protect waterbodies from deteriorating in quality. Data from 1994 and 1995 indicate that approximately 40 percent of the Nation's surveyed rivers, lakes, and estuaries are too polluted to support basic uses, such as fishing and swimming. Nutrients and metals are the most widespread pollutants impacting lakes and rivers with

agriculture as the most common sources of pollutants; however, atmospheric deposition was also identified as a source of pollutants. In estuaries, nutrients are the most widespread pollutants with industrial point sources, urban runoff, and storm sewers as the most common sources of pollutants (U.S. EPA 1998n).

Most of the Great Lakes, in contrast, are safe for swimming and other recreational activities. However, approximately 97 percent of the surveyed shoreline area shows unfavorable conditions for supporting aquatic life and is impacted by toxic organic chemicals that appear in fish tissue samples at much higher concentrations than in water samples. These impacts are partially due to persistent toxic pollutant burdens, such as PCBs, in the food web. Several of the Great Lakes States identified air pollution, discontinued discharges from industrial facilities that are no longer in use, urban runoff and storm sewers, contaminated sediments, land disposal of wastes, and unspecified non-point sources as pollutant sources contributing to water quality problems (U.S. EPA 1998n).

NOAA'S NATIONAL STATUS AND TRENDS PROGRAM

Since 1984, NOAA's National Status and Trends (NS&T) Program has conducted annual monitoring of chemical contamination and biological responses to contamination to document current conditions and long-term trends in the environmental quality of U.S. near shore waters and sediments. The Mussel Watch Project is a major component of this monitoring program. Initiated in 1986, the Mussel Watch Project is designed to provide long-term trends and large-scale monitoring of pollutant distributions, with an emphasis on temporal rather than spatial trends. The project tracks trends in pollutant concentrations in mollusks at 287 coastal and estuarine sites on the Atlantic, Pacific, and Gulf coasts (including Great Waters). Eighty-three of these sites were recently added, including several sites from the Great Lakes region. Mussels and oysters are useful for monitoring changes in pollutant levels because they are sedentary organisms and concentrations in their tissues generally reflect changes in the concentrations in the surrounding water. Trends were examined in the Mussel Watch Project for the following Great Waters pollutants of concern: DDT, chlordane, dieldrin, PCBs, PAHs, cadmium, lead, and mercury (O'Connor 1999).

Florida NS&T Sites

A recent monitoring study suggests general decreasing trends in contaminant levels in South Florida (Cantillo et al. 1997), based on concentrations of PAH, PCB, DDT, lindane, hexachlorobenzene, chlordane, lead, mercury, and cadmium in bivalves from seven South Florida sites monitored by the NS&T Program during the period 1986 to 1994. Contaminant levels in South Florida were generally lower than nationwide. Nevertheless, a recent sediment quality assessment indicates that resident benthic communities may be adversely impacted by contaminants at a number of locations in Tampa Bay (Carr et al. 1996). For instance, up to 73 percent of the sediment pore water samples were toxic to sediment biota in at least one of the toxicity tests. In addition, sediment concentrations of several metals and PAHs were correlated with sediment toxicity, and concentrations of lead, PAHs, PCBs, and DDT equaled or exceeded sediment quality guidelines in several locations.

Also as part of the Mussel Watch and Benthic Surveillance Projects, NOAA conducts sediment toxicity assessment studies to determine the extent and severity of sediment toxicity in U.S. coastal regions. Sediment toxicity studies are conducted based on the following considerations: (1) a high level of contamination in oysters or mussels sampled in the NS&T Program; (2) the likelihood of adverse biological effects of contamination based on State and local data; and (3) possible collaboration with other Federal, State, and local agencies. Since 1991 when NOAA began conducting these studies, 23 coastal bays and estuaries covering approximately 4,000 km^2 have been the subject of studies (see text box). Study sites range in size from 0.3 km^2 to 1,350 km^2. Sediment toxicity surveys include three different toxicity tests which measure different biological endpoints leading to different results: (1) the

amphipod survival test using whole sediment; (2) the sea urchin fertilization test using sediment porewater; and (3) the Microtox test which measures decreased light production by a luminescent marine bacterium, *Vibrio fischeri*, using an organic extract of the sediment. The concentration of approximately 80 chemical contaminants, including mercury, lead, cadmium, PAHs, chlorinated dioxins and dibenzofurans, DDT and its metabolites, dieldrin, chlordane, lindane, HCH, and HCB, are measured in sampled sediments. Results of these studies are discussed below (NOAA 1999).

National Trends

Data collected between 1986 and 1996 from the Mussel Watch Project indicate that concentrations of most of the Great Waters pollutants of concerns in mussel and oyster tissues are decreasing at some sites. Specifically, decreasing trends of DDT, chlordane, dieldrin, PCBs, and cadmium were evident. Concentrations of cadmium declined despite the fact that other trace metals, including mercury and lead, exhibited no overall trend during this period. Data for PAHs also indicated no overall trend. Lindane and hexachlorobenzene were so infrequently detected that no trend determinations could be made. Another study found a decreasing trend for cadmium and lead, and no overall trend for PAHs (Lauenstein and Daskalakis 1999, O'Connor 1999, NOAA 1998).

The NOAA analyzed spatial trends in sediment contamination based on data collected for the Benthic Surveillance Project and the Mussel Watch Project (NOAA 1998), and found that both trace metal and organic contamination are associated with urban areas. Sites with "high" levels of contamination were located in urbanized areas of the Northeast, San Diego, Los Angeles, and Seattle.[4] The cut-off levels that indicated "high" concentrations for DDT, PCBs, chlordane, dieldrin, PAHs, mercury, cadmium, and lead are 140 ppb, 430 ppb, 34, ppb, 9.1 ppb, 1100 ppb, 0.23 ppm, 6.2 ppm, and 4.8 ppm (mussels) and 0.84 ppm (oysters), respectively. "High" concentrations were rare in the southeast and along the Gulf of Mexico. Most sites with "high" contaminant concentrations can be linked to anthropogenic sources. Tissue concentration data can be misleading, however, because in some areas natural background levels and characteristics of chemicals can result in higher tissue concentrations. For example, high concentrations of cadmium and other trace metals in zebra mussels of the Great Lakes may be due to greater bioavailability of those metals in fresh water (NOAA 1998).

The sediment toxicity surveys, on a national level, indicate that 7 percent of the sediment in all of the study area was toxic based on the amphipod survival test; 39 percent was toxic based on the sea urchin fertilization test; and 66 percent was toxic based on the Microtox luminescence test. In general, larger estuaries and bays (areas greater than or equal to 250 km^2 or 100 mi^2) had a lower average spatial extent of sediment toxicity (i.e., 6 percent) as compared to smaller estuaries and bays (i.e., 10 percent). However, sediment toxicity varies considerably among study areas in terms of spatial extent and severity of toxicity. Furthermore, based on amphipod survival tests, NOAA found that the spatial extent of toxicity was about 35 percent in small urbanized estuaries in the Northeast and southern California and was only 3 percent in southeastern estuaries, which border coastlines, farms, forests, wetlands, and fewer industrialized areas (NOAA 1999).

[4] Areas with "high" concentrations were identified based on concentration distributions. "High" concentrations were defined as those with logarithmic values greater than the mean logarithmic value for all concentrations plus one standard deviation. Therefore, about 15 percent of all concentrations were "high."

Trends in Great Waters

Pollutant trends based on mussel and oyster tissue data collected for the Mussel Watch Project from 1986 to 1996 at several Great Waters sites are shown in Table II-27 (NOAA 1998). Similar trends were also reported in the *Second Report to Congress* (U.S. EPA 1997b). Decreasing trends are statistically significant (confidence level of 95 percent); however, quantitative information regarding the changes in pollutant levels was not provided. Therefore, the magnitudes of the increases or decreases cannot be compared between sites.

Table II-27
Trends of Pollutant Concentrations in Mussel Watch Project (1986-1996)

Waterbodies (number of sites examined)	Contaminant Trend (number of sites affected) [a]							
	Mercury	Lead	Cadmium	PCB	DDT	PAH	Chlordane	Dieldrin
Chesapeake Bay (6 sites)	↔ (6)	↔ (5) ⇓ (1)	↔ (5) ⇓ (1)	⇓ (4) ↔ (2)	↔ (3) ⇓ (3)	↔ (6)	⇓ (5) ↔ (1)	↔ (3) ⇓ (3)
Delaware Bay (6 sites)	↔ (6)	↔ (6)	↔ (5) ⇓ (1)	↔ (5) ⇓ (1)	↔ (6)	↔ (6)	↔ (4) ⇓ (2)	↔ (5) ⇓ (1)
Long Island Sound (9 sites)	↔ (9)	↔ (8) ⇓ (1)	⇓ (5) ↔ (4)	⇓ (5) ↔ (4)	↔ (5) ⇓ (4)	↔ (9)	⇓ (6) ↔ (3)	↔ (9)
Narragansett Bay (3 sites)	↔ (3)	↔ (2) ⇓ (1)	↔ (3)	↔ (3)	↔ (3)	↔ (3)	↔ (3)	↔ (3)
Tampa Bay (6 sites)	↔ (6)	↔ (5) ⇑ (1)	↔ (6)	↔ (6)	↔ (4) ⇓ (2)	↔ (6)	↔ (3) ⇓ (3)	↔ (6)
Galveston Bay (6 sites)	↔ (6)	↔ (6)	↔ (6)	↔ (6)	↔ (6)	↔ (6)	↔ (4) ⇓ (2)	↔ (4) ⇓ (2)

[a] Sites were sampled in at least 6 of 10 years; trend indicated by arrow (i.e., ⇓ = decreasing trend; ⇑ = increasing trend; ↔ = no trend). Number of sites showing trend within each waterbody is indicated in parentheses.
Source: NOAA 1998

In Tampa Bay, specifically, NS&T chemical contamination data for sediments, bivalves, and fish through 1991 indicate that there is not a bay-wide pattern of increasing or decreasing concentrations over recent years. However, increases and decreases were observed in some sample locations. Local geographic contamination trends indicate that sediment toxicity is highest in regions of northern Hillsborough Bay and moderate in regions of western Old Tampa Bay, along the western shore of Middle Tampa Bay, and in lower Boca Ciega Bay. The least toxic or nontoxic samples were taken from portions of Old Tampa Bay, and Middle and Lower Tampa Bay. Relatively high concentrations of petroleum hydrocarbons, chlorinated pesticides, other chlorinated hydrocarbons, ammonia, and trace metals were detected in the most toxic samples (Long and Greening 1999).

In the Chesapeake Bay, NS&T Program data from 186 sites from 1986 through 1995 generally indicate no trend in contamination levels in mollusks for each chemical analyzed. However, when trends were detected, decreases greatly outnumbered increases. Specifically, contamination levels decreased for chlordane, DDT, and dieldrin (Cantillo et al. 1998).

Consistent with trends on a national level, most of the Great Waters sites do not exhibit a statistically significant trend or change in pollutant levels in mollusk tissues during the 10-year period. Trends, when present, were mainly decreasing since 1986 (i.e., for cadmium, PCBs, DDT, PAHs,

chlordane, and dieldrin). According to O'Connor (1999) and NOAA (1998), decreases in levels of these chemicals are probably the result of bans on the use of chlorinated hydrocarbons (e.g., chlordane, DDT, PCBs) and the reduced use of certain chemicals.

NATIONAL SEDIMENT QUALITY SURVEY

The National Sediment Quality Survey (NSQS) describes the accumulation of chemical contaminants in the Nation's rivers, lakes, estuaries, and coastal waters. The NSQS is the first of three volumes of the first biennial *Report to Congress on the Incidence and Severity of Sediment Contamination in the United States* (U.S. EPA 1997i). The NSQS includes a screening-level assessment comparing human and ecological health benchmarks (e.g., draft sediment quality criteria, EPA cancer and noncancer human health risk levels) to concentrations of contaminants in sediment or, for human health only, to actual or estimated fish tissue concentrations. Results indicate the following.

- About 5 percent (i.e., 96) of the Nation's watersheds include APCs (Figure II-17, Appendix D includes the names of the numbered sites). The watersheds of the Great Lakes, Chesapeake Bay, and at least 17 NEP and NERRS sites are associated with APCs. An additional 39 percent of watersheds include at least one sampling station where contamination levels suggest probable adverse effects. Among that 39 percent are the watersheds of the Great Lakes, Chesapeake Bay, Lake Champlain, and some coastal waters. Only about 6 percent of watersheds did not indicate potential adverse effects on aquatic life or human health. Although there were no contaminant data for 35 percent of the watersheds, the data used were from monitoring programs targeted to areas with known or suspected contamination.

- Probable aquatic life impacts were observed 41 percent more frequently than probable human health impacts. Specifically, of the 21,096 sites sampled, 3,287 had probable aquatic life impacts and 2,327 had probable human health impacts.

- The contaminants detected most frequently at levels indicating probable adverse effects include PCBs, mercury, pesticides, and PAHs. Polychlorinated biphenyls were the sole or among the responsible indicators at 58 percent of the most contaminated (i.e., probable effects) sampling stations. Non-mercury metals were solely responsible for about 6 percent of the most contaminated sampling stations and 28 percent of the moderately contaminated (i.e., possible effects) sampling stations.

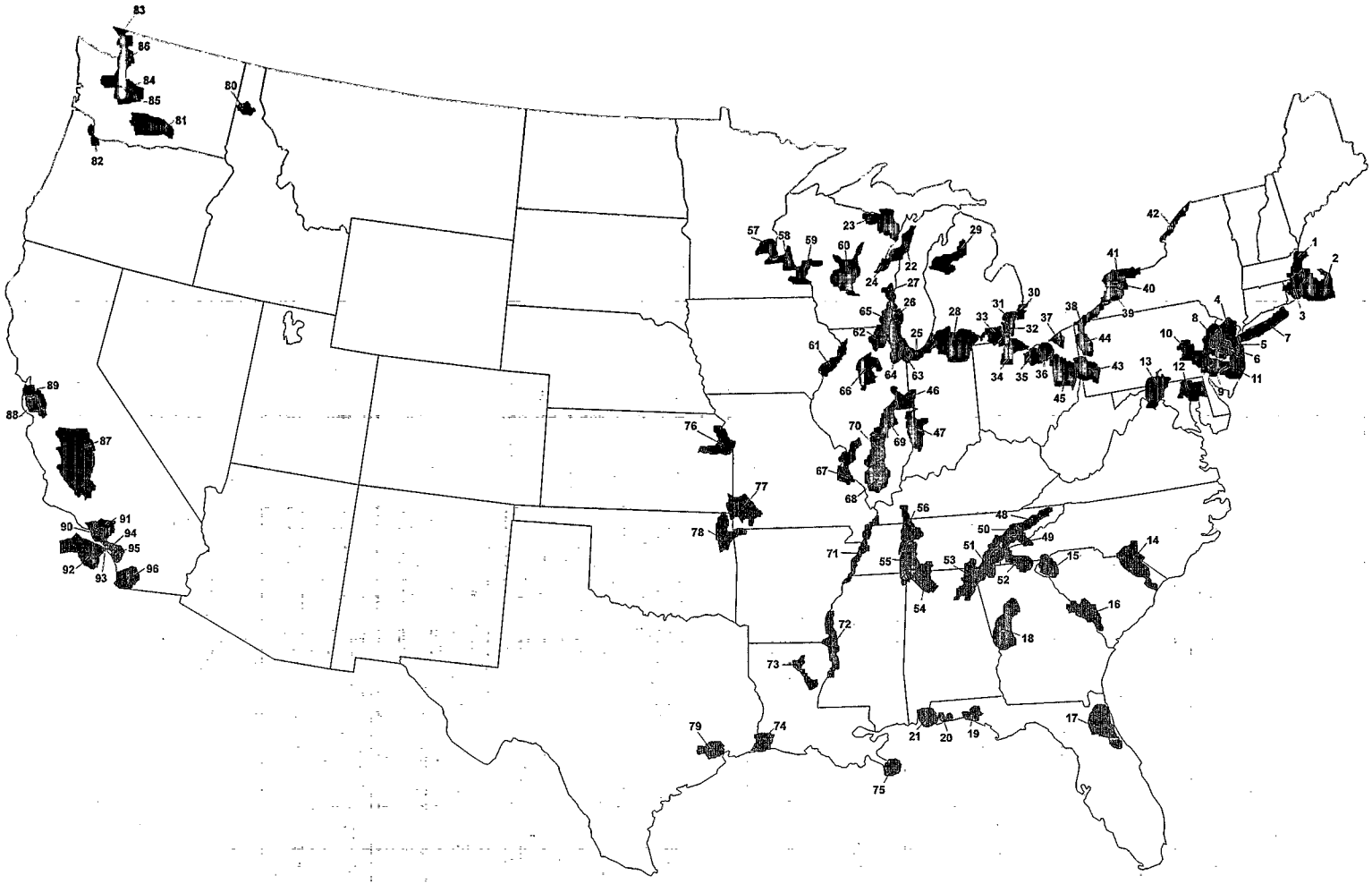
CHESAPEAKE BAY PROGRAM

As part of the Chesapeake Bay Program and in support of the goals and commitments of the Chesapeake Bay Agreement, information has been collected to assess the progress that has been made in reaching those goals.

- A recent analysis of 10 years of monitoring data indicates that the water quality of major rivers and freshwater portions of the bay has improved, probably due to reduced runoff from agricultural lands, the phosphate detergent ban, and point source reductions in phosphorous and nitrogen releases (e.g., at wastewater treatment facilities) (Chesapeake Bay Program 1998d).

- The amount and extent of submerged aquatic vegetation (SAV) is a good indicator of water quality in the Chesapeake Bay. Although the areal coverage of bay grasses, or SAV, increased by 9 percent to 69,238 acres in 1997, in 1998 the acreage had decreased by 8 percent to 63,496

Figure II-17
Locations of Areas of Probable Concern Within Watersheds



See Appendix D for names of numbered sites.
Source: U.S. EPA 1997i

acres. The reasons for the 1998 decrease are unclear, but higher-than-average flow conditions in 1998 are a likely factor. Even so, the total acreage of SAV in 1998 still represents a 70 percent increase over the 1984 low point and 56 percent of the goal to restore 114,000 acres of SAV by 2005 (Chesapeake Bay Program 1999b).

- National Toxics Release Inventory (TRI) data from 1988 to 1997 indicate a 67 percent reduction in chemical releases and transfers from industrial facilities to the bay. Releases of eight of the Chesapeake Bay Program Toxics of Concern (several of which are Great Waters program pollutants of concern), however, increased sharply in 1995, mostly due to the activities of a few facilities that were responsible for almost half of the releases (Chesapeake Bay Program 1998c).

- From 1985 to 1997, total nitrogen concentrations decreased in the Susquehanna River, which provides over 50 percent of the fresh water to the bay. Nitrogen concentrations decreased in six of the major rivers to the bay (i.e., Susquehanna, Patuxent, Rappahannock, Mattaponi, James, Appomattox) and remained unchanged in two other rivers (i.e., Potomac, Pamunkey (a tributary to theYork)). In contrast, total nitrogen concentrations increased in the upper central portion of the bay (i.e., the region around the Patapsco and Chester rivers) (Chesapeake Bay Program 1998b).

LAKE CHAMPLAIN SEDIMENT TOXICS ASSESSMENT PROGRAM

Phase II of the Lake Champlain Sediment Toxics Assessment Program investigated the three most contaminated areas of the lake: Cumberland Bay, Burlington Harbor, and Malletts Bay (McIntosh et al. 1997). Sediments of Cumberland Bay at Plattsburgh, New York are contaminated with PCBs from local historical sources and a possible source entering from the Saranac River. Water column concentrations in Cumberland Bay generally range from 0.3 to 2.1 ng/L, approximately an order of magnitude higher than in the main portion of the lake which has concentrations similar to present day levels in the Great Lakes. Water column concentrations of PCBs decreased dramatically with distance from the PCB "hot spot" near Wilcox Dock. Maximum concentrations near Wilcox Dock are up to 41 ng/L or 100 times greater than levels in the main body of the lake (McIntosh et al. 1997). The New York State Department of Environmental Protection recently completed a cleanup plan for Cumberland Bay, which will remove the most contaminated sediment near Wilcox Dock.

Surface sediment analyses at Burlington Harbor reveal that cadmium, copper, chromium, lead, and zinc display no clear spatial distribution, suggesting non-point sources (e.g., storm water, atmospheric deposition) for these metals. Bioavailability of metals appears to vary seasonally. Organic contaminants in Burlington Harbor (including PCBs, PAHs, and selected pesticides) generally exceeded the low estimate of sediment contamination levels at which adverse effects are predicted to occur among sensitive species or sensitive life stages. The PAHs and DDE exceeded the estimate of the sediment concentration levels at which toxic effects are predicted to occur among most species. Toxicity tests and benthic community analyses in Burlington Harbor showed chronic toxicity for a limited number of species, but the harbor does not appear to exhibit widespread hazardous conditions for aquatic life. In outer Mallets Bay, sediments frequently exceeded severe effects levels for some metals; however, mercury, lead, and cadmium were not the responsible contaminants (McIntosh et al. 1997).

SAN FRANCISCO ESTUARY REGIONAL MONITORING PROGRAM FOR TRACE SUBSTANCES

The San Francisco Estuary Regional Monitoring Program for Trace Substances began in 1993, and results are available from monitoring through 1996. This program collects data on water, sediment, and tissue contamination levels for a variety of pollutants, including some Great Waters pollutants of concern. Analyses of water column concentrations indicate that PCBs, PAHs, certain chlorinated pesticides, and mercury exceed water quality criteria (Table II-28). In sediments, mercury, total DDTs, and dieldrin frequently exceed levels at which adverse ecological effects are possible. An analysis of contaminant bioaccumulation by bivalves indicates that PCBs and PAHs are above the maximum tissue residue levels (MTRLs, relatively recently developed, science-based guidelines). Polychlorinated biphenyls, dioxin, mercury, dieldrin, DDT, and chlordane concentrations in fish tissue exceed EPA screening values for human consumption. Lead levels are usually below water and sediment quality guidelines and have not shown evidence of bioaccumulation or biological effects. In comparing data over several years, concentrations of mercury, lead, and chlordane are decreasing in tissues. Overall, the condition of the San Francisco estuary seems to be improving over time. Additional trends analyses will be conducted as additional data are collected (San Francisco Estuary Institute 1997).

Table II-28
Percentages of 1996 Samples that Exceeded Guidelines in the San Francisco Estuary

Pollutant	Water (Dissolved)	Water (Total)	Sediment	Bivalve Tissue
Cadmium	0	0	0	--
Mercury	0	19	81	0
Lead	0	6	6	--
PCBs	NA	93	6	100
PAHs	NA	0	38	100
Chlordane	NA	12	31	89
Dieldrin	NA	8	68	78
p,p'-DDE	NA	16	71	15

-- = no consistent guidelines
Source: San Francisco Estuary Institute 1997

CHAPTER III
MAJOR PROGRAMS AND ACTIVITIES

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This chapter summarizes major programs and activities undertaken or completed since the *Second Great Waters Report to Congress*. All of these programs and activities are helping to address issues relevant to the Great Waters program. The focus of this chapter primarily is on regulatory and policy initiatives as opposed to new science or research, which is covered in Chapter IV. Although some of the programs and activities described are being performed in conjunction with the EPA's Great Waters program, most are not. Many of the activities are led by partners outside the EPA, and have been included because they contribute to emission reductions and loadings of Great Waters pollutants of concern.

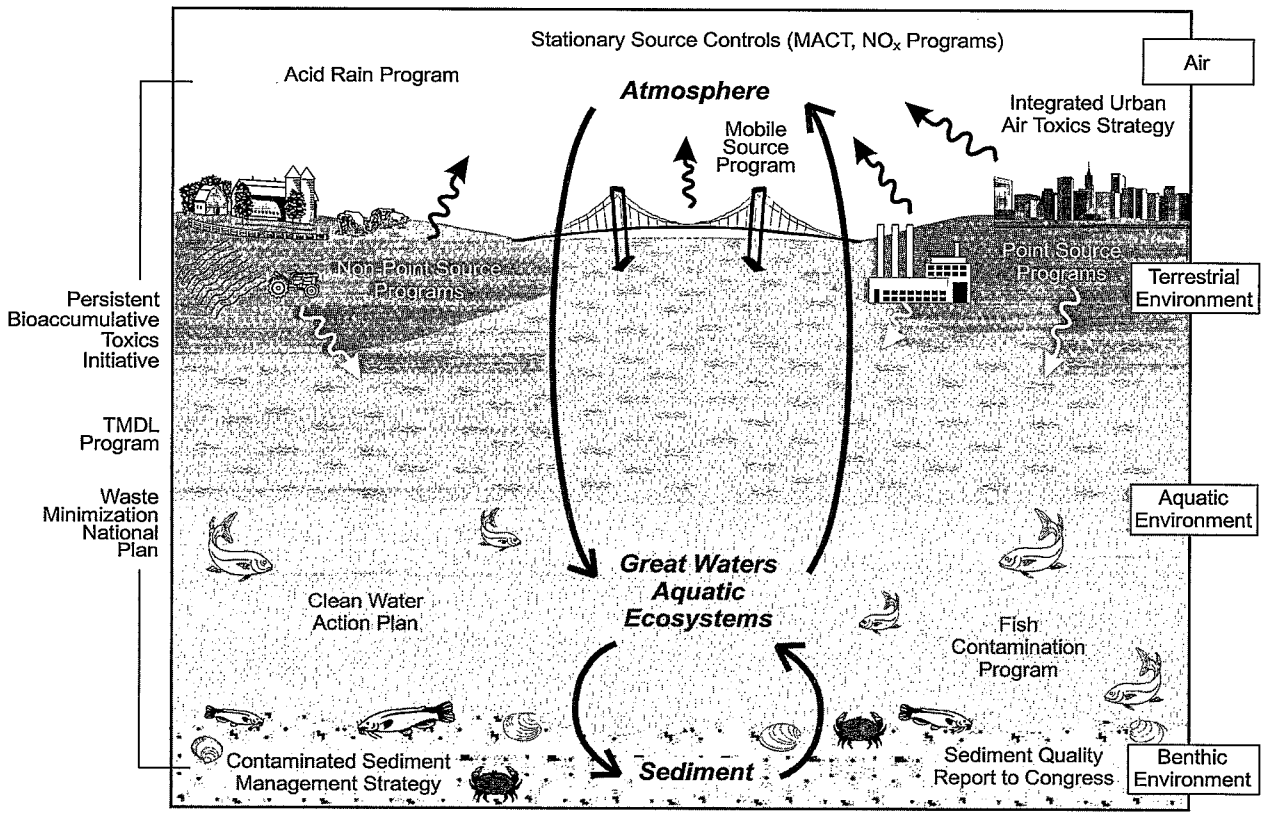
USDA Natural Resources Conservation Service, Romy Myszka

CHAPTER 3 HIGHLIGHTS

- There are more than 60 programs in progress, described in this chapter, that directly or indirectly address issues of concern to the Great Waters.
- Programs described in this chapter reduce the use of Great Waters pollutants of concern, reduce emissions of pollutants, restore (e.g., by sediment remediation) the Great Waters where they have been impacted, mitigate (e.g., by reducing consumption of contaminated fish) human health or ecological effects of Great Waters pollution, and provide a better scientific understanding of the sources, processes, and effects of and solutions to atmospheric deposition.
- Essentially all of EPA's Program Offices and most Regional Offices are involved in national, regional, or waterbody-specific activities addressing Great Waters issues.
- The EPA activities affecting Great Waters issues are performed under EPA's traditional media- and statute-specific programs (e.g., CAA programs) and under Agency-wide and inter-program multimedia initiatives (e.g., the Persistent Bioaccumulative Toxics Initiative (PBTI), Clean Water Action Plan).
- In addition to EPA, numerous other parties (including other Federal agencies; municipal, State, tribal and international governing bodies; the private sector; and, academic researchers) are leading programs which address Great Waters issues and pollutants of concern.
- The majority of the programs and activities described in this chapter involve collaborative partnerships, and many use community-based approaches to address local environmental priorities that are also Great Waters issues.

III.A NATIONAL PROGRAMS AND ACTIVITIES

A number of EPA's national programs and activities contribute to understanding and reducing pollutant impacts on the Great Waters. As discussed in Section I.B, most of the Great Waters pollutants of concern are also the focus of other national programs and activities. These programs and activities protect and enhance the quality of surface waters throughout the U.S., including the Great Waters. Recent accomplishments of these other programs and activities are summarized below, organized by whether they are considered multimedia activities, HAP-specific controls, mobile source program activities, or other national programs.



MULTIMEDIA ACTIVITIES

Persistent Bioaccumulative Toxics Initiative

The Persistent Bioaccumulative Toxics Initiative (PBTI) which was developed in an EPA-wide effort chaired by the Office of Prevention, Pesticides and Toxic Substances (OPPTS), supports Great Waters program goals by helping to reduce environmental releases of certain Great Waters pollutants of concern. The goal of the PBT Initiative is to further reduce risks to human health and the environment from existing and future exposure to persistent, bioaccumulative, and toxic (PBT) pollutants. The initiative seeks to accomplish this goal through increased coordination among EPA national and regional programs with the significant involvement of international, State, local, and tribal organizations, the regulated community, environmental groups, and private citizens. This effort fortifies existing EPA commitments related to priority PBTs, such as the 1997 Canada-U.S. Binational Toxics Strategy (BNS), the North American Agreement on Environmental Cooperation, and EPA's Clean Water Action Plan (see below for more information on these activities).

Guiding Principles for PBT Initiative

- Address problems on multimedia basis through integrated use of all EPA tools
- Coordinate with and build on relevant international efforts
- Coordinate with relevant Federal programs and agencies
- Stress cost-effectiveness
- Involve stakeholders
- Emphasize innovative technology and pollution prevention
- Protect vulnerable subpopulations
- Base decisions on sound science
- Use measurable objectives and assess performance

The PBT Initiative initially will focus on the 12 priority pollutants identified under the Binational Toxics Strategy (BNS) (see sidebar below). The initiative includes the following steps:

1. *Develop and implement national action plans for priority PBT pollutants.* Near-term activities include pollution prevention projects, enforcement and compliance assistance, development or revision of water quality criteria, research and analysis of emission and discharge controls and other topics, and collaboration with other international efforts beyond the BNS. The draft Mercury Action Plan has been developed and includes regulatory and nonregulatory initiatives.

Binational Toxics Strategy - Level I Substances

Aldrin/dieldrin*	Mercury and compounds*
Benzo(a)pyrene*	Octachlorostyrene
Chlordane*	PCBs*
DDT (DDD/DDE)*	Dioxins and furans*
Hexachlorobenzene*	Toxaphene*
Alkyl lead*	
Mirex	

*Great Waters pollutants of concern

2. *Screen and select more priority PBT pollutants for action.* The EPA will apply selection criteria for additional pollutants in consultation with a technical panel.

3. *Prevent introduction of new PBTs.* The EPA is acting to prevent new PBT chemicals from entering commerce by (1) proposing criteria for new PBT chemicals, (2) developing a rule to prevent re-introduction of phased out PBTs, (3) developing incentives to reward development of

PBT alternatives, and (4) documenting how PBT-related screening criteria are being taken into account for approval of new pesticides and re-registration of existing pesticides.

4. *Measure progress.* The EPA is in the process of defining measurable objectives to assess progress. These could include trend detection in environmental health, direct measurements of human exposure through biomarkers, tracking of chemical releases to the environment, and program activity measures such as enforcement actions.

The EPA solicited public comments on a draft PBT Initiative published in late 1998. For further information on the status of the initiative, see the web page at www.epa.gov/pbt/strtegy.htm.

Total Maximum Daily Loads

Under the Federal Clean Water Act of 1972 (CWA), EPA is required to develop effluent guidelines for specific categories and classes of point sources. The guidelines are used to set discharge limits for specific facilities that discharge pollutants to surface waters or to municipal sewage treatment systems (63 FR 22644, April 27, 1998). However, many U.S. waterbodies do not meet applicable water quality standards, which include standards for many Great Waters pollutants of concern, despite the implementation of the CWA (see sidebar), in part because of non-point source pollution, including atmospheric deposition. Total maximum daily loads established under section 303(d)(1) of the CWA, provide a framework for addressing pollution from both point and non-point sources. A TMDL is developed for a waterbody if water quality standards within the waterbody are not being met using technology-based or other effluent controls. A TMDL establishes the maximum allowable pollutant loading for a waterbody (including allocations for point source loads and non-point source loads, and a margin of safety) that will result in the waterbody meeting established water quality standards. Specifically, TMDLs assess non-point source loads, such as atmospheric deposition, in addition to point source inputs. In some cases, TMDLs attempt to identify the source of atmospheric deposition in order to implement appropriate measures to decrease the pollutant inputs to a watershed. In terms of atmospheric deposition, EPA is developing science and tools to assess the contribution of atmospheric sources to water pollution and to assist in decreasing total pollutant loadings to waterbodies.

National Water Quality Inventory

"The final National Water Quality Inventory Report to Congress for 1996 indicates that of the 72 percent of estuary waters assessed, 38 percent are not fully supporting uses/standards and 4 percent are threatened." (U.S. EPA 1998o).

Mercury TMDL Air Deposition Pilot Project

In order to assist States in preparing TMDLs for waterbodies affected by atmospheric pollutants, the EPA Office of Water (OW) and Office of Air Quality Planning and Standards (OAQPS) have initiated the Mercury TMDL Air Deposition Pilot Project. This project, a collaborative effort between OAQPS, OW, the Office of Research and Development, EPA regional offices and State agencies, will investigate tools and approaches for developing TMDLs in cases involving atmospheric pollutants. The project will design, develop, and identify uncertainties and data needs for pilot mercury TMDLs for two waterbodies receiving mercury contributions from the air. In the process, the compatibility of air and water quality modeling systems, as well as the linkages between the CWA and CAA will be evaluated. Two pilot TMDL waters have been selected for study (Devil's Lake, Wisconsin, and the Everglades Conservation Area 3a, Florida). Modeling work is under way.

As required by the CWA, States are directed to identify and directs States to identify and establish a priority ranking for waters that do not meet applicable water quality standards after application of technology-based and other controls, taking the severity of the pollution and the designated uses of the waterbody into consideration. The EPA's implementing regulations require States to submit these lists every 2 years. Once the list of priority waters is

approved by EPA, the State establishes TMDLs for each waterbody on the list to restore water quality. The TMDL specifies the amount of pollution or other stressor that needs to be reduced to meet water quality standards, allocates pollution control or management responsibilities among sources in a watershed, and provides a scientific and policy basis for taking actions needed to restore a waterbody (U.S. EPA 1998o). A TMDL may also identify the need for point source or non-point source controls. The EPA recently began development of a TMDL pilot project addressing atmospheric deposition of mercury (see sidebar).

A variety of tools have been created to assist States in the process of developing TMDLs. Many of the tools were created in response to challenges encountered in allocating non-point source inputs of nitrogen compounds to specific land uses. Additional tools have been developed to assist in decreasing loadings to waterbodies. See, for example, the discussion of the Albemarle-Pamlico Estuary (NC) under the National Estuary Program in Section III.B.

Also, to assist in the implementation of TMDLs in States, EPA published a report, *TMDL Development of Cost Estimates: Case Studies of 14 TMDLs* (U.S. EPA 1996b). The selected TMDL case studies are from a variety of geographic locations, address the most common pollutants, range from small- to large-scale projects, and represent a range of complexity levels. The report also identifies funding sources and discusses benefits of TMDLs (U.S. EPA 1998v).

Pulp and Paper Cluster Rule

In April 1998, EPA promulgated the pulp and paper cluster rule – a joint CAA and CWA rule – which is designed to protect human health and the environment by reducing releases of toxic pollutants from the pulp and paper industry to air and water. This rule is the first integrated regulation to control the release of pollutants to more than one media from one industry. Implementation of the rule will further reduce paper industry air emissions and surface water discharges of certain Great Waters pollutants of concern (e.g., dioxins and furans).

By issuing joint standards, industry can consider all regulatory requirements at one time; therefore, reducing the regulatory burden and allowing mills to select the best combination of pollution prevention and control technologies that will provide the greatest protection to human health and the environment. The cluster rule requires new and existing pulp and paper mills (1) to capture and treat toxic air pollutant emissions that occur during cooking, washing, and bleaching stages of the pulp manufacturing process and (2) to meet new effluent limits for toxic pollutants in the wastewater discharged during the bleaching process and in the final discharge from mills in the bleached papergrade kraft and soda subcategory and in the bleach papergrade sulfite subcategory. The rule limits releases of toxic air pollutants from processes that are used at 155 of the 565 U.S. pulp and paper mills (i.e., the rule applies to paper and paperboard mills, also referred to as kraft, soda, sulfite, and semi-chemical mills) along with water discharges of toxics and other pollutants from the 96 of those 155 mills that bleach pulp to make paper. The new water limits are based on substituting chlorine dioxide for chlorine in the bleaching process (63 FR 18504, April 15, 1998).

The new air and water standards under the pulp and paper cluster rule will provide significant environmental benefits, including:

- Seventy-three rivers and streams will become cleaner because of toxic release reductions;
- Emissions of over 160,000 tons of toxic air pollutants will be eliminated;

- Dioxin and furan discharges to water will be reduced by 96 percent; and,
- Ultimately, all dioxin fish consumption advisories associated with the 96 mills affected by this action will be eliminated (63 FR 18504, April 15, 1998).

The pulp and paper cluster rule also includes an innovative voluntary incentives program (i.e., the Voluntary Advanced Technology Incentives Program). Under this program, mills are voluntarily subject to more stringent standards in return for rewards, such as increased compliance time, reduced monitoring requirements and inspections, greater permit certainty, reduced penalties, and public recognition (63 FR 18504, April 15, 1998). For example, mills participating in the program are allowed 6 years instead of 3 years to comply with air standards and 6 to 16 years to comply with water discharge permit limits, depending on the performance level of the new technology or process change. This program also encourages mills to consider all technology options prior to making large investment decisions, such as purchasing new emissions control devices or implementing major process changes (U.S. EPA 1997d). In the long term, this innovative program could result in additional reductions in air toxics releases and water pollutant discharges.

Mercury Research Strategy

The EPA, in an intra-agency effort led by EPA's Office of Research and Development, is developing a *Mercury Research Strategy*, which is expected to be completed in 2000. The EPA plans for the strategy to describe:

- The key scientific questions of greatest concern to EPA for mercury risk assessment and risk management that EPA plans to investigate over the coming 5 years; and,
- A research program which would provide information, methods, models, and data to address these key scientific questions.

The research strategy is intended to guide EPA's development of research plans and decisions about future research priorities and budgets. It may also provide useful information to others in guiding their research. However, the strategy is not intended to convey information on specific projects, nor will it provide a detailed schedule of outputs or products.

Clean Water Action Plan

Completed in February 1998, the Clean Water Action Plan is an interagency, multimedia strategy to address remaining obstacles to the original goal of the Clean Water Act – "fishable and swimmable" water for all Americans (U.S. EPA 1998a). The action plan was requested by Vice President Al Gore on October 1997 to mark the 25th anniversary of the Clean Water Act. It forms the core of President Clinton's Clean Water Initiative, which was proposed in the 1998 State of the Union Address. Together, the Clean Water

**Clean Water Action Plan:
Key Actions to Assess and Reduce Air Deposition of Nitrogen**

"EPA and NOAA will work with other Federal, State, tribal, and local government agencies and others to better quantify the risks associated with atmospheric deposition of nitrogen compounds and other pollutants to waterbodies."

"EPA will work through the TMDL program to evaluate the linkage of air emissions to the water quality impacts to help determine appropriate reduction actions. EPA will work with States, tribes, and Federal land management agencies to employ both Clean Water Act and Clean Air Act authorities to reduce air deposition of nitrogen compounds and other pollutants that adversely affect water quality." (U.S. EPA 1998a)

Action Plan and Initiative outline specific actions to strengthen and expand efforts to restore and protect water resources.

The plan identifies non-point sources (including atmospheric deposition) as the most important remaining threat to water quality. Because EPA's existing water programs do not focus on control of non-point sources, the action plan emphasizes innovative approaches such as partnerships with local stakeholders and watershed-level projects. Atmospheric deposition is among the leading non-point sources addressed by the action plan. In particular, agencies pledged to work together to better assess the risks associated with atmospheric deposition of nitrogen compounds (see sidebar) and other pollutants to waterbodies and to integrate air deposition into TMDL evaluations. In addition, EPA will include air deposition in a multiagency coastal research strategy and coordinated coastal monitoring plan, expected to be issued in 2000.

Another action in the President's Clean Water Action Plan is to conduct a national survey of levels of persistent bioaccumulative toxic (PBT) chemical levels in fish and shellfish throughout the country. Specifically, EPA and NOAA are conducting a study to estimate the national distribution of the mean levels of selected PBT chemical residues in fish and shellfish tissue in U.S. waters. The study will provide information for the Agency's PBT Initiative, which seeks to identify potential areas of concern for human and/or ecological health. The study of fish tissue may reveal where PBT chemicals not previously considered a problem are present in the environment at levels of concern. For the national fish study, fish will be obtained from lakes and reservoirs which have been selected according to a probability design. The shellfish survey will be based on the data obtained by NOAA's ongoing Mussel Watch Project. Both studies will be coordinated with State and tribal efforts to maximize geographic coverage.

Sediment Quality Report to Congress

Once deposited to surface waters from the air or other sources (e.g., industrial and municipal point discharges, urban and agricultural runoff), most of the Great Waters pollutants of concern tend to accumulate in sediments where they may reach concentrations harmful to aquatic life and the food web. In recognition of environmental and economic problems associated with contaminated sediment, Congress included in the Water Resources Development Act (WRDA) of 1992 a requirement for EPA, in cooperation with NOAA and the U.S. Army Corps of Engineers, to conduct a comprehensive national survey of data regarding the quality of aquatic sediments in the U.S.

In September 1997, EPA's Office of Science and Technology within the Office of Water published the first biennial Sediment Quality Report to Congress, entitled the *Incidence and Severity of Sediment Contamination in the Surface Waters of the United States*. The report consisted of three volumes:

Volume 1: The National Sediment Quality Survey (U.S. EPA 1997i);
Volume 2: Data Summaries for Areas of Probable Concern (U.S. EPA 1997j); and,
Volume 3: The National Sediment Contaminant Point Source Inventory (U.S. EPA 1997k).

The National Sediment Quality Survey (NSQS) (U.S. EPA 1997i) describes the accumulation of chemical contaminants in river, lake, ocean, and estuary bottoms and includes a screening assessment of the potential for associated adverse effects on human and environmental health. Key findings of the NSQS are discussed in Chapter II.

In developing the NSQS, EPA compiled all available computerized data on the quantity, chemical and physical composition, and geographic location of pollutants in sediment. The database is referred to as the National Sediment Inventory (NSI) and is the largest set of sediment chemistry and related biological data ever compiled by EPA. The NSI will be updated on a regular basis in order to assess trends in both sediment quality and the effectiveness of existing regulatory programs at the Federal, State, and local levels. The NSI is discussed further in Chapter IV.

The EPA's mandate to investigate sediment contamination in the Nation's water included a directive to identify potential pollutant sources. Volume 3 of the Sediment Quality Report to Congress (U.S. EPA 1997k) evaluated point sources (i.e., direct discharges to waterbodies). In future biennial Reports to Congress, EPA will assess loadings from non-point sources, including harvested croplands, urban areas, atmospheric deposition, and abandoned and inactive mine sites (U.S. EPA 1997i). To prepare the non-point source inventory for the second Sediment Quality Report to Congress, EPA is currently compiling data from the Bureau of the Census, the U.S. Department of Agriculture, the U.S. Department of the Interior's U.S. Geological Survey and Bureau of Mines, and others.

Contaminated Sediment Management Strategy

The EPA is using data compiled for the Sediment Quality Report to Congress and other resources for a multiprogram, multimedia effort to coordinate and streamline contaminated sediment management decisions within the Agency. In April 1998, EPA's Office of Water completed the Contaminated Sediment Management Strategy (U.S. EPA 1998d), which outlines the following specific actions:

1. Control sources of sediment contamination and prevent the volume of contaminated sediment from increasing;

2. Reduce the volume of existing contaminated sediment;

3. Ensure that sediment dredging and dredged material disposal are managed in an environmentally sound manner; and,

4. Develop scientifically sound sediment management tools for use in pollution prevention, source control, remediation, and dredged material management.

The Contaminated Sediment Management Strategy identifies atmospheric deposition as an important source of sediment contamination. Specifically, the strategy directs EPA's Office of Air and Radiation to use the National Sediment Inventory (NSI) to evaluate the contribution of atmospheric deposition to sediment quality problems. This new tool will enable EPA to better assess trends in sediment pollution, including pollution from atmospheric deposition, and focus cleanup and pollution control activities. In addition, the strategy identifies the Agency's Great Waters program as a significant component of its coordinated effort to address contaminated sediment problems.

The Contaminated Sediment Management Strategy includes a research component designed to identify relationships between sediment contaminants and the viability and sustainability of benthic ecosystems. Ultimately, the research will help to formulate source control and pollution prevention strategies. In addition, the strategy outlines coordinated, multiprogram efforts of research and policy development to ensure that uniform exposure and effects assessment procedures for contaminated sediments are used throughout the Agency. For example, EPA proposed, and is currently developing, standard sediment toxicity test methods and chemical-specific sediment quality guidelines. The strategy

proposes several specific uses of the assessment procedures and sediment quality guidelines, and the Agency has begun to develop an *Equilibrium Partitioning Sediment Guidelines User's Guide*.

Waste Minimization National Plan

The Waste Minimization National Plan (WMNP), which EPA's Office of Solid Waste developed in 1994, is a voluntary, long-term effort to reduce the quantity and toxicity of hazardous waste through source reduction and recycling, including wastes bearing Great Waters pollutants of concern such as mercury and dioxin. The plan calls for a 50 percent reduction in the presence of the most persistent, bioaccumulative, and toxic (PBT) chemicals in hazardous waste by 2005 compared to a baseline year of 1991. This goal was also adopted as a Government Performance and Results Act (GPRA) measurement goal.

To assist in implementing the WMNP, the Office of Solid Waste and the Office of Pollution Prevention and Toxic Substances have developed a draft Windows-based software tool to prioritize PBT chemicals for waste minimization efforts. The Waste Minimization Prioritization Tool (WMPT) provides a screening-level assessment of potential chronic risks that chemicals, including most Great Waters pollutants of concern, pose to human health and the environment, based on their persistence, bioaccumulative potential, and human and ecological toxicity. More information about the WMPT can be found on EPA's waste minimization home page at www.epa.gov/wastemin.

The WMPT served as a starting point in developing the draft Resource Conservation and Recovery Act PBT Chemicals List (63 FR 60332, November 9, 1998). Other factors, such as quantity, prevalence, environmental presence, and degree of concern to the RCRA program, were used in the selection of chemicals for the draft list. The final list of chemicals is expected to be published in the *Federal Register* in 2000. This final list will serve to focus national waste minimization efforts and track progress toward the 2005 reduction goal.

Air Characteristic Study

The Air Characteristic Study currently being conducted by EPA's Office of Solid Waste addresses the question of whether some industrial wastes should be classified as hazardous because of risks posed by their air emissions. The overall goal of this study is to estimate the maximum waste constituent concentrations that could be present in certain waste management units and still be protective of human health.

The study is estimating potential risk to humans for 105 chemical constituents, of which 88 are HAPs under the CAA and several are Great Waters pollutants of concern (e.g., lead compounds, mercury, benzo[a]pyrene, dioxin, and others). Draft results of the risk analysis indicate that volatile toxic chemicals managed in non-storage tanks, such as aerated wastewater treatment tanks, pose the highest risk, with the waste concentrations for these aerated tanks differing from other units by an order of magnitude or more. Storage tanks, land application units, landfills, and waste piles followed aerated tanks in ranking of risk. The findings of this study, due to be completed in 2000, will assist EPA in exploring the need for regulatory changes under RCRA for these waste management units and in investigating possible options for risk reduction.

HAZARDOUS AIR POLLUTANT (HAP) CONTROLS

Under the CAA, EPA is required to regulate sources of 188 listed HAPs. All but two of the pollutants identified as Great Waters pollutants of concern are listed as HAPs. (The two Great Waters pollutants that are not HAPs are nitrogen and dieldrin.) On July 16, 1992, EPA published a list of 174 industry groups (known as source categories) that emit one or more of these air toxics. For listed categories of "major" sources (those with the potential to emit 10 tons/year or more of a listed pollutant or 25 tons/year or more of a combination of pollutants), the CAA requires EPA to develop standards under section 112(d) that require the application of air pollution reduction measures known as MACT. This performance-based approach requires EPA to set standards based on consideration of those controls in use at the best controlled facilities within an industry.

The CAA provided a 10-year schedule in which to promulgate these technology-based standards with certain standards being promulgated in the first 2 years, 25 percent in the first 4 years, an additional 25 percent no later than the 7th year, and the remaining 50 percent no later than the 10th year. The EPA has been productive in fulfilling these statutory requirements and, working in partnership with States, has built the necessary infrastructure for implementing the air toxics regulations. For the 45 source categories in the 2- and 4-year groups, EPA estimates that the regulations will reduce air toxics emissions by approximately one million tons per year. For the 42 source categories in the 7-year group, EPA has either proposed or promulgated regulations that are estimated to reduce air toxics emissions by roughly 500,000 tons per year. A list of all source categories, the MACT implementation schedule, and references to proposed and final rules is included in Appendix C of the *Residual Risk Report to Congress* (U.S. EPA 1999d).

Some regulations are already in place under section 112(d) with sources currently in compliance. The source categories affected by these rules are listed in Table III-1 below along with the emission reductions of the affected pollutants of concern, where available.

Table III-1
Source Categories With Effective Compliance Dates and Anticipated Reductions of HAP Emissions

Source Category	Compliance Date	Pollutant	Nationwide Pre-MACT Emissions (tpy)	Nationwide Expected Percent Reductions
Coke oven batteries: Charging, leaks, and bypass/bleeder stacks	01/01/98	Coke oven emissions[a]	1600	94
Secondary Lead Smelting	12/23/97	All HAPs[b]	1900	65
		Lead Compounds	120	40

[a] Coke oven emissions include POM.
[b] HAP emissions for this category include lead compounds, dioxins/furans, mercury, and POM among other pollutants. Of these, lead compounds is the only pollutant of concern for which pollutant-specific estimates are available.

Section 112(c)(6) of the CAA directs EPA to focus attention on seven specific toxic pollutants – all of which are Great Waters pollutants of concern: alkylated lead compounds, hexachlorobenzene, POM, mercury, PCBs, dioxins, and furans. The Agency is to ensure that sources accounting for at least 90 percent of the emissions of each of these pollutants are subject to standards under section 112(d)

(described above). Section 112(c)(6) of the CAA requires EPA to identify the source categories that emit 90 percent of the aggregate emissions for each of the seven specific pollutants and add any source categories not previously identified to the list discussed above.

Under section 112(c)(6), a review of the available data indicated that nearly all source categories emitting the seven pollutant groups had already been listed for regulation under the CAA or were subject to comparable regulation under other CAA authorities. However, two additional source categories were added to the source category list in a final *Federal Register* notice on April 3, 1998. These two categories are open burning of scrap tires and gasoline distribution (Stage I Aviation), which includes evaporative losses associated with the distribution and storage of aviation gas containing lead. A comment and response document is available along with the 1990 emissions inventory for the seven pollutants at www.epa.gov/ttn/uatw/112c6/112c6fac.html.

A different section of the CAA (section 129) is devoted to control of certain air toxics, as well as other pollutants, from solid waste combustion units. The pollutants of concern to the Great Waters covered are lead, cadmium, mercury, dioxins and furans, and NO_x. This regulatory program is discussed later in this section.

Under sections 112 and 129, several rules have either been proposed or finalized, but the compliance date has not yet been reached. A summary of these actions and their anticipated emission reductions are listed below in Table III-2. In addition, under the joint authority of section 112 and the Resource Conservation and Recovery Act (RCRA), EPA's Office of Solid Waste finalized on July 30, 1999 (signed by the Administrator) new emission standards for existing and new cement kilns, incinerators, and lightweight aggregate kilns that burn hazardous wastes. These combustors burn about 80 percent of the hazardous waste combusted annually within the U.S. When fully implemented in 2002, the MACT standard for these sources is expected to achieve significant reductions in emissions of several Great Waters pollutants of concern, including dioxin/furans, mercury, lead and cadmium. These standards will also satisfy our obligation under RCRA to ensure that hazardous waste combustion is conducted in a manner adequately protective of human health and the environment.

The remaining 50 percent of MACT regulations are expected to be issued within the next 2 years (by 2002). Based on our current knowledge of the remaining industries slated for regulation under section 112(d), those that emit pollutants of concern to the Great Waters include chlorine manufacturing (chlor-alkali plants), coke ovens (pushing, quenching and battery stacks), industrial boilers, institutional and commercial boilers, iron and steel, and refractory manufacturing. There are more MACT rules for solid waste combustion under section 129 noted later in this section.

In addition to the standards development requirements of the CAA, there are a number of other HAP program requirements that will help reduce emissions of the Great Waters pollutants of concern. These are briefly described below. Additional information regarding the air toxic program can be found on the Internet at EPA's unified air toxics web site at www.epa.gov/ttn/uatw.

Mercury Study Report to Congress

The *Mercury Study Report to Congress*, issued by EPA in December 1997 (U.S. EPA1997e), is a comprehensive document detailing the U.S. mercury emissions inventory, fate and transport of mercury in the environment, human health effects, an ecological risk assessment, a human and wildlife risk characterization, and an assessment of control technologies and their costs. The report also outlines research needs. Pertinent results and conclusions from this report are described in the mercury and compounds section of Chapter 2.

Table III-2
Proposed and Final Rules Affecting Pollutants of Concern

Source Category	Status	Pollutants	Nationwide Pre-MACT Emissions (tpy)	Nationwide Expected Percent Reductions
Municipal waste combustion (large combustors, > 250 tons per day)	Final rule and guidelines	Dioxins/Furans[a]	0.0025	98
		Mercury[i]	54	78
		Lead	64	75
		Cadmium	4.2	67
		NO_x	54,000	36
Medical waste incineration	Final rule and guidelines	Dioxins/Furans[a]	0.0002	95-97 [b]
		Mercury	16	93-95 [b]
		Lead	12	80-87 [b]
		Cadmium	1.3	75-84 [b]
		NO_x	1,300	0-30 [b]
Pulp and paper cluster[c]	Final rule	HAPs	240,000	58
Primary aluminum production	Final rule	POM	2,000	50
Secondary aluminum production	Final rule	HAP metals[d]	64.4	62.5
		Dioxins/Furans[a]	0.0009	86.6
Primary copper production	Proposed rule	Cadmium	0	0
		Lead	140	13
		Mercury	0	0
Pesticide active ingredient production[e]	Final rule	HAPs[e]	4,255	65
Portland cement manufacturing - nonhazardous waste -fired	Final rule	Dioxins/Furans[a]	0.0005	36
		Mercury	4	0
		HAP metals[f]	[f]	[f]
Mineral wool production	Final rule	HAP metals[g]	1.1	91
Hazardous Waste combustion (existing and new cement kilns, incinerators, and lightweight aggregate kilns that burn hazardous waste)	Final rule	Dioxins/Furans[a]	.000044	70
		Mercury	6.5	55
		Lead and Cadmium	88.5	88
		Other HAP Metals[h]	9.8	75

Table III-2 (continued)
Proposed and Final Rules Affecting Pollutants of Concern

[a] Dioxin/Furan emissions are reported on a 2,3,7,8-TEQ basis.
[b] Ranges reflect different assumptions on the number of incinerator closures.
[c] Values represent air emissions affected by the cluster rule only. Values for individual HAPs were not provided in the final rule.
[d] The HAP metals for secondary aluminum production are mercury, lead, and cadmium, as well as eight other HAP metals.
[e] This rule covers 11 of the source categories listed for regulation. Values for individual HAPs are not available. Included are emissions of HCB and chlordane, although they are not the most prevalent HAPs in this category.
[f] The rule includes a particulate matter limit, which serves as a surrogate for all non-volatile and semi-volatile HAPs, including metals. These metals are estimated to be no more than 1 percent of the total particulate matter HAPs. The rule is estimated to achieve about 20 percent reduction in particulate matter emissions.
[g] The HAP metals for mineral wool production are cadmium and lead, as well as seven other HAP metals.
[h] The other HAP metals for hazardous waste combustion include antimony, cobalt, manganese, nickel, and selenium.
[i] Emission reductions for municipal waste combustion are often cited from a 1990 baseline, other than the pre-MACT baseline presented here. For example, the rule and guidelines will reduce mercury emissions by greater than 90 percent from 1990 levels when fully implemented.

The *Mercury Study Report to Congress* is not a regulatory effort; currently it is being broadly used in support of Great Waters activities, the PBT Initiative, the Binational Toxics Strategy, the mercury research strategy, and other EPA efforts to understand and control this pollutant.

Utility Air Toxics Study and Regulatory Determination

In February 1998, EPA issued a study of the public health impacts of emissions of air toxics from utilities that burn fossil fuel (U.S. EPA 1998p). About 67 air toxics were found to be emitted from utilities, including mercury and dioxins. The report includes (1) a description of the utility industry; (2) an analysis of air toxics emissions data from coal-, oil-, and gas-fired utility plants; (3) an assessment of risks to public health from exposure to air toxics emissions through inhalation; (4) an assessment of potential risks to public health from exposure to four specific air toxics (i.e., radionuclides, mercury, arsenic, and dioxins) through other indirect means of exposure (e.g., food ingestion, dermal absorption); (5) a general assessment of the fate and transport of mercury through environmental media; and, (6) a discussion of alternative control strategies.

The report indicates that, although uncertainties in the analysis exist, on balance, mercury from coal-fired utilities is the HAP of greatest potential public health concern. The report identifies three other air toxics for which there are some potential concerns and uncertainties that may need further study: dioxins, arsenic, and nickel.

The CAA also requires EPA to make a determination, after considering the results of the utility study, as to whether emission controls for air toxics are appropriate and necessary for utility boilers. The EPA has delayed this determination until it collects additional information, and EPA's Office of Air and Radiation is currently collecting the following mercury emissions data from electric utility steam generating units:

- Current information on the type of coal they use and on their method of particulate matter (PM) and sulfur dioxide (SO_2) control at all "traditional" coal-fired electric utility steam generating units;

- Current information on the fuel they use and on their method of PM and SO_2 control at all independent power producers that could be identified as possibly burning coal;

- 1 year of mercury-in-coal analyses at all coal-fired units meeting the section 112(a)(8) definition of "electric utility steam generating unit"; and

- One series of speciated mercury emissions testing at a randomly selected subset of coal-fired units.

The regulatory determination is scheduled to be provided by December 15, 2000 (U.S. EPA 1998p).

Residual Risk Report to Congress

Under section 112(f) of the CAA, EPA is required to develop and implement a program for assessing risks remaining (i.e., the residual risk) after facilities have implemented MACT standards, and to promulgate rules, if necessary, to protect the public health with an "ample margin of safety" or to prevent adverse environmental effects. Additional risk-based regulations, if needed, are to be promulgated within 8 years after EPA promulgates an air toxics standard for a given source category. The first such risk-based regulations, if necessary, are due in 2002. If promulgated, residual risk standards could further reduce emissions of Great Waters pollutants of concern.

In March 1999, EPA issued the *Residual Risk Report to Congress*. This report reviews EPA human and ecological risk assessment methods, identifies data sources and data collection needs for conducting risk assessments, proposes methods on how to close data gaps, discusses how results of residual risk assessments will be used in the residual risk program, and includes an appendix of all MACT source categories, the MACT implementation schedule, and references to proposed and final rules. In addition, the report discusses the strategy or "framework" EPA will use in conducting residual risk assessments.

The risk assessment framework under the residual risk program was developed using knowledge gained from past risk assessments and information from other regulatory agencies and guidance from reports. This strategy calls for an iterative, tiered assessment of the risks to humans and ecological receptors through inhalation and, where appropriate, non-inhalation exposures to air toxics. The residual risk assessment framework will allow the Agency to be flexible in its decisions while ensuring that public health and the environment are protected. The EPA's objectives also include integration of all portions of the Federal air toxics program, continuing the partnership with State/local programs in the sharing of data and expertise, and including groups who may be affected by residual risk decisions (e.g., industry, public interest groups) as part of the process.

Integrated Urban Air Toxics Strategy and Report to Congress

As part of its overall efforts to reduce air toxics, EPA published the integrated urban air toxics strategy in the *Federal Register* on July 19, 1999 (64 FR 38706). The strategy presents a framework for addressing air toxics in urban areas as required by section 112(k) of the CAA. The goals of the strategy are to reduce by 75 percent the risk of cancer and substantially reduce non-cancer risks associated with air toxics while ensuring that disproportionate risks are addressed. Specifically, the strategy does the following:

- *Outlines EPA's approach for assessing health risks.* The EPA will evaluate risks considering the multiple sources of air toxics in our cities, whether they come from major industrial sources, smaller sources (like drycleaners or gas stations), or cars and trucks. This includes risks from consuming fish from waters contaminated by urban air toxic deposition.

- *Builds on the substantial emission reductions already achieved from cars, trucks, fuels, and industries such as chemical plants and oil refineries.* The strategy outlines actions to reduce emissions of air toxics and to improve EPA's understanding of the health risks posed by air toxics in urban areas.

- *Identifies a list of the 33 air toxics that pose the greatest threat to public health in urban areas, considering multipathway exposure, such as fish consumption, in the identification process.* These 33 air toxics are a subset of the 188 air toxics and include the Great Waters pollutants of concern mercury, cadmium, lead, dioxins and furans, POM, PCBs, and HCB.

- *Identifies the 30 of these 33 urban air toxics with the greatest contribution from smaller commercial and industrial operations or so-called "area" sources.* The CAA requires EPA to ensure that 90 percent of the aggregate emissions of each of the 30 identified HAPs are subject to regulation through EPA's established air toxics program. In order to address this requirement, EPA identified 29 area source categories that are significant contributors to the emissions, including sources of mercury, cadmium, lead, POM, dioxins and furans. Currently, EPA has regulations under development or completed for 16 of these area source categories and intends to develop regulations for the remaining 13 area source categories over the next 5 years. The EPA intends to list additional area sources by 2003 as better inventory data become available.

The strategy also addresses the Agency's efforts to date to assess the public health risk from air toxics from mobile sources and highlights EPA's expectation for additional regulations targeting toxics emissions from motor vehicles and fuels. In the strategy, EPA describes plans to consider diesel emissions in the upcoming mobile source air toxics regulation and to issue a rule (the proposed "Tier II rule"; see page III-29) which will reduce levels of diesel emissions significantly in both urban and rural areas.

Rules for Solid Waste Combustion, Including Large Municipal Waste Combustors and Hospital/Medical/Infectious Waste Incinerators (CAA Section 129)

Section 129 of the CAA directs EPA to control solid waste combustion and set emission limits for dioxins and furans, cadmium, lead, mercury, and NO_x (all pollutants of concern to the Great Waters), as well as particulate matter, opacity, sulfur dioxide, carbon monoxide, and hydrogen chloride. For existing solid waste combustion units, section 129 requires EPA to develop emission guidelines. These guidelines do not directly regulate the units. Rather, they establish requirements for State plans, which are the vehicle by which States implement the guidelines. For new units, section 129 requires EPA to develop technology-based performance standards following section 111 of the CAA. Section 129 further subjects solid waste combustion units to the section 112(f) residual risk program, which was discussed earlier in this section.

Final rules are now in place for large municipal waste combustors (MWC) and for hospital/medical/infectious waste incinerators (HMIWI, or often called medical waste incinerators). There are also rules under development for small municipal waste combustors, commercial/industrial waste incineration, and other solid waste incineration. The commercial/industrial waste incineration rule is planned to be finalized by November 15, 2000; the small municipal waste combustor rule is planned to be finalized by 2001.

Large MWC are those units with a capacity of at least 250 tons of waste per day. The EPA initially promulgated standards for new units and guidelines for existing units on December 19, 1995 and revised them on August 25, 1997. The 24 States with large MWCs were required to submit emission guidelines implementation plans to EPA by December 19, 1997. The State plans include source and emission inventories, testing and monitoring, as well as generic or site-specific compliance schedules. The MWC Federal Plan adopted in November 1998 applies to large MWCs until State plans are approved. The Federal Plan ensures that large MWCs are on track to complete pollution control equipment retrofit schedules to meet the final compliance date of December 19, 2000. The emission guidelines affect 70 large MWCs and will reduce toxic air pollutant emissions by 112,000 tons per year. Table III-2 provides the nationwide emission estimates and expected percent reductions for pollutants of concern to the Great Waters. The control equipment expected to be used at a typical existing plant reduce dioxin emissions by 99 percent, mercury emissions by over 90 percent, NO_x emissions by 40 percent, and will sharply reduce other air pollutants like lead and cadmium, as shown in Table III-3.

Table III-3
Emission Reductions Expected from Control Equipment Used to Retrofit A Typical Existing MWC Plant

Pollutant	Typical Uncontrolled Level	Typical Controlled Level	Percent Reduction
Dioxin/furan (ng/dscm) total mass	1,000	3	99+
Particulate matter (mg/dscm)	3,700	4	99+
Cadmium (mg/dscm)	1.2	0.001	99+
Lead (mg/dscm)	25	0.01	99+
Mercury (mg/dscm)	0.65	0.02	90+
Sulfur dioxide (ppmv)	160	5	90+
Hydrochloric acid (ppmv)	500	10	95+
NO_x (ppmv)	225	130	40+

Source: U.S. EPA 1998h

For HMIWI, the emission guidelines and performance standards were published in the *Federal Register* in September 1997. The guidelines will apply to about 2,400 existing HMIWI; full compliance with them is no later than September 2002. In addition, EPA developed a new source performance standard (NSPS) that applies to new HMIWI that commence construction after June 20, 1996 or that commence modification after the effective date of the NSPS (i.e., 6 months after promulgation). In the first 5 years after promulgation, the NSPS are expected to apply to about 10 to 70 new HMIWI. The pollutants addressed, regulatory baseline emissions, and expected reductions of the emission guidelines and the NSPS are presented in Tables III-4 and III-5, respectively.

STATIONARY SOURCE CONTROLS ADDRESSING NO_x

The CAA provisions specifically addressing NO_x have had the greatest effect on controlling stationary source nitrogen compound emissions. Primarily because of these provisions, nationwide NO_x emissions are projected to decrease gradually for the next few years, ultimately leveling off at around 19 million metric tons per year around 2005, representing a decrease from the 1996 level of around 21.2 million metric tons/year (U.S. EPA 1998l). Nitrogen oxide emissions are projected to remain at about

that level through 2010. Figure III-1 indicates projected trends in NO_x emissions through 2010. The EPA plans to update these projections in 2000 using newer models for mobile and stationary sources. Among other benefits, these reductions will reduce rates of atmospheric nitrogen deposition affecting the Great Waters.

This section summarizes recent developments in key CAA programs that have recently or will in the near future reduce NO_x emissions from stationary sources. It lists and briefly describes, in Table III-6, CAA regulatory controls that will result in NO_x emission reductions. It also presents the sources affected, compliance dates, and the emission reductions that each regulation is expected to achieve. Finally, the section discusses the effect on NO_x emissions of possible new 8-hour ozone and $PM_{2.5}$ standards, as well as the regional haze rule.

Table III-4
Emission Reductions Expected from Existing HMIWI

Pollutant	Baseline Emissions	Nationwide Emission Reduction	Nationwide Emission Reduction (percent)[a]
Particulate matter (Mg/yr)	940	820-870	88-92
Carbon monoxide (Mg/yr)	460	340-380	75-82
Total Dioxin/Furan[b] (g/yr)	7,200	6,900-7,000	96-97
Dioxin/Furan TEQ[b] (g/yr)	148	141-143	95- 97
Hydrochloric acid (Mg/yr)	5,700	5,600	98
Sulfur dioxide (Mg/yr)	250	0-74	0-30
NO_x (Mg/yr)	1,200	0-350	0-30
Lead (Mg/yr)	11	8.6-9.4	80-87
Cadmium (Mg/yr)	1.2	0.91-1.0	75-84
Mercury (Mg/yr)	14.5	13.5-13.8	93-95

[a] These reductions represent reductions from the regulatory baseline. Percent reductions have been calculated based on the actual (unrounded) values for baseline emissions and nationwide emissions reduction.
[b] Total dioxin/furan reflects total tetra- through octa-chlorinated dibenzo-p-dioxins and dibenzofurans, as measured by EPA Reference Method 23. TEQ reflects the toxic equivalent quantity of 2,3,7,8-tetrachlorinated dibenzo-p-dioxin using international toxic equivalency factors.
Source: U.S. EPA 1998g

Table III-5
Emission Reductions Expected at New HMIWI after 5 Years of NSPS Implementation

Pollutant	Baseline Emissions	Nationwide Emission Reduction	Nationwide Emission Reduction (percent)[a]
Particulate matter (Mg/yr)	28	23-25	85-92
Carbon monoxide (Mg/yr)	14	0-7.0	0-52
Total Dioxin/Furan[b] (g/yr)	47	35-41	75-87
Dioxin/Furan TEQ[b] (g/yr)	1.1	0.80-0.93	74-87
Hydrochloric acid (Mg/yr)	64	61-62	95-98
Sulfur dioxide (Mg/yr)	28	0-15	0-52
NO_x (Mg/yr)	130	0-69	0-52
Lead (Mg/yr)	0.39	0.33-0.36	85-92
Cadmium (Mg/yr)	0.051	0.042-0.046	83-91
Mercury (Mg/yr)	0.21	0.10-0.16	45-74

[a] These reductions represent reductions from the regulatory baseline. Percent reductions have been calculated based on the actual (unrounded) values for baseline emissions and nationwide emissions reduction.
[b] Total dioxin/furan reflects total tetra- through octa-chlorinated dibenzo-p-dioxins and dibenzofurans, as measured by EPA Reference Method 23. TEQ reflects the toxic equivalent quantity of 2,3,7,8-tetrachlorinated dibenzo-p-dioxin using international toxic equivalency factors.
Source: U.S. EPA 1998j

Figure III-1
Projected National NO_x Emission Trends, 1996-2010
(U.S. EPA 1998l)

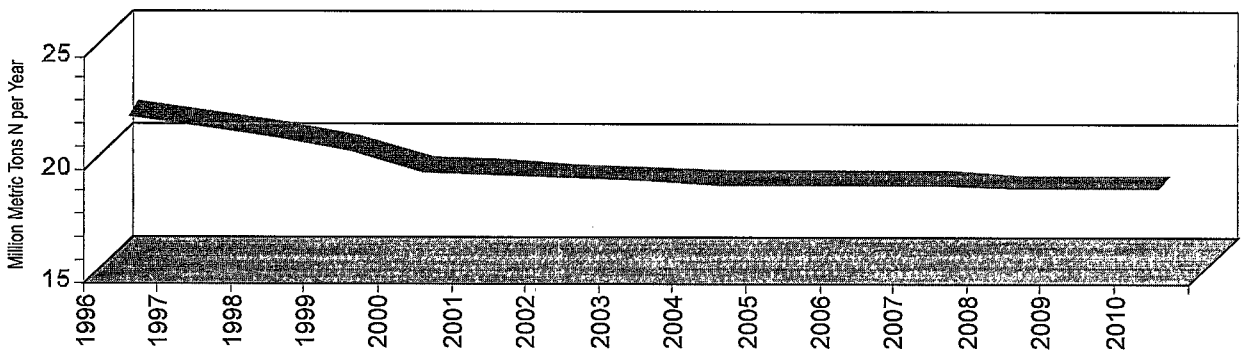


Table III-6
Recent Regulations Affecting Stationary Source NO_x Emissions

Regulation	Compliance Date	Affected Sources	NO_x or Related Requirement	Projected NO_x Emission Reductions (year by which reductions will be achieved)
NO_x RACT[1]	May 31, 1995	Boilers and combustion units (varies from State-to-State)	Varies from State-to - State	Estimated 30 to 50 percent reduction from applicable sources. [some overlap with Title IV Acid Rain reductions]
Title IV Acid Rain NO_x	Group 1 (Phase I): January 1, 1996 Group 1 (Phase II) and Group 2: January 1, 2000	Group 1: Coal-fired dry bottom wall-fired boilers, tangentially fired boilers; Group 2: wet bottom boilers, cyclones, cell burner boilers, and vertically-fired boilers (nationwide).	Group 1– Phase I: Dry-bottom wall-fired boilers: 0.50 lb/mmBtu Tangentially-fired boilers: 0.45 lb/mmBtu Group 1 -- Phase II: Dry-bottom wall-fired boilers: 0.46 lb/mmBtu Tangentially-fired boilers: 0.40 lb/mmBtu Group 2: Wet-bottom boilers: 0.84 lb/mmBtu Cyclones: 0.86 lb/mmBtu Cell burner boilers: 0.68 lb/mmBtu Vertically-fired boilers: 0.80 lb/mmBtu	2.06 million metric tons/yr (2000) [some overlap with NO_x RACT reductions]

[1] Although the NO_x RACT compliance date was over 4 years ago and the NO_x RACT was covered in the *Second Great Waters Report to Congress*, it is included here again for reference because it relates to some of the more recent regulations listed in this table.

Regulation	Compliance Date	Affected Sources	NO_x or Related Requirement	Projected NO_x Emission Reductions (year by which reductions will be achieved)
Ozone Transport Commission Memorandum of Understanding	Phase I (NO_x RACT--see above): May 31, 1995 Phase II: May 1, 1999 Phase III: May 1, 2003	Fossil fuel-fired boilers and indirect heat exchangers with a maximum rated heat input capacity of 250 mmBtu/hour or more (applies to northeast Ozone Transport Region, including Washington, DC and the 11 northeastern States)	Phase I: Varies from State-to-State (see NO_x RACT above) Phase II: Less stringent of 65 percent reduction for Inner Zone and 55 percent reduction for Outer Zone from baseline levels or 0.20 lb/mmBtu Phase III: Less stringent of 75 percent reduction (Inner Zone and Outer Zone) - from baseline levels or 0.15 lb/mmBtu Northern zone - Less stringent of 55 percent reduction from baseline levels or 0.20 lb/mmBtu	0.32 million metric tons/yr (2003)[2]

[2] Based on the difference between 1990 levels (490,000 tons) and NO_x budget for 2003 (143,000 tons).

Regulation	Compliance Date	Affected Sources	NO_x or Related Requirement	Projected NO_x Emission Reductions (year by which reductions will be achieved)
NO_x SIP Call[3]	State NO_x Budget Programs (and NO_x reductions) must be implemented by May 1, 2003; budgets to be achieved by 2007	22 States and the District of Columbia (DC) [4]	The States and DC are directed to develop regulations to meet $_{NOX}$ budgets as defined by EPA and reduce $_{NOX}$ and ozone transport. The EPA will implement these requirements under a Federal implementation plan if the States and DC do not.	1.05 million metric tons per ozone season (2007)[5]

[3] The EPA also proposed the Federal implementation plan in September 1998 in the event that any of the 22 States or the District of Columbia do not submit revised SIPs to meet the NO_x SIP call. At the same time, EPA proposed actions on petitions filed under section 126 of the CAA by eight northeastern States. The petitions requested that EPA make a finding that NO_x emissions from certain stationary sources contribute to ozone non-attainment problems in the petitioning States. The final action on the section 126 petitions was completed on April 30, 1999, but revised in December 1999 in light of rulings by the DC Circuit Court of Appeals related to the NO_x SIP call and the 8 hour ozone standard.

[4] The most likely sources to be affected include electric generation units (EGUs) including units of 25 MW or less; non-EGUs, boilers, and turbines; stationary internal combustion engines; and, cement manufacturing plants. As a result of action by the DC Circuit Court of Appeals in March 2000, WI and portions of GA and MO were removed from the areas subjected to the SIP call.

[5] Emission reductions are for the ozone season (May 1 to September 30). Includes original estimate of $_{NOX}$ reductions from EGUs, non-EGUs, mobile, and area sources from areas subjected to the SIP call. This number will be slightly less with the removal of WI and portions of GA and MO.

Regulation	Compliance Date	Affected Sources	NO_x or Related Requirement	Projected NO_x Emission Reductions (year by which reductions will be achieved)
NSPS for New Fossil Fuel-Fired Steam Generating Units (NSPS Subpart Da and Db)	November 16, 1998 (effective date)	New[6] electric utility, industrial, commercial, and institutional steam generating units	New utility boilers: 1.6 lbs/megawatt hour of energy output (gross output)[7] Modified or reconstructed existing utility boilers: 0.15 lbs/million Btu emission limit[8] New and existing industrial boilers: 0.20 lbs/million Btu	0.04 million metric tons/yr (2002)
Section 126 Petition Rule	May 1, 2003	Utilities (capacity of 25 megawatts or greater) and industrial boilers and combustion turbines (capacity of 250 million Btu/hour or greater) in 12 eastern States and the District of Columbia (NY, NJ, PA, DE, MD, DC, VA, WV, NC, KY, IN, OH, MI)	Utilities: 0.15 NO_x/ million Btu Boilers/turbines: 60 percent reduction in NO_x from controlled levels	510,000 tons of NO_x per ozone season (2007)

[6] For which construction, modification, or reconstruction commenced after July 9, 1997.

[7] This limit was changed from an input-based format (where emissions are linked to the fuel used) to an output-based format (where emissions are linked to the amount of energy generated).

[8] This emission limit was revised to be equivalent to the new unit output-based limit.

Regulation	Compliance Date	Affected Sources	NO_x or Related Requirement	Projected NO_x Emission Reductions (year by which reductions will be achieved)
Emission Guidelines for Municipal Waste Combustors	No later than December 19, 2000	Existing MWC[9] with combustion capacity > 250 tons/day of MSW per unit not specifically covered by an approved State plan	NO_x emission limits for affected facilities: Mass burn waterwall –205 ppmv Mass burn rotary waterwall – 250 ppmv Refuse-derived fuel combustor – 250 ppmv Fluidized bed combustor – 240 ppmv Mass burn refractory -- no limit	0.02 million metric tons/yr (2000)
Emission Guidelines for Existing Hospital/Medical/ Infectious Waste Incinerators (HMIWI) and NSPS for new HMIWI	Existing sources: no later than September 15, 1997 (effective date) New sources: March 15, 1998 or 6 months after start-up (effective date)	New HMIWI that commenced construction after June 20, 1996, existing HMIWI (built on or before June 20, 1996), and existing units that commence modification after March 15, 1998	NO_x emission concentrations for new and existing facilities: 250 ppmdv	New HMIWI: 0 to 69 metric tons/yr or 0 to 52 percent per year reduction. Existing HMIWI: 0 to 390 metric tons/yr or 0 to 30 percent per year reduction
Mobile Source Regulations	Tier I Tailpipe standards: 1996	Tier I Tailpipe standards: light duty vehicles and trucks	Tier I Tailpipe standards	Tier I Tailpipe standards: 850,000 metric tons of NO_x per year (2010)

[9] Constructed on or before September 20, 1994.

Regulation	Compliance Date	Affected Sources	NO_x or Related Requirement	Projected NO_x Emission Reductions (year by which reductions will be achieved)
	Tier II Gasoline Sulfur Program: 2004 for gasoline sulfur content nationwide; 2004-2009 for tighter NO_x standards for vehicles	Gasoline nationwide, and cars, light trucks, and SUVs up to 10,000 pounds gross weight sold outside California	Establishes NO_x standard of 0.07 gram per mile for all passenger vehicles , including light duty trucks, up to 10,000 pounds, phased in over several model years with interim standards more stringent than currently for light trucks, which are allowed the longest time to meet the 0.07 gram per mile standard. Averaging is allowed using a bin-based approach. These standards are comparable to California's LEV-2 program.	4.049 million metric tons of NO_x per year (2030)
	National Low Emission Vehicle (NLEV) Standards: 1999 in NE ozone transport region; 2001 nationwide	National Low Emission Vehicle (NLEV) Standards: light-duty vehicles and light light-duty trucks	NLEV Standards: Establishes the adoption of the California Low Emission Vehicle (CAL LEV) program throughout the Northeast Ozone Transport Region for the 1999 and 2000 model years, and across the entire Nation starting with the 20001 model year	NLEV Standards: 181,000 metric tons of NO_x per year (2007)

Regulation	Compliance Date	Affected Sources	NO_x or Related Requirement	Projected NO_x Emission Reductions (year by which reductions will be achieved)
	Heavy-duty highway diesel standards: 2004	Heavy-duty highway diesel standards: heavy-duty highway diesel engines	Heavy-duty highway diesel standards: 2.4 g/bhp-hr for NMHC+NO_x or, 2.5 g/bhp-hr for NMHC+NO_x with a limit of 0.5 g/bhp-hr on NMHC	Heavy-duty highway diesel standards: 1 million metric tons of NO_x per year (2020)
	Heavy-duty non-road diesel standards: 1999 - 2006	Heavy-duty non-road diesel standards: heavy-duty diesel construction, agricultural, industrial engines	Heavy-duty non-road diesel standards: 9.2 g/kW-hr (different start years depending on size of vehicle)	Heavy-duty non-road diesel standards: 1.1 million metric tons of NO_x per year (2010)
	Small spark-ignition engine standards, phase I: 1997 Small spark-ignition, non-handheld engine standards, phase II: 2001-2007	Small spark-ignition engine standards, small spark-fired engines Small spark-ignition, non-handheld engine standards	Small spark-ignition engine standards: (See 40 CFR 90.103) Small spark-ignition, non-handheld engine standards	Phase I and II: 9,000 metric tons of NO_x per year (2020)
	Locomotive engine standards: 2000	Locomotive engine standards: new and rebuilt locomotive engines	Locomotive engine standards: (See 63 FR 18982, April 16, 1998)	Locomotive engine standards: 449,000 metric tons of NO_x per year (2010)

Acid Rain Program NO_x Reduction

Title IV of the CAA requires reductions in NO_x emissions from the electric power generating industry. The acid rain NO_x requirements incorporate a two-phased strategy to reduce NO_x emissions from boilers. In the first phase, which became effective January 1, 1996, certain Group 1 boilers (i.e., dry-bottom wall-fired boilers and tangentially-fired boilers) were required to comply with specific NO_x emissions limitations.[10] All additional Group 1 boilers must comply in the second phase, which became effective on January 1, 2000. Also included in the second phase are NO_x emissions limits for all Group 2 boilers (i.e., wet-bottom wall-fired boilers, cyclones, boilers using cell-burner technology, and vertically-fired boilers).[11]

In April 1995, EPA promulgated the rule establishing NO_x emission limits for Group 1 boilers. These regulations also allowed Phase II Group 1 units to use an "Early Election" Compliance Option. Under this regulatory provision, Phase II Group 1 NO_x affected units can demonstrate compliance with the higher Phase I limits for their boiler type from 1997 through 2007 and not meet the more stringent Phase II limits until 2008. If the utility fails to meet this annual limit for the boiler during any year, the unit is subject to the more stringent Phase II limit for Group 1 boilers beginning in 2000 or the year following the exceedance, whichever is later. As a result of these rules, NO_x reductions were projected to be approximately 400,000 tons per year in 1996 through 1999 (Phase I) and 2,060,000 metric tons per year in 2000 and subsequent years (Phase II).

NO_x SIP Call, Section 126 Petitions, and Federal Implementation Plans

Many States have found it difficult to attain the ozone national ambient air quality standard (NAAQS) because of widespread regional transport (i.e., from sources in other States) of ozone and its precursors, NO_x and volatile organic compounds (VOCs). In 1995, the Ozone Transport Assessment Group (OTAG) was formed to address the regional transport problem in the eastern half of the U.S. (i.e., the 37 easternmost States). The OTAG process was a collaborative effort among 37 affected States, the District of Columbia, EPA, and interested members of the public, including environmental groups and industry representatives. The OTAG concluded that further regional reductions in NO_x emissions are needed to reduce the transport of ozone and its precursors. Furthermore, OTAG recommended in July 1997 that major sources of NO_x emissions (i.e., utility and other stationary sources) be controlled under State NO_x budgets and that an emissions trading program be developed.

In response to the OTAG recommendations, EPA issued the NO_x State implementation plan (SIP) call on October 27, 1998 (63 FR 57356). The SIP call limits summer season NO_x emissions for 22 States and the District of Columbia that are significant contributors to ozone in downwind areas. The EPA directed the 23 jurisdictions to amend their SIPs to ensure that the NO_x budgets are met. The EPA set these budgets by assessing the reductions that could be obtained through cost-effective controls on electricity generating units and large industrial boilers. However, in order to meet the SIP requirements,

[10] The affected dry-bottom wall-fired boilers must meet a limitation of 0.50 lbs of NO_x per mmBtu averaged over the year, and tangentially-fired boilers must achieve a limitation of 0.45 lbs of NO_x per mmBtu averaged over the year.

[11] The limits are 0.68 lb/mmBtu for cell burners, 0.86 lb/mmBtu for cyclones greater than 155 MWe, 0.84 lb/mmBtu for wet-bottom boilers greater than 65 MWe, and 0.80 lb/mmBtu for vertically-fired boilers.

States can adopt NO_x trading programs. These programs will be similar to the successful SO_2 trading program under EPA's Acid Rain program. The NO_x SIP call is expected to reduce atmospheric nitrogen emissions by up to 1.05 million tons per ozone season, which should reduce loadings into the Great Waters in the eastern U.S. [NOTE: In March 2000, in response to arguments made before the court, the Circuit Court of Appeals for the District of Columbia removed Wisconsin and portions of Georgia and Missouri from the list of States subject to the call. The emission reduction estimate will be slightly less with these removals.]

At the same time that EPA promulgated the NO_x SIP call rule, EPA also proposed that NO_x Federal implementation plans (FIPs) may be needed if any State fails to respond to the final NO_x SIP call. In addition, a number of northeastern States petitioned EPA, as allowed by section 126 of the CAA, to address air pollution transported from upwind States and requested that EPA make a finding that NO_x emissions from certain major stationary sources significantly contribute to ozone nonattainment problems. Such a finding would require EPA to establish Federal emissions limits for these sources. On April 30, 1999, EPA took final action on the petitions and identified upwind sources that significantly contribute to ozone nonattainment problems. In December 1999, EPA revised the April 126 petition rule in light of the rulings by the DC Circuit Court of Appeals related to the NO_x SIP call and the 8 hour ozone standard. The FIPs and the section 126 petition action would directly impose regulatory requirements on these emissions sources, including a capped, market-based trading program for certain stationary sources.

New Source Performance Standards

New source performance standards (NSPS) require emission reductions in both attainment and nonattainment areas. Section 111 of the CAA requires EPA to identify "source categories" emitting criteria air pollutants (e.g., ozone) or precursors of criteria pollutants (e.g., NO_x and VOCs) and to establish emissions limits for new, modified, and reconstructed sources of emissions.[12] Emissions limits must be based on the "best demonstrated technology," and must apply to all new sources in the country after the effective date of the rule. To date, EPA has promulgated approximately 100 NSPS, of which approximately ten directly control NO_x emissions.

In September 1998, under court order, EPA finalized an NSPS for fossil fuel-fired utility and industrial boilers. Specifically, the final standards revised the NO_x emission limits for electric utility, industrial, commercial, and institutional steam generating units for which construction, modification, or reconstruction commenced after July 9,1997. These final revised NO_x emission limits will reduce the projected growth in NO_x emissions from new sources by approximately 42 percent (41,500 metric tons/year) from levels allowed under current standards.

New Source Review and RACT

Under the CAA, States must apply similar requirements to major stationary sources of NO_x emissions as are applied to major stationary sources of VOCs because these two pollutants are precursors to ozone. These new NO_x provisions require (1) existing major stationary sources to apply reasonably available control technology (RACT) in certain ozone nonattainment areas and ozone transport regions, (2) new or modified major stationary sources to offset increased emissions and to install controls representing the lowest achievable emission rate (LAER) in areas that do not attain the ozone NAAQS

[12] Few sources emit ozone; rather it is formed in the atmosphere through the reaction of VOCs and NO_x. To attain the ozone standard, States typically require VOC and NO_x controls.

(i.e., ozone nonattainment areas) and ozone transport regions, and (3) new or modified major stationary sources to install the best available control technology (BACT) in ozone and NO_2 attainment areas.

MOBILE SOURCE PROGRAM ACTIVITIES

Collectively, mobile sources are major contributors of nitrogen compounds to the atmosphere. The EPA's Office of Mobile Sources (OMS) is responsible for regulatory oversight of air pollution emitted from mobile sources, primarily automobiles, but also including marine, aircraft, locomotive, and small engines such as lawn and garden equipment. The regulatory strategies often focus on both vehicle emissions and fuel composition.

Historically, OMS has led the effort to eliminate lead from gasoline and require more stringent tailpipe emissions and fuel changes that benefit air quality. Recent accomplishments by OMS that affect Great Waters pollutants of concern are focused primarily on nitrogen compounds. These include the following.

- Between 1994 and 1996, OMS phased in Tier I tailpipe emission standards affecting light-duty vehicles and trucks. The EPA expects the standards to reduce NO_x emissions by 850,000 metric tons per year by 2010. The Tier II tailpipe emission standards, which will further limit emissions, were proposed on May 13, 1999 and, if finalized, will reduce NO_x emissions by an additional 2.8 million tons by 2030 (see below and the *Federal Register* at 64 FR 26004, May 13, 1999).

- The national low emission vehicle, or NLEV, standard begins with model year 1999 vehicles in the Northeast Ozone Transport Region and throughout the Nation in 2001. Compliant vehicles will meet California emission standards and will reduce NO_x emissions by 181,000 metric tons per year by 2007.

- Recent regulations for heavy-duty highway diesel engines will result in one million metric tons per year reductions in NO_x emissions by 2020. Heavy-duty non-road diesel standards covering construction, agricultural, and industrial engines will be phased in between model years 1999 and 2006 and will result in reductions of 1.1 million metric tons per year of NO_x by 2010.

- New regulations covering small spark-ignition engines will reduce NO_x emissions by 9,000 metric tons per year in 2020.

- New requirements for locomotive engines, both new and rebuilt, will come into effect in 2000 and result in NO_x reductions of 449,000 metric tons per year by 2010.

Tier II Emission Standards for Vehicles and Gasoline Sulfur Standards for Refineries

In December, 1999 (65 FR 6698), EPA issued new, more protective standards for tailpipe emissions from all passenger vehicles (including sport utility vehicles (SUVs), minivans, and pick-up trucks) and new standards to reduce sulfur levels in gasoline to ensure the effectiveness of low emission-control technologies in vehicles. These new standards were in response to EPA's July 1998 Tier II Report to Congress which concluded that more stringent vehicle standards are needed to meet the ozone and particulate matter air quality standards, and that technology would be available to meet such standards cost-effectively. The EPA designed the new standards in close consultation with the auto and

oil industries, emissions control manufacturers, the States, and public health, consumer, and environmental groups (U.S. EPA 1999c).

Under the Tier II standards, SUVs, minivans, and pickup trucks are required to meet the same protective standards as passenger cars, regardless of the type of fuel used. The standards also reduce the amount of sulfur in gasoline, which will ensure the effectiveness of low emission-control technologies in vehicles and reduce harmful air pollution. When fully implemented in 2030, the new tailpipe and gasoline standards are expected to reduce NO_x emissions from vehicles by 2.8 million tons, emissions of particulate matter (i.e., soot) by 35,000 tons, and SO_2 emissions from vehicles by 334,000 tons. The significant environmental benefits of this program are expected to come at an average cost increase of less than $100 per car and less than $200 per light-duty truck. Consumers would pay less than 2 cents per gallon more for gasoline, or about $100 more over the life of an average vehicle (U.S. EPA 1999c). Additional information is available at http://www.epa.gov/otaq/tr2home.htm.

OZONE AND PM NAAQS AND THE REGIONAL HAZE RULE

Since the *Second Great Waters Report to Congress*, EPA made revisions to the particulate matter (PM) and ozone NAAQS. In addition, in April 1999, EPA issued the final regional haze rule to address visibility impairment in national parks and wilderness areas (also known as Class I areas) caused by numerous sources located over broad regions. Some of these Class I areas are associated with Great Waters waterbodies, such as Isle Royal National Park in Lake Superior and Swan Quarter National Wildlife Refuge in the Albemarle-Pamlico Estuary. Implementation of the NAAQS in conjunction with the regional haze program is anticipated to improve visibility across the country as well as reduce NO_x emissions and consequently nitrogen deposition to coastal waters, particularly in the eastern U.S. The EPA will have a better understanding of the NO_x emission reductions resulting from these programs when emissions and monitoring data are collected from the States, nonattainment areas are designated, and the States submit implementation plans (U.S. EPA 1998f).

However, on May 14, 1999, in response to a suit by the American Trucking Associations, Inc., a panel of the Circuit Court of Appeals for the District of Columbia issued a decision vacating the revised PM_{10} standard and stopping implementation of the new ozone standard. The U.S. Department of Justice has appealed this decision. The court did not, however, prevent EPA from designating nonattainment areas for the new ozone standard, and therefore EPA is considering doing so in 2000. For the new $PM_{2.5}$ standards, which the court ruled should stay in place, EPA currently plans to designate attainment and nonattainment areas as soon as air quality data are collected and analyzed, which is anticipated to be in 2004 or 2005.

OTHER NATIONAL PROGRAMS

Fish Contamination Program

The EPA's Fish Contamination Program (FCP) provides technical assistance to States, tribes, and others on matters related to persistent bioaccumulative toxics in fish and wildlife and associated potential health risks to consumers. Since 1992, the FCP has worked with State and tribal agencies to establish nationally-consistent methods and protocols for assessing contaminants in fish and wildlife for the purpose of developing and managing consumption advisories. Additional activities of the FCP include publishing guidance documents, maintaining national databases (e.g., of fish consumption advisories), sponsoring conferences and training workshops, providing grants for advisory development

and special studies, developing outreach materials, and assisting States and tribes in the issuance of consumption advisories.

Since 1993, the FCP has published an annual report on trends in the number of fish and wildlife consumption advisories. *The National Listing of Fish and Wildlife Advisories* (NLFWA) identifies all State-, tribal-, and Federally-issued fish consumption advisories in the U.S. Recently, it has been expanded to include Canadian provinces and territories. According to the 1998 NLFWA, the number of consumption advisories in the U.S. rose by 125 in 1997 to a total of 2,299, a 5 percent increase from 1996. The number of waterbodies under advisory in 1997 represented 16.5 percent of the Nation's total lake acres and 8.2 percent of the Nation's total river miles. The total number of advisories in the U.S. increased for three major pollutants – mercury, dioxin, and DDT. The increase in advisories issued by the States generally reflects an increase in the number of assessments of the levels of chemical contaminants in fish and wildlife tissues, rather than an increase in contaminant levels (U.S. EPA 1998m).

Environmental Justice Initiatives

Research indicates that people of different racial and ethnic backgrounds and income levels often do not eat the same kinds and amounts of food (U.S. EPA 1995). For example, Native Americans and the urban poor are at a greater risk for adverse health effects due to high rates of consumption of potentially contaminated fish. Fetuses and young children are at risk because they are more vulnerable to the effects of the pollutants of concern. Thus, these subpopulations may be disproportionately affected by deposition of air pollutants to the Great Waters.

The EPA recognizes the relationship between health risks, environmental pollutants, and diet as a potential environmental justice issue. Since 1992, EPA's Office of Environmental Justice has served as the point of contact for environmental justice outreach and educational activities, has provided technical and financial assistance, and has disseminated environmental justice information. In conjunction with regional and headquarters offices, this office has initiated many programs to address the environmental concerns among minority, low-income, and Native American and Alaska Native communities (U.S. EPA 1995). Likewise, EPA created the American Indian Environmental Office in 1994 for the purpose of coordinating the EPA-wide effort to strengthen public health and environmental protection on Native American lands (U.S. EPA 1998u). An example of EPA's efforts is the passage of a resolution by the National Environmental Justice Advisory Council of EPA's Office of Environmental Justice in December 1998 that was developed by the Indigenous People Subcommittee pertaining to the effects of mercury contamination on American Indian populations. This resolution requires EPA's Office of Pollution Prevention and Toxic Substances to share the 1998 Mercury Action Plan with tribes, to provide educational and health information to tribes, to adopt a Mercury Action Plan and regulatory authority to eliminate anthropogenic mercury emissions by 2010, to establish baseline emission standards, and to adopt enhanced reporting requirements for mercury emission sources.

Recent studies continue to examine the relationship between increased health risks in certain subpopulations and the consumption of fish from the Great Lakes. Study results show that some subpopulations are not as aware of fish advisories as other populations, and that human health effects from consumption of fish from contaminated areas vary. Chapter II describes additional relevant research relating to exposure and effects of Great Waters Pollutants of concern and sensitive or highly-exposed subpopulations.

Children's Health Initiatives

Children face environmental health threats from many of the Great Waters pollutants of concern. In addition, child exposures to pollutants tend to occur through multiple exposure routes, including inhalation, ingestion, dermal contact, and prenatal (transplacental) exposure. For example:

- Prenatal and childhood exposure to contaminants, such as lead, PCBs, and mercury, via multiple exposure pathways may inhibit a child's intellectual development and ultimately may result in behavioral problems.
- Exposure to endocrine disrupting chemicals, such as organochlorine pesticides and PCBs, may cause birth defects and alterations of normal childhood growth and development (Browner 1998, U.S. EPA 1998w).

Children Experience Increased Risk From Environmental Hazards Because:

Their systems are still developing making them more susceptible to environmental threats.

They eat more food, drink more fluids, and breathe more air per pound of body weight making them more exposed to environmental hazards.

Their behavior, such as crawling on the ground, and lack of ability to protect themselves exposes them to hazards that adults can easily avoid.

In an effort to protect children from environmental health threats, EPA published its *National Agenda to Protect Children's Health from Environmental Threats* in April 1996. This agenda calls for the consideration of children's risks in all appropriate agency actions and a greater emphasis on research to support children's risk assessment activities (U.S. EPA 1996a). In addition, EPA established its Office of Children's Health Protection (OCHP) in May 1997 to ensure the implementation of the President's 1997 Executive Order to Protect Children from Environmental Health and Safety Threats. The OCHP's mission is to make the protection of children's health a fundamental goal of public health and environmental protection in the U.S. The office supports and facilitates EPA efforts to protect children from environmental threats (U.S. EPA 1998t).

The President's Executive Order requires all Federal agencies to address health and safety risks to children, coordinate research priorities on children's health, and ensure that their standards take into account special risks to children. The EPA documents its current actions in regard to children's health in *The EPA Children's Environmental Health Yearbook* (U.S. EPA 1998q). The yearbook includes sections on asthma and respiratory effects, childhood cancer, developmental and neurological toxicity, health effects of pesticides, and potential risks from contaminated surface and ground water. To coordinate research efforts, EPA and the National Institute of Environmental Health Services developed a grant program to support the establishment of Centers for Children's Environmental Health and Disease Prevention Research. The purpose of these centers is to foster the advancement of children's health through enhancing the public's understanding of basic disease mechanisms and promoting community-based prevention activities related to children's respiratory disorders, childhood learning, and growth and development (U.S. EPA 1998t).

The EPA, in coordination with other Federal agencies, has begun several efforts to address these specific threats. Most notably, EPA has conducted an Agencywide Risk Assessment Forum colloquium on children's risk and has begun to review and revise several of its risk assessment guidance documents to identify areas where children's health protection is or should be considered. Mercury, lead, dioxin, HCB, and PAHs are among the chemicals included in this risk characterization (Browner 1998). As part of this effort, EPA requested that the Federal Children's Health Protection Advisory Committee (CHPAC) recommend existing standards that may merit reevaluation in order to further protect children's environmental health. One recommendation was to reevaluate the chlor-alkali NESHAP

(mercury). In response, EPA has begun a process to revise this standard, including a risk assessment of mercury emissions from chlor-alkali plants (64 FR 5277, February 3, 1999). Also, EPA, the Department of Health and Human Services, and other Federal agencies have begun to develop a comprehensive cross-government strategic plan to address the causes of children's asthma and the scope of the problem (Browner 1998).

III.B REGIONAL AND WATERBODY-SPECIFIC PROGRAMS

All of the Great Waters are affected by the policies and activities of multiple communities and governments on their shores. The Great Lakes, for example, are affected by the environmental management decisions of two nations, one Canadian Province, eight States, a number of tribes, and countless municipalities. Intergovernmental or multistakeholder institutions (e.g., the Lake Michigan Forum) have been established for many of the Great Waters to coordinate resource management decision making and resolve conflicts. In addition, EPA and NOAA, as directed by Congress, administer several programs to address particular regional and waterbody-specific environmental challenges. These programs lead or support many efforts to evaluate or control the impacts of pollution, including pollution via atmospheric deposition, on the Great Waters ecosystems.

The EPA has found that regional environmental challenges, such as those facing the Great Waters, are often best solved through collaboration with local stakeholders and with a holistic approach that addresses human social and economic needs as they relate to environmental quality. The EPA has used these approaches in a number of place-based (i.e., geographically-based) programs, including the National Estuary Program; Great Lakes, Chesapeake Bay, and Clean Lakes Programs; and, the Regional Geographic Initiative. These approaches are further developed in EPA's Community-Based Environmental Protection (CBEP) program. The EPA's Strategic Plan (EPA 1997c) recognized CBEP as the Agency's main tenet for "reinventing" its approach to environmental protection by considering environmental problems across organizational and political boundaries and in a multimedia fashion. The Agency is now using the CBEP approach in several of the regional and waterbody-specific programs and activities described in this section.

New York Sea Grant Extension, Pat MacNeill

GREAT LAKES PROGRAM

Administered by EPA's Great Lakes National Program Office (GLNPO), the Great Lakes Program consists of programs and activities initiated by EPA, States, tribes, and their partners that are designed to address challenges facing the Great Lakes ecosystem. Several of these activities involve atmospheric deposition and Great Waters pollutants of concern.

The GLNPO has provided funds for monitoring of toxics in conjunction with the Episodic Events/Great Lakes Experiment (EEGLE) Study research effort. The EEGLE Study is being funded by the National Science Foundation and NOAA to study nutrient transport in a plume that occurs in Lake Michigan annually. This effort enables the study of air-water exchange of toxics in this plume. This information will be used in support of the Lake Michigan Mass Balance Study (LMMBS) by providing insight into the air/water exchange of PCBs and PAHs. The project will also provide information necessary to determine the spatial and temporal variation of loadings across large lakes. This project is a pilot for future air/water toxics sampling projects, such as planned additional over-water measurements for the Integrated Atmospheric Deposition Network (IADN) program.

In addition to these activities, the Great Lakes Program is continuing to utilize remedial action plans (RAPs) for areas of concern (AOCs) and lakewide management plans (LaMPs) to target ecological problems on a geographic basis, in accordance with the 1978 Great Lakes Water Quality Agreement (GLWQA) between Canada and the U.S. The LaMPs and RAPs are tools for reducing the input of pollutants to the Great Lakes and restoring the environmental quality of the Great Lakes basin.

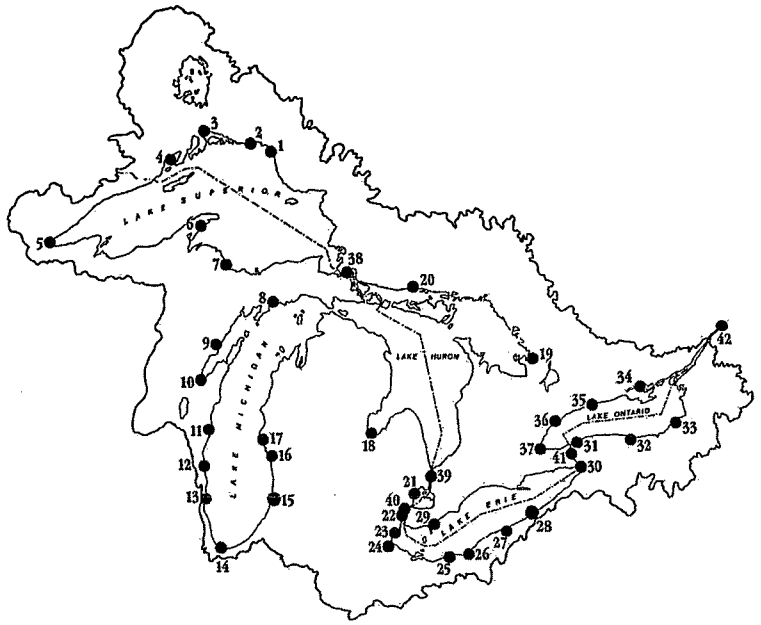
The LaMPs and RAPs target ecological problems on a geographic basis and provide a community-based approach to identifying and solving environmental problems. Both tools were originated in response to the GLWQA goals of restoring and maintaining the chemical, physical, and biological integrity of aquatic ecosystems. The RAPs were first established in 1985 to provide more uniform guidance on how to restore uses in AOCs. Rivers, connecting channels, harbors, and embayments of the Great Lakes are designated as AOCs if there is an impairment of beneficial use or the area's ability to support aquatic life. Unlike RAPs, the development and implementation of LaMPs for each of the five Great Lakes was a specific objective of the GLWQA. The LaMPs are frequently integrated with RAPs and other efforts that are best suited to address issues of local concern.

IJC Identified Seven AOCs That Have Developed Particularly Successful Remediation Strategies

- √ Black River (Ohio)
- √ Grand Calmumet River/Indiana Harbor Ship Canal
- √ Hamilton Harbor (Ontario)
- √ Ashtabula River (Ohio)
- √ Bay of Quinte (Ontario)
- √ Manistique River (Michigan)
- √ Muskegon and White Lakes (Michigan)

The Great Lakes Water Quality Board of the International Joint Commission (IJC) established 42 AOCs in the Great Lakes basin (Figure III-2): 26 within the jurisdiction of the U.S., 12 within Canadian jurisdiction, and 5 shared by both countries. The RAPs are being developed for each of these AOCs to address impairments to any one of the 14 beneficial uses (e.g., restrictions on fish and wildlife consumption, dredging activities, or drinking water consumption) associated with these areas. The RAPs are prepared and implemented by the eight Great Lakes States and the Province of Ontario, with help from Federal agencies and organizations, local governments, industry, environmental groups, and individuals. Although there has been significant progress in developing and implementing most RAPs (including the delisting of the Collingwood Harbor AOC in Canada), considerable challenges remain.

Figure III-2
The 42 Areas of Concern in the Great Lakes Basin



Lake Superior

1. Peninsula Harbour
2. Jackfish Bay
3. Nipigon Bay
4. Thunder Bay
5. St. Louis Bay/R.
6. Torch Lake
7. Deer Lake-Carp Creek/R.

Lake Michigan

8. Manistique River
9. Menominee River
10. Fox River/Southern Green Bay
11. Sheboygan River
12. Milwaukee Estuary
13. Waukegan Harbor
14. Grand Calumet River/Indiana Harbor Canal
15. Kalamazoo River
16. Muskegon Lake
17. White Lake

Lake Huron

18. Saginaw River/Saginaw Bay
19. Severn Sound
20. Spanish River

Lake Erie

21. Clinton River
22. Rouge River
23. River Raisin
24. Maumee River
25. Black River
26. Cuyahoga River
27. Ashtabula River
28. Presque Isle bay
29. Wheatley Harbour

Lake Ontario

30. Buffalo River
31. Eighteen Mile Creek
32. Rochester Embayment
33. Oswego River
34. Bay of Quinte
35. Port Hope
36. Metro Toronto
37. Hamilton Harbour

Connecting Channels

38. St. Marys River
39. St. Clair River
40. Detroit River
41. Niagara River
42. St. Lawrence River (Cornwall/Massena)

Source: Adapted from Hartig et al. 1997

One of the major problems facing the AOCs today is toxic contamination of sediments, contributing to beneficial use impairments.

Both the U.S. and Canadian governments are charged with developing LaMPs for each of the Great Lakes, with the exception of Lake Michigan. Because Lake Michigan lies entirely within the boundaries of the U.S., the Lake Michigan LaMP was developed solely by the U.S. government. The LaMPs are in various stages of development for each of the Great Lakes (see sidebar). Not all of the LaMPs have been completed; however, commitments have been made by key stakeholders in the respective basins to pursue toxics reductions and actions are being taken to achieve these goals. Each LaMP addresses a different list of critical pollutants, commonly including mercury, PCBs, hexachlorobenzene, dioxins, furans, chlordane, DDT and metabolites, and dieldrin, all of which are Great Waters pollutants of concern.

Current Status of LaMPs in the Great Lakes

Superior	Binational Program to Restore and Protect the Lake Superior Basin announced (1991) Stage 1 LaMP submitted to IJC (1995) Stage 2 LaMP released (1999) Stage 3 LaMP in development
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) Stage 1 LaMP released (1998)

The Lake Superior LaMP is unique in that it is being developed in stages. The Stage 1 LaMP was submitted to the IJC in 1995. The Stage 2 LaMP, which addresses critical pollutants, is available on EPA's web site at www.epa.gov/grtlakes/lakesuperior/stage2lamp.html. The Stage 3 LaMP, which is currently in development and is available as a review draft on EPA's web site at www.epa.gov/grtlakes/lakesuperior/stage3/review.html, addresses selection of remedial measures and management strategies to achieve critical pollutant load reduction targets.

LAKE CHAMPLAIN BASIN PROGRAM

Since the *Second Report to Congress*, the Lake Champlain Basin Program (LCBP) has continued to develop and implement a comprehensive pollution prevention and restoration plan for the lake and its watershed, as called for by the Lake Champlain Special Designation Act of 1990. In October 1996, the LCBP finalized *Opportunities for Action, An Evolving Plan for the Future of the Lake Champlain Basin* (LCBP 1996a, b). The final plan differs little from the draft plan, which was described in detail in the *Second Report to Congress*. Environmental issues addressed by the plan include high phosphorus levels, toxic substances (most notably PCBs and mercury) in biota and sediment, and invasive non-native species. Atmospheric deposition of mercury to Lake Champlain basin is the subject of research efforts described in Chapter II.

The LCBP supported and published several technical reports on the Lake Champlain basin and the Lake Champlain ecosystem. For example, in October 1997, the LCBP published Phase II of the Lake Champlain Sediment Toxics Assessment Program (McIntosh et al. 1997). The first phase, which was discussed in the *Second Report to Congress*, accomplished a lakewide screening of surface sediments for an array of organic and inorganic trace contaminants, more intensive evaluations at nine sites with elevated contaminants levels, and an assessment of PCB bioaccumulation from sediment by the macroinvertebrate *Mysis relicta*. Phase II further targeted investigations to the three most contaminated

areas of the lake: Cumberland Bay, Malletts Bay, and Burlington Harbor. Results of the Phase II investigation are presented in Chapter II above.

Other recent research projects sponsored by LCBP include development and compilation of Geographic Information Systems data for the basin (VCGI 1996, Millette 1997); hydrodynamic and water quality modeling and monitoring (ASA 1996, Lake Champlain Basin Program 1998); food web modeling (LeBar and Parrish 1996); and, other ecological subjects. Numerous other LCBP-supported publications address economic, educational, recreational, and other resource management subjects. Future research will focus on environmental indicators. In addition, the Lake Champlain Steering Committee, which evolved from the Management Conference, includes the Province of Quebec as a member. Involvement of Quebec will ensure that both U.S. and Canadian concerns are addressed.

CHESAPEAKE BAY PROGRAM

Chesapeake Bay Program Partners

- State of Maryland
- Commonwealth of Virginia
- Commonwealth of Pennsylvania
- District of Columbia
- Chesapeake Bay Commission
- U.S. EPA (representing all Federal agencies, e.g., NOAA, U.S. FWS)

The Chesapeake Bay Program (CBP) is a unique regional partnership (see sidebar) that has been responsible for directing and implementing the restoration of the Chesapeake Bay since 1983. Since that time, the highest priority has been placed on restoring the living resources of the bay, including finfish, shellfish, bay grasses, and other aquatic life and wildlife. Examples of specific actions undertaken by the CBP include agricultural best management practices, pesticide collection and disposal programs, public education, Biological Nutrient Removal at wastewater treatment facilities, and a phosphate detergent ban (Chesapeake Bay Program 1998d). In addition, the CBP is working with NOAA's Chesapeake Bay Environmental Effects Committee which supports research on contaminated sediment to better understand issues related to the management of contaminated sediments.

As discussed in the *Second Report to Congress*, the 1994 Chesapeake Bay Basinwide Toxics Reduction and Prevention Strategy is an integral part of the CBP. The primary goal of the strategy is a "Chesapeake Bay free of toxics by reducing or eliminating the input of chemical contaminants from all controllable sources to levels that result in no toxic or bioaccumulative impact on the living resources that inhabit the bay or human health." The strategy contains commitments in the following five areas: (1) regional focus – calls for assessing the status of chemical contaminant effects on the living resources of the bay and its tidal waters and implementing reduction and prevention activities in those areas; (2) directed toxic assessments – calls for the characterization of chemical contaminant conditions in the bay, the assessment of low level toxics exposure to living resources as well as the update of the Toxics Loading and Release Inventory to identify toxics sources; (3) regulatory program integration – calls for Chesapeake Bay Program activities to complement and enhance Federal, State, and local regulatory programs; (4) pollution prevention – includes facility-based pollution prevention, pesticide management, and consumer/household hazardous waste activities and goals; and, (5) strategy implementation – outlines how the strategy will be implemented.

The strategy addresses non-point source pollution, committing the CBP signatories to "establish more complete loadings baselines and source identification for storm water runoff, atmospheric deposition, and acid mine drainage, and set reduction targets from that baseline to be achieved over the next decade." The CBP will use the updated *1999 Chesapeake Bay Basinwide Toxics Loading and Release Inventory*, which provides updated chemical contaminant loadings estimates for atmospheric

Reducing Pesticide Use in the Chesapeake Bay Watershed

The CBP has been working toward goals to reduce pesticide use in the Chesapeake Bay watershed. Recent accomplishments include the following:

✔ Pesticide collection and disposal programs have been offered in all Virginia and Pennsylvania counties and 75 percent of Maryland counties in the watershed. Over 1.1 million pounds of pesticides have been collected;

✔ Between 1993 and 1998, nearly 600,000 pesticide containers have been collected and recycled.

✔ Integrated Pest Management is now used on nearly 4.4 million acres (61 percent) of agricultural cropland in the watershed.

deposition and other point and non-point sources to address this commitment. The inventory reports atmospheric deposition loads from chemical contaminants in the air that are deposited onto the bay and its tidal rivers. These estimates are updated and expanded using recent field measurements and improved theoretical understanding of deposition processes. Volatilization of organic contaminants from the surface waters to the air is considered for the first time in calculating a "net" atmospheric loading to the bay and tidal rivers. Initial estimates of the contribution of urban areas to atmospheric deposition loads to the bay and tidal rivers are also reported. Only loads to tidal waters (below the fall line) are reported. The TRI database for industrial air releases was not included in this inventory, as it was in 1994, since the improved and expanded atmospheric loadings data (to tidal waters) are based on measured data and are a much better representation of loads than the TRI data estimates of releases. The inventory reports that atmospheric deposition loads to the tidal waters increase in areas of the bay and tidal rivers adjacent to urban areas (Chesapeake Bay Program 1999a).

To focus toxic reduction and prevention efforts, the CBP developed a list of Chesapeake Bay toxics of concern (i.e., chemicals that cause or have a potential to cause adverse impacts on the bay system, such as mercury, PAHs, and PCBs -- see sidebar). By 2000, the CBP is directed to reevaluate and revise the 1994 toxics strategy. Future plans for the Chesapeake Bay Program include research in support of regional action plans for areas with known toxics problems, with a particular emphasis on how to deal with contaminated sediment. In addition, data collected over the past decade will continue to be analyzed to determine which chemicals have been detected in water, sediment, shellfish, and finfish to identify other toxics problems in the bay (Chesapeake Bay Program 1998c).

Chesapeake Bay Program Toxics of Concern

Atrazine	Chromium
Benz[a]anthracene (PAH)*	Chrysene (PAH)*
Benzo[a]pyrene (PAH)*	Copper
Cadmium	Fluoranthene (PAH)*
Chlordane*	Lead*
Naphthalene (PAH)*	Mercury*
Tributyltin	PCBs*

* Great Waters pollutants of concern

Nitrogen reduction in the bay is an ongoing focus of the CBP. Recent modeling efforts indicate that approximately 21 percent of the nitrogen entering the bay is from atmospheric deposition. Therefore, the CBP is working to quantify and address atmospheric nitrogen and toxics emissions and sources along with their associated impacts on the bay resources. A current effort involves assessing the benefits that will be experienced due to the implementation of the CAA. The CBP is also supporting scientific research which is being conducted to better understand the integrated, multimedia relationships of the ecosystem. In addition, the Chesapeake Bay Program is developing a strategy to better understand and quantify the various forms of nitrogen which may be affecting living resources and water quality in the Chesapeake Bay. Part of an integrated basinwide monitoring effort, this strategy will help to fill in gaps in our knowledge of atmospheric

deposition of nitrogen compounds, focusing in the near term on measuring deposition of ammonia and ammonium in the coastal areas (Chesapeake Bay Program 1998a).

In addition, recent actions taken under the Clean Water Act resulted in listing portions of the Chesapeake Bay and its tidal rivers as impaired waters. These actions have emphasized the regulatory framework of the Clean Water Act along with the ongoing cooperative efforts of the Bay Program as the means to address the nutrient enrichment problems within the Bay and its rivers. In response, the Bay Program partners have committed to a process for integrating the cooperative and statutory programs of the Chesapeake Bay and its tributaries. In the new Chesapeake Bay Agreement, the partners are committed to developing goals for improving water quality in the Bay and its tributaries so these waters may be removed from the impaired waters list prior to the timeframe when regulatory mechanisms under section 303(d) of the Clean Water Act would need to be applied.

The CBP is helping bay partners to incorporate air pollution impacts in the management of lakes, rivers, and streams. For example, the CBP is encouraging States to account for air deposition in TMDL development (for background information on TMDLs, see the TMDL discussion on page III-5) and helping bay States account for atmospheric deposition of nitrogen compounds in developing tributary strategies to protect the bay. Tributary strategies are "clean-up plans" for each major river that flows into the bay. The Commonwealth of Virginia is developing tributary strategies for their southernmost bay tributaries, and the CBP is providing modeled information on how different management scenarios for atmospheric nitrogen emissions will affect deposition loads. This will give States an idea of different options for cleaning up lakes, rivers, and streams. For example, an understanding of how much nitrogen will not enter the bay by implementing certain air controls will allow States to count the cost of all of the options of reducing nitrogen inputs. In comparing methods of nutrient reduction in waters in the Chesapeake Bay area, it may be that cleaning up the air is more cost-effective than some water-based controls, such as additional storm water management in cities.

Despite the progress made to date in reducing inputs of nitrogen to the Chesapeake Bay, the Chesapeake Executive Council announced that unless current efforts are accelerated, the nitrogen reduction goal of the Chesapeake Bay Agreement will not be met by the year 2000. In 1997, the Executive Council developed three new directives to accelerate the reduction of nitrogen inputs to the bay.

1. The *Baywide Nutrient Reduction Progress and Future Reductions* directive outlines a series of actions aimed to further commitments made in the Chesapeake Bay Agreement. One of the actions is to "Work toward additional reductions of airborne nitrogen delivered to the Bay and its watershed from all sources including States outside the watershed, and seek improved understanding of how airborne nitrogen affects the Bay and its watershed.." The directive includes a time line for completing refinements of computer modeling as well as water quality monitoring. Outputs from monitoring and modeling efforts will be used to help

Growing Attention to Sources of Ammonia and Urea to the Chesapeake Bay

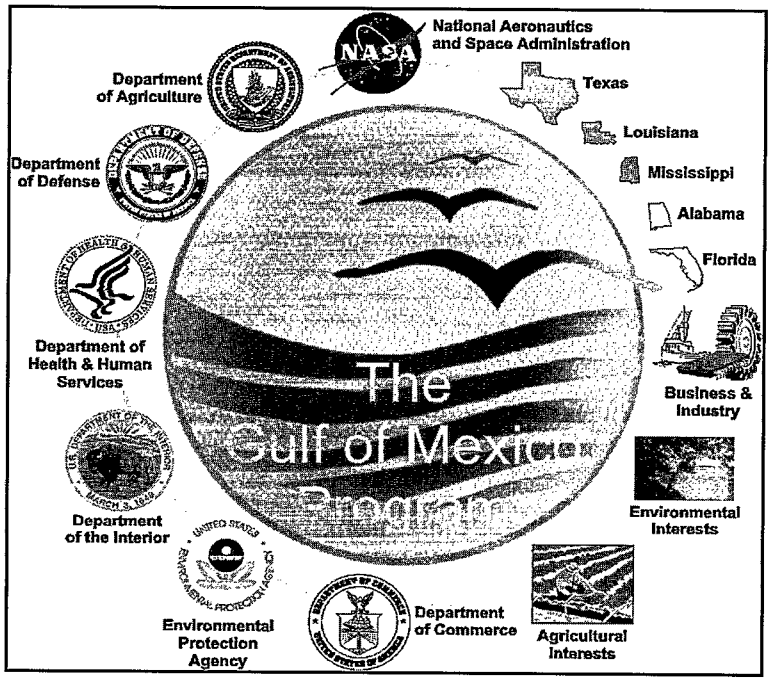
Ammonia and urea are other forms of nitrogen that are receiving increased attention from researchers and regulatory agencies, in part because these forms are more biologically available. One of the sources of ammonia and urea is manure from animal farming operations. With the increase in animal farming in the bay watershed and surrounding States, particularly hog and poultry farming, it is important to investigate pollutant emissions to the air and the distances they travel in the air before being deposited to land or water surfaces. The CBP sponsored a workshop on atmospheric organic nitrogen (e.g., urea) and is coordinating with NOAA to determine atmospheric concentrations of ammonia, estimate ammonia deposition to land and water surfaces, and evaluate the importance of ammonia transport and deposition.

set nutrient goals for Virginia tributaries and to develop a protocol to determine whether nutrient reduction efforts can be further targeted to areas of persistent high loadings.

2. The *Wetlands Protection and Restoration Goals* directive provides quantifiable wetland restoration goals to assure no net loss of wetlands and to move in the direction of a net gain of wetland areas.

3. The *Community Watershed Initiative* will develop a community watershed strategy to ensure that Chesapeake Bay Program goals and objectives are integrated at the community watershed scale (Chesapeake Bay Program 1998b).

GULF OF MEXICO PROGRAM



The Gulf of Mexico Program emphasizes community-based, ecosystem management approaches to environmental protection, including (1) equal partnership among government agencies and private and non-government interests to define problems and implement solutions, (2) use of the best science and knowledge available to support decisions and guide actions, and (3) public involvement in all phases of the program to generate the consensus needed for action. The Gulf of Mexico Program is not a regulatory program, although some of the partner agencies at the Federal and State levels have regulatory responsibilities. The program provides a forum whereby issues that cross political or social boundaries can be clearly identified, discussed, and collaboratively resolved to benefit the ecological and economic resources of the Gulf of Mexico.

Given the vast geographic scope of the gulf, protection of these critical resources requires a long-term commitment and focused attention. A strategic assessment process is being implemented to focus future efforts, identify resources at greatest risk, and establish quantitative goals to measure progress. Currently, the Gulf of Mexico Program is addressing four priority environmental concerns, two of which are relevant to the Great Waters program: (1) protecting the public from contaminated shellfish and recreational waters, and (2) excessive nutrient enrichment.

Excessive nutrient enrichment is attributable to a multitude of terrestrial and atmospheric sources throughout the Gulf States and the watersheds (e.g., the Mississippi River basin) that drain into the gulf. The Gulf of Mexico Program, as a multiagency effort, is working with State and community partners on several projects to protect the gulf from the deleterious effects of nutrient enrichment.

For example, the Gulf of Mexico Program is working with the Gulf States to address nutrient enrichment problems in the gulf, such as a zone of hypoxia along the Louisiana coast. Hypoxia in the northern gulf represents one of the largest zones of oxygen-deficient bottom waters in the western Atlantic Ocean. Nitrate and other nutrients discharged from the Mississippi River are the probable cause, making agricultural and municipal runoff and atmospheric deposition potential sources to investigate.

In addition, the Gulf of Mexico Program is supporting two initiatives of a multiagency scientific team established by the White House's Committee on Environment and Natural Resources (CENR). In particular, the Gulf of Mexico Program is supporting studies to characterize the ecological and economic consequences of hypoxia in the gulf and nutrient sources and loads to the gulf from the Mississippi River. Further discussion of the CENR process can be found in Chapter IV of this report.

NATIONAL ESTUARY PROGRAM

Coastal waters addressed by the Great Waters program include all estuaries covered by the National Estuary Program (NEP). In 1987, Congress established the NEP as part of the Clean Water Act. The NEP's mission is to protect and restore the health of the estuaries while supporting economic and recreational activities. The EPA periodically calls for nominations of estuaries to the NEP from State governors. If an estuary meets the Agency's criteria, EPA may then designate it as an estuary of national significance. As depicted in Figure I-2, there are currently 28 estuaries around the country and in Puerto Rico in the National Estuary Program.

NEPs Conducting Atmospheric Deposition Studies

- Albemarle-Pamlico Estuary (NC)
- Casco Bay (ME)
- Charlotte Harbor (FL)
- Coastal Bend Bay and Estuary (TX)
- Delaware Inland Bays (DE)
- Indian River Lagoon (FL)
- Long Island Sound (NY, CT)
- Massachusetts Bay (MA)
- Maryland Coastal Bays (MD)
- Mobile Bay (AL)
- New York/New Jersey Harbor (NY, NJ)
- Peconic Bay (NY)
- San Francisco Bay (CA)
- Santa Monica Bay (CA)
- Sarasota Estuary (FL)
- Tampa Bay (FL)

To date, at least 19 NEPs have identified atmospheric deposition of pollutants as a threat to the health of their estuaries. Many of these NEPs either have initiated studies on the contribution of atmospheric deposition to annual loadings of nitrogen and/or other pollutants, or have expressed serious interest to EPA in conducting such projects. In 1999, EPA provided funds to establish new atmospheric deposition monitoring sites in five NEPs, expanding the National Atmospheric Deposition Network in the coastal waters and improving the ability to compare coastal data to data collected from inland sites. Peconic Bay NEP and Maryland Coastal Bays NEP are monitoring for nitrogen and sulfur compounds, San Francisco Bay is monitoring for mercury compounds, and Mobile Bay NEP is monitoring for sulfur, nitrogen, and mercury compounds. In addition, a site measuring dry deposition of sulfur and nitrogen compounds (part of the CASTNet monitoring network) is being established near Indian River Lagoon NEP. The following describes other NEP sites and their associated atmospheric deposition research activities to date.

- **Albemarle-Pamlico Estuary (NC).** Nitrogen deposition studies in eastern North Carolina are primarily focusing on the emissions and deposition of ammonia. Concern has been spurred by the explosive growth of large-scale hog farming operations in the coastal plain over the last few years. For example, long-term analysis of National Atmospheric Deposition Program (NADP) data from a site near the center of an intensive animal operations (i.e., Sampson County, NC) indicate at least a doubling of NH_4^+ deposition since the early 1980s (Paerl 1997b). Monitoring efforts led by researchers from the University of North Carolina at Chapel Hill (UNC-CH) and

North Carolina State University (NCSU) are aimed at quantifying atmospheric levels of both gas and aerosol forms of ammonia to aid in the development of regional-scale, air quality models (Robin Dennis, NOAA/EPA). Monitoring began in fall 1998, but data obtained in the 1999 summer season will be critical in understanding seasonal emission and deposition patterns. Additional efforts are aimed at developing nitrogen budgets for the Neuse River basin. These projects began with State funding and, when completed, will put the atmospheric contribution into the context of the overall nitrogen load to the Neuse River basin (W. Robarge, NCSU; H. Paerl, UNC-CH). The EPA is also funding research to examine the biological ramifications (e.g., eutrophication) of atmospheric nitrogen inputs in the Neuse River basin and the adjacent coastal waters.

- **Casco Bay (ME).** The Casco Bay Air Toxics Deposition Study, begun in 1998, is a multiyear collaborative effort by the Casco Bay Estuary Project, the Maine Department of Environmental Protection, EPA Region I, and university research scientists (University of Massachusetts, Lowell). The study focuses on atmospheric deposition of five contaminant groups (i.e., mercury, toxic trace elements, PAHs, nitrogen, and fine particulates) and is funded by the Great Waters program as part of the national strategy to determine the environmental health and status of key NEP ecosystems. The objectives of the study are to characterize seasonal and annual depositional patterns of toxic air compounds to Casco Bay and to develop a generic assessment method that can be used by other community-based programs.

- **Charlotte Harbor (FL).** The Charlotte Harbor NEP atmospheric deposition study received funding in 1999 and will begin activities in 2000.

- **Coastal Bend Bay and Estuary Program (TX).** The concentrations of nutrients and organic contaminants (including PAHs, PCBs, and some pesticides) in wet and dry atmospheric deposition is being measured or calculated at two representative sites on Corpus Christi Bay. An EPA grant expanded the pollutants measured at one station to include organic contaminants. The wet nutrient deposition data are comparable to other air monitoring programs, including the NADP, but this is one of the only NEP studies that measures the deposition rate of organic contaminants. This study is being conducted in conjunction with other studies in the area (including EPA's Environmental Monitoring and Assessment Program (EMAP) and NOAA's National Status and Trends Program) to measure the inputs to the bay and estuary of organic pollutants, trace metals, and nutrients from other sources (e.g., other non-point sources, point sources). This will allow the Coastal Bend Bay and Estuary Program to calculate the importance of atmospheric deposition for each pollutant and target control measures where they are most effective.

- **Delaware Inland Bays (DE).** The University of Delaware Graduate College of Marine Studies is undertaking three studies to address atmospheric deposition issues related to the Delaware Inland Bays. The first study, currently in progress and funded by the Delaware Department of Natural Resources and Environmental Conservation (DNREC), has two primary objectives: (1) to accurately quantify the atmospheric loading of nitrogen to the Delaware Inland Bays; and (2) to assess, in cooperation with the University of Delaware Center for Climatic Studies, the meteorological transport patterns which contribute to the observed nitrogen deposition. The second study, also funded by the DNREC, will examine the episodic impact of large precipitation events on the loading of nitrogen to the Delaware Inland Bays by both direct (deposition to the water surface) and indirect (via watershed transmission) pathways.

The third study, which is funded by the EPA National Estuary Program, will address the impacts of local sources (e.g., a coal-fired power plant, poultry-rearing facilities) on nitrogen deposition to the Delaware Inland Bays and distinguish the impacts of local sources from the impacts of regional sources. Research to date indicates a 60 percent increase in the wet deposition of ammonia over the past two decades. Although the explanation is uncertain, the working hypothesis is that the increase is related to the large increase in poultry production on the Delmarva Peninsula. The NEP grant will test this hypothesis. An analogous situation exists in coastal North Carolina where the approximate doubling of the concentration of ammonia in precipitation over the past 10 years has been attributed to the proliferation of hog farms in the region (Paerl 1997b). Such increases in atmospheric ammonia deposition are not only important because of the additional sources of nitrogen to surface waters, but also because ammonia represents the most readily-available form of nitrogen for most aquatic organisms.

- **Long Island Sound Estuary Program (NY, CT).** The Long Island Sound Study (LISS) Estuary Program has been evaluating atmospheric nitrogen sources leading to the development of a final nitrogen control plan. Wet and dry deposition monitoring studies have expanded in recent years through a cooperative effort with the Connecticut Department of Environmental Protection (CTDEP) and the University of Connecticut (UConn). The UConn now maintains eight sampling stations spread throughout Connecticut where wet and dry monitoring of nutrients and mercury is conducted. The data have been key to estimating nitrogen deposition loads, which are about 10 lb/acre-year in the Long Island Sound region. The anthropogenic component of the atmospheric deposition delivered to Long Island Sound is estimated to be around 6,700 tons of nitrogen annually including about 3,700 tons that fall directly on the sound. This combined direct and indirect deposition represents about 15 percent of the total load of nitrogen to Long Island Sound from the New York and Connecticut portions of the watershed. Additional nitrogen loadings come from atmospheric deposition onto the Long Island Sound drainage basins north of Connecticut, the watersheds of the New York/New Jersey Harbor and Narragansett Bay, and from direct deposition on the Atlantic Ocean that currents transport into Long Island Sound. In February 1998, the States of New York and Connecticut and EPA agreed to a reduction target of 58.5 percent below a 1990 baseline for point and terrestrial non-point source enrichment. While achieving that target will greatly improve oxygen conditions in the sound, it will not attain existing State water quality standards for dissolved oxygen. In a TMDL analysis being prepared by Connecticut and New York, additional actions are identified, including atmospheric reductions of nitrogen planned under the CAA. The analysis identifies that reducing atmospheric sources of nitrogen will be key to long term efforts to attain water quality standards.

- **Massachusetts Bays (MA).** Wet and dry deposition of toxic compounds, including metals and PAHs, were measured from September 1992 to September 1993 at two sites, one in the northern bay and one in the southern bay on Cape Cod. Dry deposition was greater at the northern site (close to Boston) for most metals. Wet deposition, on the other hand, was greater at the southern site for the metals. The high dry deposition rates at the northern site are probably due to its proximity to Boston. The high wet deposition rates on Cape Cod are probably from sources upwind in southern New England, New York, and New Jersey. Both dry and wet deposition of PAHs were higher at the northern site, also probably from sources in the Boston metro area, including Logan Airport. Dry deposition was highest at both sites in the winter. No PCBs were found at either site.

  Wet nitrogen deposition data from four regional (three in Massachusetts and one in coastal Maine) NADP sites were also analyzed from the early 1980s (1980, 1981, or 1982, depending on the site) through 1993. Dry deposition data were collected from a literature search. Direct

deposition to the bays was estimated to be 6-8 percent of the total nitrogen load. Approximately two-thirds were in the form of wet deposition. The percentage of nitrogen in the surface layer, where a large portion of the biological activity occurs, was also estimated in an attempt to quantify the biological availability of atmospherically deposited nitrogen. Direct deposition was estimated to be approximately 2 percent of this surface-layer nitrogen during the winter months. However, this is probably the lowest percentage that occurs during the year, and deposition may be an important source of nitrogen in the summer months. Uncertainties related to *in situ* dry deposition sampling and wet dissolved organic nitrogen sampling require additional research.

- **New York/New Jersey Harbor Estuary Program (NY, NJ).** New York-New Jersey Harbor is currently the focus of several studies relating to sources of nutrient and toxic pollution loadings to the harbor. These studies will help to quantify pollutant loadings under the TMDL determination. As part of that effort, four air deposition monitoring stations were set up in the harbor area for limited monitoring for PCBs, PAHs, dioxin, heavy metals, and nitrogen. This information will then be available to determine the total loadings of contaminants that are not meeting criteria from all sources. Control options for meeting the TMDLs may include a reduction of air sources. This work is being conducted in cooperation with the New Jersey Department of Environmental Protection and the Hudson River Foundation.

- **San Francisco Estuary (CA).** The San Francisco Estuary Project (SFEP) is working with the Bay Area Stormwater Management Agencies Association (BASMAA) to identify sources of air emissions resulting in deposition of pollutants onto the land and to quantify the contribution of air pollutants reaching the estuary in storm water runoff. The pollutants of concern are primarily toxics, including copper, mercury, PCBs, and PAHs. This study is being coordinated with the San Francisco Estuary Regional Monitoring Program for Trace Substances (see page II-76). The San Francisco Estuary Institute (SFEI) coordinates the Regional Monitoring Program, which includes water, sediment, and tissue monitoring and is now being expanded to monitor air deposition. The SFEI is conducting a pilot study to evaluate pollutants which are being deposited from the air directly onto the estuary waters. Based on these studies, local and State agency partners will be able to assess the cost-effectiveness of emission reduction options and quantify the benefits associated with emission reduction strategies.

- **Santa Monica Bay (CA).** The Santa Monica Bay NEP has proposed an air transport/deposition study to (1) quantify emissions of the toxic materials and nitrogen in the Los Angeles air basin that are subsequently deposited in the bay and its watershed; (2) identify pollutant sources and their relative contributions to total pollutant loading to the bay; and, (3) evaluate the relative impacts of air deposition and the benefit of various emission reduction options in order to recommend the most cost-effective measures to control the identified sources. Initially, the Santa Monica Bay study will quantify the wet and dry toxic and nitrogen deposition to the bay surface. Indirect deposition over the landscape will be calculated using a model developed locally for the region that uses air concentrations, local meteorology, and surface types (trees, pavement) to calculate deposition velocities and loadings. This study will measure air concentrations over water, a difficult process that is not often done but that is necessary to improve the understanding of direct deposition processes. The study will also measure the impact of air deposition during "events" (fire storms, rain storms, Santa Ana winds) to understand how these weather patterns contribute to local air deposition.

- **Sarasota Estuary (FL).** The Sarasota Bay NEP is involved in four large research projects.

  Atmospheric Monitoring Site on Sarasota Bay. For the average rainfall year, it is estimated that atmospheric deposition directly to the water surface provides 26.5 percent of the total nitrogen load to the bay. Under cooperative agreements with EPA, the Southwest Florida Water Management District (SWFWMD), and local governments, the Sarasota Bay NEP initiated an intensive, 1-year atmospheric deposition monitoring effort (within the national NADP/AIRMoN program) in September 1998. The intensive monitoring program is designed to establish relationships between emission sources or source regions and deposition to specific receptors.

  Atmospheric Transport and Dispersion Model. Preliminary modeling by EPA using the Regional Atmospheric Deposition Model (RADM) at an 80 km grid suggested that 70 percent of the atmospheric nitrogen deposited to Sarasota Bay may originate from outside the watershed. Therefore, the Sarasota Bay NEP contracted with the University of South Florida to develop a regional atmospheric transport and dispersion model to determine the impact of NO_x emissions on Sarasota Bay water quality. Investigators modeled atmospheric dispersion, transport, chemical transformation, and deposition of NO_x, nitric acid, and nitrate from stationary and mobile sources using CALMET/CALPUFF, a Lagrangian puff model. A regional domain of 250 km by 500 km with a 20 km grid was modeled and included emissions from the metropolitan areas of Tampa, Orlando, Miami, and Fort Myers. The model indicated that Sarasota Bay shared the same airshed as Tampa Bay and the airshed encompassed the entire modeling domain. The model further indicated that mobile source emissions may be responsible for the majority (81 percent) of atmospheric nitrogen sources to Sarasota Bay. One caveat of the modeling, however, was that modeled wet deposition was approximately a factor of five lower than measured fluxes. Furthermore, utilities were found to contribute disproportionately to wet deposition. Therefore, the total contribution of utilities to atmospheric deposition may be underestimated and that of mobile sources may be overestimated.

  Biological Effects of Atmospheric Deposition. Areas of the Sarasota Bay that receive the greatest percentage of nitrogen loading from atmospheric sources are also associated with the highest water quality; however, total nutrient loads to these segments are lower. Therefore, an investigation of the effects of atmospheric deposition on algal assemblages was initiated by the Sarasota Bay NEP through cooperative funding by SWFWMD and is being conducted by Mote Marine Laboratory. The growth response of phytoplankton to rainwater and nutrient additions is being determined by changes in major taxon composition, changes in particle size distribution, and through high performance liquid chromatography of photosynthetic pigment composition. The final results of this study should yield information on the major taxon composing a nutrient-rich (nearshore) and nutrient-depleted (offshore) algal regime, changes in growth rates as a result of rainfall and nutrient additions, and the potential of rainfall to act as a trigger for algal blooms in each regime. This research should provide information on the biological effects of atmospheric deposition.

  Stable Isotopes to Trace Nitrogen Sources. This on-going study funded by EPA will use stable nitrogen isotope ratios ($^{15}N/^{14}N$) to determine the relative contributions of different types of sources (e.g., wastewater treatment plant effluent, fertilizer runoff, animal waste, and combustion processes), including air deposition of nitrogen. Both nitrogen isotopes are naturally-occurring, but the ratio of $^{15}N/^{14}N$ varies depending on the source. Measuring the ratio in emissions from different sources, in rainwater (wet deposition), and in phytoplankton, macroalgae, seagrasses, and the water column will help researchers identify the sources of atmospherically-deposited

nitrogen and its effects once it reaches the estuary. This is the only NEP air deposition study that measures nitrogen in the food web.

- **Tampa Bay Atmospheric Deposition Projects (FL).** The EPA and its partners in the Tampa Bay NEP (TBNEP) are currently working on eight separate but related projects to characterize the sources and impacts of atmospheric deposition to Tampa Bay and its watershed. A brief summary and status for each of these projects follows:

  (1) An intensive monitoring site, sponsored by TBNEP and EPA's Great Waters program, was created to quantify nitrogen loading from atmospheric deposition to the surface of Tampa Bay, estimate relative contributions from wet and dry deposition, assess temporal variability of wet and dry deposition, and assess the relative contribution of different nitrogen species to atmospheric deposition in Tampa Bay. Preliminary results indicate that atmospheric deposition directly to the bay's surface accounts for approximately one-third of the new nitrogen delivered from all sources to the bay.

  (2) The TBNEP initiated a study to estimate the contribution of atmospherically-deposited nitrogen to storm water loading from residential basins and estimate attenuation of nitrogen from atmospheric deposition in residential basins. The results indicated that approximately 15-20 percent of atmospherically-derived nitrogen was discharged from these basins per rainfall event.

  (3) Toxic materials sampling, sponsored by TBNEP, EPA, Florida Department of Environmental Protection (FDEP), and the Environmental Protection Committee of Hillsborough County (EPCHC), is being initiated to quantify metal concentrations and other contaminants in ambient air and to estimate potential loadings to water and the watershed.

  (4) The TBNEP and the Great Waters program will fund ammonia sampling to map the pattern of ambient air ammonia concentrations. Initial results from a pilot study indicate a strong gradient in ambient ammonia from the highly industrialized east bay to background levels at the existing intensive monitoring site. The overall objective is to develop a surface map showing relative concentrations of ammonia across the northern Hillsborough Bay area.

  (5) Beginning in the fall of 1998, the FDEP funded a study in which the Florida State University and the University of Virginia used N-isotopic ratios to identify nitrogen source types affecting Tampa Bay. In particular, the researchers are using isotopic ratios to attribute the relative contribution from different source types to atmospheric nitrogen, including combustion engines, coal-fired power plants, and diesel engines.

  (6) Local governments and the TBNEP have cooperatively developed and are operating a long-term spatial monitoring network for atmospheric deposition in the Tampa Bay region. The purpose of the monitoring network is to track the contribution and temporal trends of atmospheric nitrogen loading throughout the region.

  (7) The EPA is using RADM to identify relative contributions to nitrogen deposition in Tampa Bay from near and far sources. This effort also examines deposition to the bay and watershed.

(8) In 1999, the FDEP is scheduled to initiate the Bay Regional Atmospheric Chemistry Experiment (BRACE) to (1) refine estimates of annual deposition of nitrogen species (HNO_3, NH_3, NH_4NO_3) to Tampa Bay and its watershed; (2) predict urban ambient ozone and $PM_{2.5}$ concentrations; and, (3) estimate contributions of primary emissions from motor vehicles and stationary sources. This project will include high resolution deposition-daily event samples analyzed for nutrients and trace metals (including mercury), and a 1-year field monitoring program using a Differential Optical Absorption Spectrometer to measure sulfur dioxide, ozone, nitric oxide, nitrogen dioxide or nitrite, benzene, toluene, organics, metals, pesticides, and xylenes.

NOAA ACTIVITIES

National Estuarine Research Reserve System

Under the 1972 Coastal Zone Management Act, Congress created the National Estuarine Research Reserve System (NERRS) to enhance the scientific understanding and management of the Nation's estuaries and coastal habitats. The NERRS is a network of protected estuarine areas in which Federal, State, and local partnerships work to promote stewardship, education, and research. As of 1999, 23 reserves were designated as NERRS sites, encompassing about 960,000 acres of estuarine waters, wetlands, and uplands. Four additional sites have been proposed and are in the process of development and designation. See Figure I-2 for the location of NERRS sites. All NERRS estuaries are included in the definition of "Great Waters."

The NOAA's Estuarine Reserves Division is working with all NERRS sites to implement a System-wide Monitoring Program (SWMP) to track the status and trends in coastal ecosystem health. This national monitoring program will be coordinated with other national and regional programs (i.e., NEP, EMAP, National Status and Trends). The overall goal of SWMP is to identify and track short-term variability and long-term changes in the integrity and biodiversity of representative estuarine ecosystems and coastal watersheds for the purpose of contributing to effective national, regional, and site specific coastal zone management.

Currently, SWMP is focusing on compiling water quality and weather data. Within 22 reserves in the system (Kachemak Bay NERR, Alaska was designated in February 1999 and is not yet implementing SWMP), two locations – one non-impacted (baseline) and one non-point source impacted – are designated as water quality monitoring sites where water quality parameters are measured every 30 minutes. In addition, meteorological data collection began at each NERRS site in February 1998 to allow local weather events to be related to water quality conditions (NOAA NERRS 1998). Although SWMP does not include atmospheric deposition monitoring, it will provide data useful for tracking the ecological health of the coastal Great Waters.

Assessing Relative Nitrogen Inputs to Coastal Waters From the Atmosphere

With funding from EPA's Office of Water and Great Waters program, NOAA's Air Resources Laboratory (ARL) is performing a comprehensive assessment of nitrogen deposition to estuaries. In 1998, NOAA held two workshops involving experts from government, academia, and other institutions to assemble and evaluate nitrogen deposition data and assessment procedures for approximately 40 estuaries on the Atlantic and Gulf of Mexico coasts. Workshop participants evaluated the adequacy of existing nitrogen deposition data and attempted to develop standard nitrogen loading and mass balance

assessment methods. Standard assessment methods are needed to improve comparisons between studies and locations. The report is being produced in 2000.

Coastal Zone Management Program

The Coastal Zone Management Act of 1972 established the national coastal zone management (CZM) program, a voluntary partnership between the Federal government and the coastal States and territories of the U.S., with the following goals:

- Preserve, protect, develop, and (where possible) restore and enhance the resources of the Nation's coastal zone for this and succeeding generations;
- Encourage and assist the States and tribes to effectively exercise their responsibilities in the coastal zone to achieve the wise use of land and water resources of the coastal zone, giving full consideration to ecological, cultural, historic, and aesthetic values as well as the needs for compatible economic development;
- Encourage the preparation of special area management plans to provide increased specificity in protecting significant natural resources, reasonable coastal-dependent economic growth, improved protection of life and property in hazardous areas, and improved predictability in governmental decision making; and,
- Encourage the participation, cooperation, and coordination of the public, Federal, State, tribal, local, interstate, and regional agencies and governments affecting the coastal zone.

Since 1974, at least 32 Federally-approved CZM State programs have protected more than 99 percent of the Nation's 95,000 miles of oceanic and Great Lakes coastline.

As a component of the overall CZM effort, NOAA and EPA are currently developing a Coastal Non-Point Pollution Control Program for each CZM State program. The EPA has created pollution management and control measures for five non-point source categories: agricultural runoff, urban runoff, forestry runoff, marinas, and hydromodification.

OZONE TRANSPORT COMMISSION (OTC)

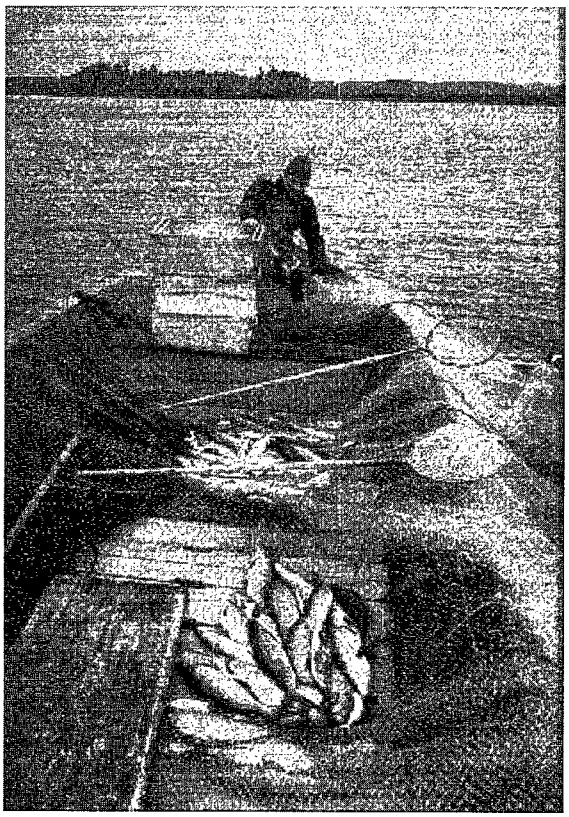
Section 184 of the CAA delineates a multistate ozone transport region (OTR) in the Northeast and requires specific additional NO_x and VOC controls for areas in this region, including attainment areas. In addition, section 184 of the CAA established the Ozone Transport Commission (OTC) to assess the degree of ozone transport in the OTR and to recommend strategies to mitigate the interstate transport of pollution. States in the OTR include Connecticut, Delaware, Maine, Maryland, Massachusetts, New Hampshire, New Jersey, New York, Pennsylvania, Rhode Island, Vermont, parts of northern Virginia, and the District of Columbia. The OTC has concluded that regional reductions of NO_x emissions are particularly important in reducing ozone.

To further control NO_x emissions in the OTR, the OTR States agreed to implement RACT on major stationary sources of NO_x and to a phased approach for additional controls, beyond RACT, for power plants and other large fuel combustion sources. This agreement, the OTC Memorandum of Understanding (MOU) for stationary source NO_x controls, was approved on September 27, 1994. All OTC States, except Virginia, are signatories to the MOU. The MOU establishes an emissions trading

system to reduce the costs of compliance with the control requirements. In addition, in developing State budgets for the NO_x SIP call, EPA considered the NO_x reductions each OTR State committed to in the MOU.

III.C STATE, LOCAL, AND TRIBAL ACTIVITIES

This section describes State, local, and tribal activities that will make significant contributions to understanding or reducing atmospheric deposition of toxic air pollutants to the Great Waters. Unlike the regional and waterbody-specific programs and activities described in Section III.B, these programs are led by State, local, or tribal agencies, not Federal agencies. However, EPA and other Federal agencies are partners in some of the programs.



Minnesota Sea Grant, Jeff Gunderson

STATE AND LOCAL ACTIVITIES

The projects described below are examples of State and local projects that support the goals of the Great Waters program. It is likely that there are many additional relevant State and local programs that were not identified for this report.

Ammonia Study in North Carolina: An Example of Progress in Understanding

The State of North Carolina recognizes nitrogen enrichment and eutrophication as a serious environmental concern for certain coastal plains, nitrogen-sensitive estuaries, and coastal waters (see, for example, the discussion of the Albemarle-Pamlico Estuary on page III-42). Atmospheric emissions and deposition of ammonia from intensive livestock operations are the subject of particular attention, in addition to atmospheric nitrates and water-borne discharges and runoff. A workshop on atmospheric

nitrogen compounds was held at North Carolina State University (NCSU) in March 1997 (Aneja et al. 1998).

The State of North Carolina Department of Environment and Natural Resources, in conjunction with NCSU, University of North Carolina at Chapel Hill, U.S. Department of Agriculture, and EPA, has begun a coordinated research program on ammonia emissions from large-scale livestock operations, and on the transport and deposition of ammonia, with possible effects of eutrophication in aquatic systems and on forest and crop production. Ammonia and other NH_x compounds have much different atmospheric lifetimes and interactions in the environment than do oxidized nitrogen compounds or NO_x (primarily NO and NO_2). The research program on nitrogen compounds will quantify the emissions, verify or improve existing emission factors, and begin modeling studies of deposition patterns, especially in the Coastal Plains of North Carolina.

Several aspects of the research program are already under way. New data on emissions of ammonia from waste lagoons and animal barns/houses have been gathered and are under review. Studies of deposition, deposition velocities, and movements of ammonia in the environment have begun, and a nitrogen balance is being developed to better understand sources, sinks, and exchanges of nitrogen compounds. Emissions data for ammonia and NO_x sources have been produced. Modeling using the Regional Atmospheric Deposition Model (RADM) and Models-3 is under way. A general conference on ammonia and other atmospheric nitrogen compounds was held in June 1999, in addition to intensive reviews of the North Carolina research program as individual studies are completed. These analyses will contribute to an understanding of which sources or source categories are generating the most environmental impact and which should be the focus of additional management efforts (Personal Communication with George Murray, NC DENR, September 9, 1998).

Maryland's Power Plant Research Program

The Maryland Department of Natural Resources (DNR), Power Plant Research Program, in cooperation with other partners, has several projects ongoing in the Chesapeake Bay watershed. These include applying the CALPUFF model to develop estimates of Maryland's contribution of atmospheric deposition of nitrogen to the Chesapeake Bay; using CALPUFF to assess the implications of possible utility emissions trading under title I of the CAA and impacts of deregulation on power plant emissions. The DNR also supports studies on air toxics, particularly mercury from coal-fired power plants, the migration of metals and nitrate through watersheds (including coastal wetlands and forests), and economic resource valuations associated with implementation of the CAA.

Florida's Mercury Rule: Progress in Reducing Atmospheric Mercury Emissions

The amount of mercury in Florida's municipal solid waste stream has been dropping rapidly due to the implementation of statutes and rules designed to reduce or replace mercury in the manufacture of widely used products, recycle mercury-containing items such as fluorescent lamps, and control mercury at the point of release. One tool the Florida Department of Environmental Protection uses to reduce atmospheric mercury emissions is a rule (i.e., "The Mercury Rule") adopted in 1993 that limits mercury emissions from municipal waste combustors (MWCs) to a level that is more stringent than the applicable EPA standard. Florida's strict mercury emissions standard is currently being met by over half of the MWCs in the State, and all MWCs in the State are expected to meet the standard in the year 2000. The rule also requires that every MWC unit perform a mercury stack test at least once each year. With over 5 years of test data available, it is clear that the uncontrolled (i.e., no up-front sorting of waste and no fine

particulate control) MWC units in Florida are emitting about 65 percent less mercury than just 5 years ago. With post-combustion controls (e.g., carbon injection) designed to capture mercury, the stack emission rates at some Florida MWCs have been reduced an additional 80 percent. In 1997, the average mercury emission rate for south Florida MWC units was 31 μg/dry standard m^3 at 7 percent oxygen (Memorandum from Michael M. Hewett to Howard L. Rhodes, August 26, 1998).

South Florida Mercury Science Program

Mercury bioaccumulation in wildlife is extensive in Florida (FDEP 1996). High levels of mercury in several species of fish have resulted in bans or restrictions on their consumption in over half of the fresh waters of the State. The entire Florida Everglades is covered by fish consumption bans. In addition, the mercury problem places at risk a variety of wildlife within the Everglades, most notably top predators such as the endangered Florida Panther. The contribution of atmospheric deposition of mercury to this regional environmental problem is a subject of several current research efforts.

Participants in the South Florida Mercury Program

- Florida Department of Environmental Protection
- Florida Game and Fresh Water Fish Commission
- South Florida Water Management District
- U.S. EPA
- U.S. Geological Survey
- National Park Service
- U.S. Fish and Wildlife Service
- U.S. Army Corps of Engineers
- Florida Electric Power Coordinating Group
- Electric Power Research Institute
- Florida Power & Light Company
- Florida International University
- Florida State University
- University of Florida

The South Florida Mercury Science Program is a broad, multidisciplinary effort by scientists from State and Federal agencies, State universities, industry groups, and others (see sidebar) working together to understand and address mercury bioaccumulation in South Florida (FDEP 1996). The program is designed to determine the following:

- Potential risks to humans and wildlife from mercury in South Florida;
- How mercury enters the aquatic food chain and concentrates in predators;
- Chemical and biological pathways for transformation of inorganic mercury into methylmercury;
- The origin of mercury in South Florida's atmosphere and waters;
- How mercury moves through air, water, and soil; and,
- Actions that could be taken to reduce levels of mercury in fish and wildlife.

The Florida Everglades is currently the focus of the most in-depth and comprehensive research on mercury in the environment. Scientific findings in the areas of atmospheric mercury deposition, aquatic chemistry and cycling of mercury, and bioaccumulation of mercury in food chains have broad application to many of the Great Waters (see Chapter II for information on mercury in the Great Waters). In addition, scientists studying mercury in South Florida hope to incorporate their findings into a model that could be applied to other ecosystems.

Studies to date identified the atmosphere as the primary source (>95 percent) of mercury impinging on the Everglades. Much remains to be learned, however, about the sources of mercury as well as its fate in the environment. Several of the key projects currently under way are described below.

Florida Atmospheric Mercury Study (FAMS)

The FAMS was a large, regional-scale study conducted from 1992-1996 and designed to measure long-term temporal and spatial trends in atmospheric mercury transport and deposition in Florida. While most of the sites were remote, some were extremely close to or in urban areas (e.g., the Fort Meyers site). Data were collected over a 3-year period at nine sites (seven in South Florida), including monthly integrated samples of mercury in rainfall and weekly integrated vapor phase and particulate phase mercury samples. Preliminary analysis of the FAMS data indicated the following:

- More than 90 percent of the mercury found in the Everglades was from atmospheric deposition, while less than 10 percent was from agricultural runoff;
- Wet deposition of mercury to South Florida was high – approximately double what has been observed at other North American remote sites;
- Particulate mercury levels were relatively low;
- Mercury deposition exhibited a strong seasonal trend – 85 percent of annual deposition occurs in the summer; and,
- Slight spatial trends were evident in South Florida.

Although the FAMS study design limited its ability to differentiate between local, regional, and global sources of atmospheric mercury, the researchers conducting the study suggested that local source contributions of mercury to the Everglades are less dominant (<30 percent) than regional and/or global contributions (Guentzel et al. 1995).

South Florida Atmospheric Mercury Monitoring Pilot Study (SoFAMMS)

The SoFAMMS was a short-term, very intensive pilot study focused on determining the ability of state-of-the-art sampling, measurement, and modeling techniques to track mercury from sources to receptors. As a complement to FAMS, SoFAMMS was carried out to assess the influence of local mercury sources in the developed Southeast Florida Coast on the atmospheric deposition of mercury to the Everglades. Over a 1-month period (August 6, 1995 - September 6, 1995), SoFAMMS measured mercury emissions from three source types (municipal waste incinerator, medical waste incinerator, and coal-fired cement plant), meteorological conditions across the study area (surface and upper air), and several forms of atmospheric mercury and deposition at 17 ambient monitoring sites. The data were subjected to extensive dispersion, receptor, elemental composition, and meteorological modeling. The study found a wide range in spatial variability of mercury wet deposition. Volume weighted mean concentrations ranged from 13 to 31 ng/l across the 17 sites. The highest mercury concentrations were observed in the urban areas (19-31 ng/l). The sites in the Everglades were lower but still elevated, ranging from 13-20 ng/l. Those precipitation events in the Everglades with high mercury concentrations were also found to contain elevated concentrations of other trace element species known to be tracers for anthropogenic sources. The precipitation data were subjected to extensive meteorological, atmospheric dispersion, and source apportionment modeling. The results of the modeling indicated that greater than 70 percent of the mercury wet deposited to the Everglades were accounted for by waste incineration and

oil combustion sources, contrary to the preliminary FAMS results that indicated that local sources contribute less than 30 percent (Dvonch et al. 1998). Furthermore, monitoring data from these three sources were the first data to indicate that a high percentage of emissions from incinerators are in the form of divalent (reactive) mercury. Based on these results, EPA's Office of Research and Development has begun methods development efforts for ambient and source mercury speciation techniques.

Mercury Cycling in the Florida Everglades Project

The overall objective of this project is to provide resource managers scientific information on the hydrological, biological, and geochemical processes controlling mercury cycling in the Everglades (Krabbenhoft 1996). Specific areas of research include geochemical studies of mercury, mercury methylation and demethylation studies, interactions between dissolved organic carbon and mercury, mercury accumulation in sediments, physical and chemical processes in peat, sulfur cycling studies, biological uptake of mercury and lower food chain transfer pathways, and groundwater/surface water exchange. The USGS is leading this research effort, and participating scientists are from USGS, SFWMD, FDEP, EPA, Wisconsin Department of Natural Resources, and University of Wisconsin-Madison.

South Florida Ecosystem Assessment Project

This project is part of the EPA Region IV Regional Environmental Monitoring and Assessment Program (R-EMAP), which was designed to monitor the condition of ecological resources in South Florida (Stober et al. 1996). The project was intended to address several issues that threaten the Everglades ecosystem, including mercury contamination.

The project assessed mercury concentrations (e.g., in water, soil, algae, mosquitofish) at approximately 700 sampling sites. Interim findings provide an indication of the spatial distribution of mercury within the Florida Everglades as well as the levels of mercury contamination at various trophic levels in the food chain (Stober et al. 1996). The spatial distribution of mercury within the Everglades is relevant to the Great Waters research because it helps define the environmental conditions under which methylation and bioaccumulation occur. For example, the highest concentrations of methylmercury were found in fish, birds, and algae from the marsh sites between Alligator Alley and Tamiami Trail. North of Alligator Alley, the organic compounds and reduced sulfate are believed to bind the mercury and methylmercury so it is not available for uptake by organisms. South of Tamiami Trail, lower concentrations of sulfate and total phosphorous probably limit microbial methylation and organic production rates, respectively. In addition, researchers have found methylating bacteria associated with periphyton (attached algae) mats, which are more common in the marsh sites between Alligator Alley and Tamiami Trail.

Additional sampling has been conducted, and an updated report is expected soon. The study results will be used to answer the seven questions identified for mercury. The EPA Region IV is also studying the complex interactions between mercury contamination and other issues, such as eutrophication, habitat alteration, and hydropattern modification.

Midwestern Pollution Prevention Activities

Since the *Second Great Waters Report to Congress*, State and local pollution prevention activities have helped to reduce releases of Great Waters pollutants of concern. Six such activities in the Midwest are described below.

State Pesticide Clean Sweep Programs

Over time, unwanted pesticides have accumulated in the barns, sheds, and storage areas of farmers, ranchers, golf courses, pest control operators, and other pesticide users. The pesticides may be unwanted for a variety of reasons – the products could be banned, unusable, or not needed. Farmers and other small businesses may have difficulty determining how to properly manage these pesticides since some (but not all) may be hazardous wastes when they are disposed.

States have addressed the problem of accumulated unwanted pesticides by establishing waste pesticide collection and disposal programs, commonly called "Clean Sweeps." These programs provide a simple way to properly dispose of unwanted pesticides at little or no cost to the participants. Because each State has designed its program to fit its own needs and funding sources, there is no single "typical" Clean Sweep program. Some of the variations include the following:

- *Format* – The pesticides may be collected by holding single-day collection events, picking up pesticides from individual farms, or establishing permanent collection sites;
- *Type of waste collected* – Waste pesticide collections may be combined with household hazardous waste programs either by consolidating all of the waste or by collecting both waste types at a single site but handling them separately;
- *Organizer* – The programs may be run by a State regulatory agency, the agricultural extension service, a county, or a combination of these;
- *Funding source* – Clean Sweep programs may be funded through State pesticide registration fees, State legislature appropriations, Federal grants, or fees assessed to participants; and,
- *Participants* – Some programs are limited to farmers, while others are open to households and/or small businesses.

State Clean Sweep programs have been extremely successful in removing pesticides from the environment and ensuring the proper management of these materials. A few highlights of their accomplishments through 1997 include the following:

- Clean Sweep programs have collected and disposed of more than 12 million pounds of pesticides;
- Over 40 States have collected and disposed of some pesticides;
- About 20 States have had on-going Clean Sweep programs since 1995 (or earlier); and,
- The collections bring in an average of 200-300 pounds of pesticide per participant (where 100 pounds is equivalent to about 11 gallons of liquid pesticide).

In addition, Table III-7 presents the amounts of pesticides that have been collected nationwide based on the limited data that have been collected. Despite this progress, efforts in this area need to continue. There is a large, but unquantified, amount of unused and/or unwanted pesticides that needs to be collected.

Table III-7
Pounds of Pesticides Collected through Clean Sweep Programs Nationwide

Pesticide	Average Percent (per collection)	Total Amount in U.S. (pounds)
DDT	3.86	463,200
Toxaphene	2.98	357,600
Chlordane	1.46	175,200
Mercury	1.53	183,600
Dieldrin	0.48	57,600

Illinois Clean Sweep Partners for PCB and Mercury Wastes

The presence of mercury and PCBs in the environment is partially attributable to their widespread use in commercial and consumer products, particularly electrical equipment. While newer technologies in products, such as transformers, capacitors, thermostats, and switches, are PCB and mercury free, older products are not and still pose potential health and environmental risks. Currently, much of the PCB- and mercury-containing equipment encountered during maintenance, remodeling, and demolition work is disposed of in the municipal solid waste stream. Because mercury and PCBs may be released into the environment throughout the disposal process – from the point of disposal, the garbage truck, a transfer station, and the solid waste landfill – the PCB and Mercury CleanSweep Partnership in Cook County, Illinois is attempting to reduce the amount of PCB- and mercury-bearing equipment entering the municipal solid waste stream.

The Cook County PCB and Mercury CleanSweep Partnership is a nonregulatory program sponsored by public and private entities. The CleanSweep Partners have joined resources to help small businesses and local governments identify and properly manage PCB- and mercury-containing materials through a convenient and cost saving program. Through literature and training, the CleanSweep Partners' goal is to educate and assist small businesses and local agency field personnel in a voluntary, public-private initiative to educate and motivate small business operators, particularly electrical and demolition contractors, to manage and dispose of mercury- and PCB- bearing equipment in identifying, handling, transporting, and disposing of mercury- and PCB-bearing equipment. For more information, call the CleanSweep Partners hotline at 1-888-SWEEP22 or visit the web site at www.erc.uic.edu/cleansweep.

The CleanSweep Partners are Commonwealth Edison, Electric Association, City of Chicago Department of the Environment, Cook County Department of Environmental Control, Metropolitan Water Reclamation District of Greater Chicago, Illinois Environmental Protection Agency, EPA, Clean Harbors Environmental Services, National Oil Recyclers Association, Safety-Kleen Corp., North Business - Industrial Council (NORBIC), and the University of Illinois at Chicago School of Public Health.

Michigan Mercury Pollution Prevention Task Force

The Michigan Mercury Pollution Prevention task force, which first convened in August 1994, has been active in many mercury pollution prevention activities throughout Michigan. Significant accomplishments include (1) a household hazardous waste collection program in 22 counties sponsored

by the Michigan Department of Environmental Quality (MDEQ), resulting in the collection of 200 pounds of mercury; (2) distribution of 16,000 copies of the "Merc Concern" brochure throughout Michigan; (3) development of a mercury pollution prevention web page at http://www.deq.state.mi.us/ead/p2sect/mercury; and, (4) distribution of mercury outreach materials to science teachers. Additional accomplishments of the Michigan Mercury Pollution Prevention task force are described below.

- The Michigan Mercury Pollution Prevention task force worked with the automobile manufacturers to phase out the use of mercury in automobiles, including identification of several uses of mercury in automobiles (e.g., in switches, anti-lock brakes, active ride control devices). To date, the manufacturers have made great progress in eliminating mercury switches from automobiles.

- A cooperative effort initiated by the Detroit Wastewater and Sewage Department that included the National Wildlife Federation, the Michigan Dental Association, and MDEQ collected approximately 1,400 pounds of elemental mercury from 400 dentists at 11 drop-off sites.

- The MDEQ, Michigan Department of Agriculture, Michigan Farm Bureau, Michigan Department of Community Health, Michigan Milk Producers Association, Independent Cooperative Milk Producers, and Michigan State University collaborated on a dairy farm mercury manometer pilot collection effort in two counties in Michigan. A total of 16 out of 18 manometers were replaced with a mercury-free substitute, and 12 pounds of mercury were collected and properly disposed. This program may be expanded Statewide.

- Detroit Edison identified 1,500 pounds of mercury used in current product applications and eliminated its use. Consumers Energy identified over 2,900 pounds of mercury used in product applications in 1996 and is now replacing mercury-containing products with mercury-free alternatives.

Indiana Statewide Mercury Awareness Program

The Indiana Statewide Mercury Awareness Program is a State and local partnership dedicated to identifying commercial uses of mercury, investigating pollution prevention opportunities, and developing and implementing outreach strategies. In October 1998, the Indiana Department of Environmental Management initiated an effort to collect and recycle household items containing mercury.

Minnesota Mercury Reduction Initiative

In 1997, the Minnesota Pollution Control Agency (MPCA) began the Mercury Contamination Reduction Initiative, aimed at reducing mercury contamination in fish in Minnesota lakes. A major part of this effort is to receive advice and comments from the public regarding the goals of the initiative. The MPCA established a Mercury Advisory Council that includes representatives from government, business, and environmental groups.

The council's charter is to devise a package of recommendations to reduce mercury contamination in the environment. In January 1999, the council agreed to adopt a goal of reducing mercury releases to Minnesota's air and water by 70 percent (compared to 1990 levels) by 2005, to be established in statute in the upcoming legislative session.

The recommendations that the council voted to forward to the MPCA include the following:

- Encouragement of voluntary commitments on the part of sources of mercury emissions (e.g., power plants, taconite facilities, sewage sludge incinerators) to reduce or work toward reducing mercury emissions;

- Development of a package of seven strategies that the State will advance at the national level to encourage States and the Federal government to act in concert to reduce national mercury emissions; and,

- Development of a package of strategies to persuade consumers to reduce their purchases and use of mercury-containing products and encourage counties to collect more mercury-containing waste in their household hazardous waste pickups.

Western Lake Superior Sanitary District (WLSSD) Pollution Prevention Efforts

The WLSSD is the largest wastewater treatment facility that discharges to the Lake Superior watershed. The WLSSD developed a multimedia mercury zero discharge pilot project with hospitals, clinics, educational institutions, laboratories, and dental practices. As part of this effort, WLSSD partnered with the Northeast District Dental Society to develop recycling procedures for materials containing amalgam particles. In the first year of the project, over 500 pounds of waste material containing amalgam was collected for recycling. Based on the results of the WLSSD pilot project, WLSSD compiled the *Blueprint for Mercury Elimination*, which is a document designed for use by other wastewater treatment facilities in developing and implementing mercury reduction programs.

Northeast States and Eastern Canadian Provinces Mercury Study

During 1996 and 1997, the Northeast States (i.e., Connecticut, Maine, Massachusetts, New Hampshire, New Jersey, New York, Rhode Island, Vermont) and Canadian Eastern Provinces (i.e., New Brunswick, Newfoundland and Labrador, Nova Scotia, Prince Edward Island, and Quebec) held a series of meetings and workshops to address shared mercury pollution issues. In June 1997, the New England Governors and Premiers of Eastern Canada subsequently signed a Mercury Resolution that called for cooperative efforts including the completion of the Northeast Mercury Study. The study, which was completed in February 1998, reflects the combined contribution of State and provincial air, waste, and water management agencies throughout the northeastern U.S. and eastern Canada. It is an informational resource and serves as the foundation for future regional activities, including the development of a coordinated action plan (see below) to reduce the environmental and public health impacts of mercury pollution.

The study reports on emission inventories, transport and deposition modeling, multimedia monitoring and assessment, communication (public and political outreach), and control strategies and effectiveness of controls. The report recommends (1) identifying mercury as a hazardous air contaminant under State air regulations to achieve the most stringent emission rate; (2) conducting an emissions inventory of airborne sources of mercury; (3) implementing the Federal standards for municipal waste combustors and medical waste incinerators by the year 2000; and, (4) forming in-State task forces to assess, evaluate, and communicate mercury-related public health and environmental information.

Northeast Mercury Action Plan

In June 1997, the Conference of New England Governors and Eastern Canadian Premiers charged its Committee on the Environment with developing a regional Mercury Action Plan. The plan was released in May 1998 with the endorsement of the New England Governors and Eastern Canadian Premiers. The Mercury Action Plan identifies steps to address those aspects of the mercury problem in the Northeast that are within the control or influence of the region. The ultimate goal of the plan is the virtual elimination of anthropogenic mercury releases in the northeastern U.S. and eastern Canadian Provinces. In all, the plan lays out 45 specific recommendations addressing the following:

- The establishment of a Regional Mercury Task Force to coordinate the implementation of the plan;
- Mercury emission reduction targets for identified sources such as municipal solid waste combustors, medical waste incinerators, sludge incinerators, utility and non-utility boilers, and industrial and area sources;
- Source reduction and safe waste management practices, including recycling;
- Outreach and education, especially for high-risk populations;
- Research, analysis, and strategic monitoring to further identify and quantify sources of mercury deposition and to monitor deposition patterns and develop meaningful environmental indicators to measure and track progress; and,
- Mercury stockpile management.

TRIBAL ACTIVITIES

Deposition of toxic air pollutants to the Great Waters adversely affects resources (e.g., fisheries) that are of particular cultural and economic importance to many Native American tribes. This section describes partnerships between tribal, State, and Federal governments that have enabled tribes to better assess the ecological and human health risks posed by exposure to the Great Waters pollutants of concern. Financial and/or technological support from Federal and State sources (through projects such as the Effects on Aboriginals from the Great Lakes Environment (EAGLE), the Baseline Assessment Project, and the American Indian Lands Environmental Support Project) and programs initiated by tribal governments (such as aquaculture, CWA section 106 programs, or educational programs) are enabling tribes to successfully conduct better quality assessments of their environment.

Effects on Aboriginals from the Great Lakes Environment (EAGLE)

The EAGLE, a partnership between the Assembly of First Nations and the Medical Services Branch of Health Canada, is a community-based epidemiological project to research health effects of environmental contaminants potentially affecting approximately 100,000 people in 63 First Nation communities in the Great Lakes basin. The EAGLE's main activities to date include (1) a survey of fish and wild meat consumption in Great Lakes First Nations communities, (2) a program to establish safe fish consumption guidelines for First Nations communities, and (3) a health survey accompanied by blood and tissue sampling. Recognizing that even the perception of contamination can have a tremendous impact on the relationship that First Nation communities have with the land, studies to assess

the socio-cultural impact of environmental contamination are also being conducted. Future activities for the EAGLE project will focus on communication/outreach strategies and helping communities develop environmental plans.

Baseline Assessment Project

The EPA initiated a Baseline Assessment of Indian Country in order to provide easy-to-use and accessible environmental data to assist tribal governments and EPA in making sound environmental decisions. A work group, led by the American Indian Environmental Office (AIEO), is gathering and analyzing the existing information on environmental conditions in Indian country. In addition, EPA's program offices have identified 37 priority data sets that need to be developed to track environmental management activities (see sidebar for priority data sets relevant to the Great Waters program). Next, EPA will develop a data management system to meet the data needs. In addition, EPA's Office of Water is reassessing the 2,200 hydrologic unit basins of the U.S. so that tribal lands can be geographically located within specific watersheds.[13] Therefore, environmental conditions on tribal lands, as evaluated through the baseline assessment, will be comparable to conditions on non-tribal lands within the same watersheds.

Draft Baseline Assessment Priority Data Sets

- Ambient air monitoring
- Air toxics
- Point and non-point source loadings
- Fish consumption advisories
- Ecological status of wetlands
- Contaminated sediment
- Pesticide use
- Blood lead screening
- PCB ballast in buildings

American Indian Lands Environmental Support Project

Established by EPA's Office of Enforcement and Compliance Assurance, the American Indian Lands Environmental Support Project (AILESP) is designed to assess the impact of toxic chemicals from permitted point sources on tribal lands. The AILESP integrates release data for multiple sources and media, and information on the potential impact of a variety of contaminants with compliance histories of facilities within 3.1 miles of tribal lands. The information is assembled into a geographic information system (GIS) to help users understand the sources and impacts of pollutants on tribal lands. Preliminary AILESP data include release of trace metals (including cadmium, lead, and mercury) and nitrogen compounds (including nitrogen dioxide, ammonia, nitrate, and NO_x) from certain facilities.

Aquaculture

Interest in aquaculture in tribal communities has recently been stimulated by the desire to preserve ancestral traditions while avoiding health risks associated with the consumption of contaminated fish. Aquaculture is based on the premise that uncontaminated fish can be obtained by breeding and rearing them at uncontaminated sites (e.g., isolating them from contaminated sediments) and feeding them high-quality commercially-supplied food (i.e., circumventing the contaminated food chain). Buttner (1997) aided aquaculture efforts by three tribal communities: Akwesasne Mohawks (St. Regis Mohawks), Mohawks of the Bay of Quinte, and Ojibway of Sucker Creek. Although the programs had varying degrees of success, it was clear that aquaculture had the potential for producing "clean" fish, while creating jobs and minimally impacting the environment.

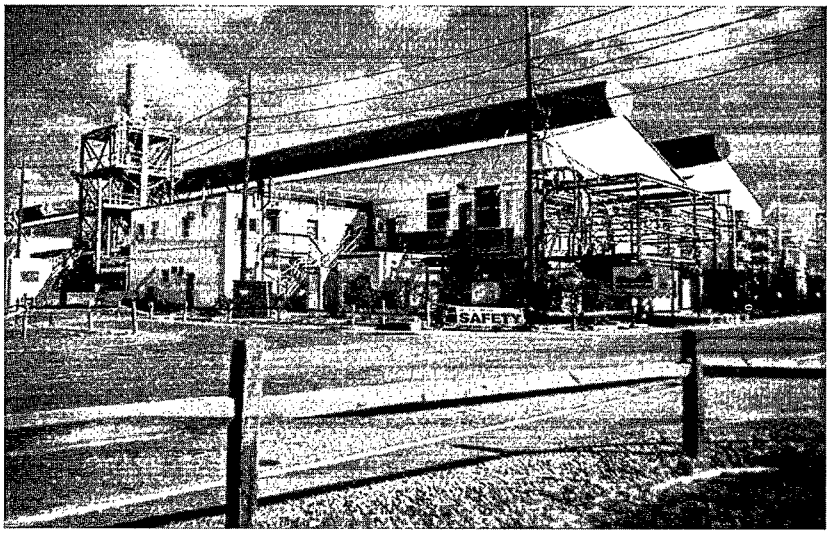
[13] Personal Communication with Ed Liu, EPA. October 26, 1998.

Tribal Air Quality Programs

In 1998, EPA issued a rule that authorizes tribes to develop air quality programs under the CAA. The EPA has also increased its financial support and technical assistance to tribes that choose to adopt air quality programs. Numerous tribes have begun to develop these programs, including programs for collecting air quality monitoring data and programs that address toxic pollutants that are generated in Indian country. The EPA will regulate larger sources of air pollution in Indian country until tribes develop their own regulatory programs. The EPA is also updating data on the number, type, and location of sources of toxic pollutants that are located in Indian country.

III.D INDUSTRY ACTIVITIES

A number of industry activities currently contribute to reduced emissions of Great Waters pollutants of concern. These industry activities were developed by cooperative partnerships involving industry groups, EPA, and other agencies. Some of the activities are noted under other sections of this chapter, such as under the Michigan Mercury Pollution Prevention Task Force. The EPA has found that nonregulatory partnerships can be an effective means of achieving or surpassing environmental goals. In addition, industry is phasing out the use of some Great Waters pollutants of concern in the manufacture of certain products. For example, the amount of mercury used for the manufacture of electric switches and thermostats has been decreasing because of the shift to solid state devices and other alternatives (see Volume II of EPA's 1997 *Mercury Study Report to Congress* (U.S. EPA 1997e)).



The Chlorine Institute

CHLOR-ALKALI INDUSTRY MERCURY REDUCTION GOAL

In July 1997, the Chlorine Institute, on behalf of its members, committed to reduce mercury use in the chlor-alkali industry by 50 percent to help the U.S. achieve the mercury reduction goals of the Binational Toxics Strategy (see page III-66). The baseline average annual mercury usage by mercury cell chlor-alkali plants for the 1990-1995 period was 160 tons per year. The industry's goal is to reduce mercury usage to 80 tons per year by 2005. In addition, as part of the agreement, the Chlorine Institute will submit an annual progress report to EPA. The first annual report was submitted to EPA in May 1998.

To ensure that appropriate oversight is provided for monitoring the progress in achieving the commitment, the Chlorine Institute's Board of Directors established ad hoc committees for technical and management issues. All chlor-alkali producers using mercury cell technology are represented on both committees. In addition, seven technical task groups were formed to address specific issues such as identification of new mercury reduction control techniques and preparation of guidance documents to assist industry members in achieving mercury reduction goals.

VOLUNTARY MERCURY AGREEMENT WITH NORTHWEST INDIANA STEEL MILLS

On September 25, 1998, the Lake Michigan Forum, the Indiana Department of Environmental Management (IDEM), and EPA signed a voluntary agreement with three northwest Indiana steel mills, including Bethlehem Steel Burns Harbor, Ispat Inland Inc. Indiana Harbor Works, and U.S. Steel Gary Works. The mills agreed to inventory mercury in equipment, materials, storage, and waste streams, and to develop facility-specific plans for mercury pollution prevention. The companies signed the agreement as part of the Lake Michigan Primary Metals Project, which is a pollution prevention effort initiated by the Lake Michigan Forum. The Lake Michigan Forum is a stakeholder group that provides input to EPA on the Lake Michigan Lakewide Management Plan and includes representatives from academia, business, environmental and sportfishing groups, and local governments.

The agreement will result in facility-specific reduction plans outlining pollution prevention activities through equipment substitutions, purchasing practices, recycling, better management, and employee education. The EPA (including the Mercury Work Group of the Binational Toxics Strategy) and IDEM will provide the companies with information on typical mercury sources, substitutions for mercury in equipment, and recycling options. Both agencies and the Lake Michigan Forum will receive progress reports from the mills. The reports will also be available to the public. The forum will promote the initiative and its results throughout the Lake Michigan basin. This effort could serve as a model for other companies and industries that use mercury-containing devices.

AMERICAN HOSPITAL ASSOCIATION MOU

On June 24, 1998, the American Hospital Association (AHA), which consists primarily of health care provider organizations, established a Memorandum of Understanding (MOU) with EPA's Office of Prevention, Pesticides, and Toxics and EPA Region V. The MOU is intended to provide AHA members with enhanced tools for minimizing the production of pollutants and reducing the volume of waste generated. The information should also reduce the waste disposal costs incurred by the health care industry.

Hospitals Produce Mercury Wastes

"Medical waste incinerators are the fourth largest releasers of mercury to the environment, constituting approximately 10 percent of all emissions sources, and hospitals are responsible for producing 1 percent of the total municipal solid waste in the entire country." (U.S. EPA 1998d)

The MOU outlined multiple primary goals and activities designed to aid in the exchange of information between EPA and the health care industry. Highlights include the following:

- Development of a Mercury Waste Virtual Elimination Plan to eliminate mercury-containing waste from the health care industry waste stream by the year 2005;
- Development of a Total Waste Volume Reduction Plan to reduce the volume of waste generated by the health care industry by 33 percent by 2003 and by 50 percent by the year 2010; and,
- Investigation of pollution prevention opportunities with respect to ethylene oxide and other persistent, bioaccumulative, and toxic pollutants.

This MOU specifically supports the goals and objectives of the PBT Initiative, the Mercury Action Plan, and the Waste Minimization National Plan, and is also expected to help reduce atmospheric deposition of mercury and other persistent toxic pollutants to the Great Waters (U.S. EPA 1998d).

ELECTRIC POWER RESEARCH INSTITUTE STUDIES

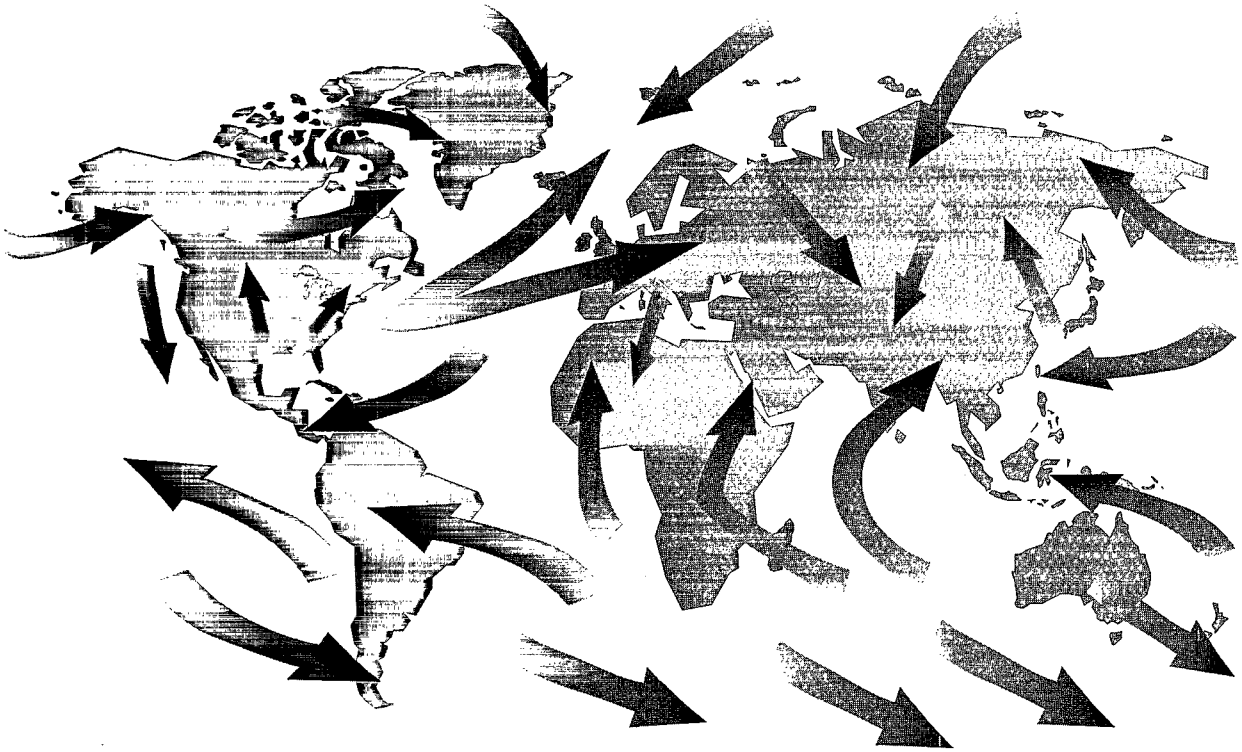
The Electric Power Research Institute (EPRI) has a broad-based research program which conducts a large amount of research cooperatively with Federal and State agencies. Research sponsored by EPRI on air toxics and nitrogen is the largest privately-funded program in the U.S. Current air toxics studies focus on mercury, nickel, dioxins, and arsenic. These studies include atmospheric global, regional, and plume modeling of mercury; measurement of natural mercury fluxes; historic patterns of mercury deposition (sediment and peat cores); environmental effects and mercury cycling in lakes; human health effects of mercury exposure; and, mercury and multimedia risk assessment. Research characterizing emissions of air toxics has, and is, leading to better emission inventories relevant to a number of air quality issues. The EPRI also conducts or sponsors research in atmospheric chemistry and physics, including atmospheric modeling and measurement of PM, ozone, and their precursors (including nitrogen species). On-going nitrogen research of specific interest to the Great Waters includes a small study on measurement of organic nitrogen in precipitation near the Chesapeake Bay; development of a nutrient model for the Chesapeake Bay airshed, watershed, and bay; a study on the feasibility of using isotopic composition of ammonium in wet deposition for source attribution; and, research on the effects of nitrogen speciation in atmospheric deposition on phytoplankton community composition and productivity. In addition, EPRI is a contributor to a study with substantial funding from EPA and NOAA that is being carried out by the Ocean Studies Board and Water Science and Technology Board of the National Research Council's Commission on Geosciences, Environment, and Resources to assess eutrophication, coastal processes, and watershed management. The study report, due in spring 2000, will review existing knowledge and make recommendations for action and research to reduce eutrophication in coastal ecosystems through more effective watershed management.

Businesses for the Bay

Since its launching by the Chesapeake Executive Council in 1996, more than 230 businesses have joined the *Businesses for the Bay*. This includes not only private industries, but State and local government facilities as well. Under this voluntary program, businesses commit to pollution prevention activities and goals. In 1998, member facilities voluntarily reported that they had reduced or recycled 222 million pounds of waste. Of these, 13 facilities reported a resulting cost savings of $1.4 million, and 15 of the facilities offered pollution prevention training to 6,300 employees. Businesses have also volunteered more than 70 of their technical experts to act as mentors to offer pollution prevention advice to other companies on an as-needed basis. In 1998, *Businesses for the Bay* received 2 national awards from the National Pollution Prevention Roundtable and the National Environmental Education and Training Foundation for its successes as a model program.

III.E WORK WITH OTHER COUNTRIES

The U.S. works with other nations on many issues concerning shared resources (e.g., the Great Lakes) and transboundary environmental problems. A number of international activities concern the Great Waters and Great Waters pollutants of concern. International activities relevant to air deposition of pollutants to the Great Waters are discussed below.



CANADA - U.S. BINATIONAL TOXICS STRATEGY

On April 7, 1997, the U.S. and Canada signed the Great Lakes Binational Toxics Strategy (BNS). The BNS sets forth a collaborative process by which Canada and the U.S. will work toward the goal of virtual elimination of persistent toxic substances resulting from human activity from the Great Lakes basin, in order to protect and ensure the health and integrity of the Great Lakes ecosystem. The goal of virtual elimination will be achieved through a variety of programs and actions that encourage cooperation among all relevant sectors of society and which place primary emphasis on pollution prevention.

This coordinated strategy provides the framework to achieve quantifiable goals in a specified timeframe. As noted in the discussion of the PBT Initiative (see page III-4), there are 12 Level I pollutants that represent an immediate priority and are targeted for reduction and eventual elimination through pollution prevention and other incentive-based actions. Nine of these 12 pollutants are Great Waters pollutants of concern.

Both the U.S. and Canada have set "challenge" goals to achieve reductions in releases of the targeted pollutants. One of these challenges is the commitment of both countries to work together to assess atmospheric inputs of persistent toxic substances to the Great Lakes with the goal of evaluating and reporting jointly on the contribution and significance of long-range transport of these substances from worldwide sources. In addition, Environment Canada will complete inventories of ten selected air pollution sources to support assessment of environmental impacts of air toxics by 1999 and will demonstrate alternative processes to reduce emissions from five predominant sources by 2001. The BNS includes several specific reduction goals or challenges for the Level I pollutants. For the U.S., these reductions will be based on the most recent and appropriate inventory for each pollutant (e.g., the mercury inventory is based on 1990 levels). Canada plans to use an inventory from 1988.

BNS Pesticides in the Great Lakes

In December 1998, EPA's Great Lakes National Program Office (GLNPO) released a draft report entitled, *Draft Pesticides Report in Response to the Great Lakes Binational Toxics Strategy.* A final report will be released in fall 1999. The report presents and analyzes data on the environmental presence of five banned pesticides (i.e., chlordane, aldrin/dieldrin, DDT, mirex, toxaphene) in the Great Lakes, along with probable and suspected sources. The report fulfills a "challenge" created by the Binational Toxics Strategy for EPA to confirm by 1998 the elimination of uses and releases of the pesticides from sources that enter the Great Lakes.

The report concludes that although environmental concentrations of the pesticides in the Great Lakes basin have gradually declined for 20 years, they remain at levels of concern in water, sediment, and fish. The EPA found no evidence of environmentally-significant, purposeful releases of the pesticides in the U.S. and concluded that continuing inputs to the Great Lakes are likely to originate from remaining stockpiles of the pesticides held by consumers, long-range transport from countries where the pesticides are not banned, and releases of the pesticides from reservoir sources (e.g., contaminated sites). Based on these findings, EPA concluded that there is a continuing need to pursue activities set forth under the Binational Toxics Strategy.

At the initial June 1997 BNS stakeholder meeting, participants developed a plan to implement the strategy, which applied the following steps to address each priority substance or category of substance: (1) information gathering, (2) assessment of current regulations and programs, (3) identification of cost effective options for further reductions, and (4) recommendations and implementation of actions.

Since the initial stakeholder meeting, substance-specific work groups have been established and are gathering information about baseline levels and sources of pollutants, as well as current programs affecting the pollutants. In addition, some work groups are attempting to identify cost-effective options to achieve reductions. Specific highlights of the mercury work group activities include the AHA MOU (see page III-64), work with the chlor-alkali industry (see page III-63), and an agreement with the steel industry (see page III-64). The PCB work group has supported Clean Sweep programs (e.g., the Illinois Clean Sweep Program described on page III-57) to reduce existing stockpiles. With the International Joint Commission, the BNS participants developed a draft report on sources, pathways, and transformations of the BNS compounds. This report, *Identifying Source Regions of Selected Persistent Toxic Substances in the U.S.*, identifies and ranks source regions for BNS pollutants, identifies regulatory and voluntary programs to control emissions of these compounds, and determines the emissions inventory and control gaps that exist for the BNS compounds. In addition, EPA's Great Lakes National Program Office released a *Draft Pesticides Report in Response to the Great Lakes Binational Toxics Strategy* in December 1998 (see sidebar). Many additional activities that support the Binational Strategy have been implemented by a wide variety of stakeholder groups and are outlined in the *Draft Great Lakes Binational Toxics Strategy: Activities by Partners* (U.S. EPA and Environment Canada 1998). Table III-8 describes the BNS challenge goals for the U.S. for each Level I substance and summarizes recent activities with respect to those goals.

Table III-8
U.S. BNS Challenge Goals and Activities for Level I Substances

Level I Substance	U.S. Challenge Goal	Progress/Activities
Mercury and Compounds	By 2006, a 50 percent reduction in deliberate use and a 50 percent reduction in release from human-activity sources. This release reduction applies to the aggregate releases to the air nationwide and to releases to the water in the Great Lakes basin.	BNS work group activities are focusing on voluntary actions. Formal collaborative efforts are under way with the chlor-alkali industry, the American Hospital Association, and three Indiana steel mills. Outreach projects are ongoing with manufacturers and users of mercury relays and switches, utilities, and laboratories.
Dioxins and Furans	By 2006, a 75 percent reduction in total releases from human-related activities. This release reduction applies to the aggregate releases to the air nationwide and to releases to the water in the Great Lakes basin.	The BNS dioxin work group is coordinating closely with the PBT Initiative dioxin efforts, including a Great Lakes State pilot to target air emissions using cross-media authorities. Voluntary reduction efforts are also planned.
PCBs	By 2006, a 90 percent reduction nationally of high-level PCBs (>500 ppm) used in electrical equipment. Ensure that all PCBs retired from use are properly managed and disposed of to prevent accidental releases.	The BNS PCB work group is developing a work plan. Voluntary actions are being pursued through expanding EPA Region V's PCB phasedown program, encouraging national replication of the phasedown program, implementing a clean sweep pilot in Chicago, and encouraging a national PCB reduction effort.
Chlordane, DDT, Aldrin/Dieldrin, Mirex, Toxaphene, Octachlorostyrene	Confirm, by 1998, that there is no longer use or release from sources that enter the Great Lakes basin. If ongoing long-range sources from outside the U.S. are confirmed, use existing international frameworks to reduce or phase out releases.	A final BNS status report on use and release from Great Lakes basin sources is due fall 1999. The BNS work group is also developing a work plan. The EPA will continue clean sweeps to reduce stockpiles in the Great Lakes basin and will work with stakeholders and Great Lakes States to reduce pesticide reliance. The BNS octachlorostyrene work group is focusing on defining sources, releases, and environmental loadings (and, to some extent, toxicity and bioaccumulation).
Alkyl Lead	Confirm no use in automotive gasoline by 1998. Support and encourage stakeholder efforts to reduce alkyl lead releases from other sources.	The EPA issued a "confirmation of no use in automotive gasoline" report under the BNS in December 1998, broaden stakeholder involvement, encourage stakeholder minimization of use/release from other sources (e.g., aviation, racing,) and track efforts to develop unleaded alternatives for aviation and racing fuel.
Hexachlorobenzene	Seek, by 2006, reductions in releases that are within or may have potential to enter the Great Lakes basin from sources resulting from human activity (percentage goal not yet established).	An initial step under the BNS is to quantify loadings to set a realistic percentage goal. The BNS work group will consider approaches to reduce releases during pesticide manufacturing and use, chlorinated solvent manufacturing, and possibly aluminum manufacturing.

Level I Substance	U.S. Challenge Goal	Progress/Activities
Benzo[a]pyrene	Seek, by 2006, reductions in releases that are within or may have potential to enter the Great Lakes basin from sources resulting from human activity (percentage goal not yet established).	The BNS work group was developing a work plan in 1998.

INTERNATIONAL JOINT COMMISSION

Originally created in 1909 for the purpose of resolving disputes between the U.S. and Canada, the International Joint Commission (IJC) is charged with the responsibility of evaluating and assessing the progress of commitments made by Canada and the U.S. under the 1978 Great Lakes Water Quality Agreement (GLWQA). In keeping with this responsibility, the IJC prepares a biennial report outlining its findings and recommendations. These recommendations are based on information compiled from the Great Lakes Water Quality Board (WQB), Science Advisory Board (SAB), International Air Quality Advisory Board (IAQAB), Council of Great Lakes Research Managers, various task forces, and through a variety of public consultation activities. In 1985, the WQB established a list of 11 critical pollutants which remain the focus of IJC's efforts today. Nine of the critical pollutants overlap with the Great Waters pollutants of concern and the PBT Initiative: DDT/DDE, dieldrin, hexachlorobenzene, lead, mercury, PCBs, dioxins and furans, and toxaphene.

The IJC's *Ninth Biennial Report on Great Lakes Water Quality*, published in June 1998, focused on the issue of persistent toxic substances in the Great Lakes ecosystem, which has been the major focus of IJC's biennial reports since 1990. The IJC continues to stress the importance of eliminating these substances. As in previous biennial reports, IJC developed targeted recommendations to aid Canada and the U.S. to achieve the objectives under the GLWQA. Historically, these recommendations have been incorporated into existing or planned programs, and a few have achieved specific and direct results, including *The Great Lakes Binational Toxics Strategy*. Recommendations to the U.S. and Canada from the 1998 report include the following:

- Accelerate the development of integrated, binational programs to reduce and eliminate sources of persistent toxic substances to the atmosphere;
- Develop and communicate a comprehensive strategy for reducing mercury and NO_x emissions associated with energy production and use;
- Expand research into endocrine disrupting chemicals in humans and wildlife;
- Support the development and application of ecosystem models;
- Identify, assess, and support surveillance and monitoring programs essential to track contaminant loadings to, and concentration trends for, each of the Great Lakes; and,
- Focus reduction and elimination efforts on dioxins, furans, mercury, and PCBs.

The IJC presented additional recommendations on agricultural practices, communication of scientific information, radioactivity, ecological economics, and contaminated sediment in areas of concern (AOCs).

UNITED NATIONS ECONOMIC COMMISSION FOR EUROPE LRTAP PROTOCOLS ON HEAVY METALS AND POPS

In 1979, members of the United Nations Economic Commission for Europe (UN-ECE) created the Long-Range Transboundary Air Pollution (LRTAP) convention to provide a framework for participating countries to limit, gradually reduce, and eventually prevent air pollution. Today, the 57 countries included in the UN-ECE region are the Russian Federation, the Newly Independent States, Central and Eastern Europe, Western Europe, Canada, and the U.S. Protocols to the LRTAP convention negotiated since its creation establish more specific and legally-binding controls and emission reduction targets for certain air pollutants.

In June 1998, the members of the UN-ECE signed protocols on persistent organic pollutants (POPs) and heavy metals. The POPs are defined as organic substances that possess toxic characteristics, are persistent, bioaccumulate, are prone to long-range transboundary transport and deposition, and are likely to cause significant adverse human health and environmental effects. The POPs protocol bans the production and use of eight compounds (i.e., aldrin, chlordane, dieldrin, endrin, hexabromobiphenyl, kepone, mirex, and toxaphene) and limits the production and use of five compounds (i.e., DDT, heptachlor, hexachlorobenzene, lindane, and PCBs). In addition, the POPs protocol requires countries to apply best available technology methods to limit air emissions from stationary sources of dioxins, furans, PAHs, and hexachlorobenzene. The protocol on heavy metals regulates cadmium, lead, and mercury. The protocol bans the use of lead in gasoline and the use of mercury in batteries and requires the application of best available technology to limit air emissions from major stationary sources of all three metals.

Both of these protocols to the LRTAP convention incorporate less stringent obligations for countries with economies in transition, and the protocols offer alternative compliance options to allow some parties to apply different control strategies, provided these strategies achieve equivalent emission reductions. The protocols also commit participating parties to reduce total national air emissions to below the levels reported for a reference year (between 1985 and 1995).

Most recently, in December 1999, the U.S. and Canada, along with European members, signed the LRTAP Protocol to Abate Acidification, Eutrophication and Ground-level Ozone. This Protocol is the most sophisticated environmental agreement so far because its creates the first comprehensive, multinational structure to simultaneously reduce the long range transport of the various pollutants that, in different combinations, cause acid rain, smog and other serious air pollution problems. The signing of this agreement also initiates a new phase within LRTAP to increase emphasis on implementation, compliance, review and extension of existing protocols. In order to accommodate the domestic (acid rain) and bilateral (ozone) processes which are currently under way in both countries, both Canada and the United States will incorporate their emission reduction commitments for sulphur dioxide, nitrogen oxides and volatile organic compounds into the Protocol at the time of its ratification. This accommodates the timing of the bilateral initiative to complete negotiations in the year 2000 of an ozone annex to the U.S. - Canada Air Quality Agreement (see below).

Further information on the LRTAP Convention and its Protocols can be found at http://www.unece.org/env/lrtap.

UNITED NATIONS ENVIRONMENT PROGRAM GLOBAL POPS INITIATIVE

At its 19th session in February 1997, the United Nations Environment Program (UNEP) Governing Council concluded that international action, including a global, legally-binding instrument, is needed to reduce the risks to human health and the environment arising from the release of 12 POPs: aldrin, dieldrin, DDT, endrin, chlordane, hexachlorobenzene, mirex, toxaphene, heptachlor, PCBs, dioxins, and furans. The Governing Council decided that immediate international action should be initiated to reduce and/or eliminate the emissions and discharges of the 12 POPs, and, where appropriate, eliminate production and subsequently the remaining uses of those POPs that are internationally produced. Accordingly, the first session of the Intergovernmental Negotiating Committee (INC) for an International Legally Binding Instrument for Implementing International Action on Certain Persistent Organic Pollutants was held in Montreal in June 1998. The INC is expected to complete an Internationally Legally Binding Instrument by the middle of the year 2000.

Currently, the INC is establishing an expert group for the development of science-based criteria and a procedure for identifying additional POPs as candidates for future international action. In addition, UNEP has initiated a number of immediate actions, such as studies to identify alternatives to POPs, current PCB inventories, sources of dioxins and furans, and available POP destruction capacity.

NAFTA COMMISSION ON ENVIRONMENTAL COOPERATION SOUND MANAGEMENT OF CHEMICALS PROGRAM

On January 1, 1994, the U.S., Canada, and Mexico officially established the North American Agreement on Environmental Cooperation (NAAEC) to foster greater cooperation on environmental issues, including the management and control of several Great Waters pollutants of concern. Subsequently, the NAAEC created the Commission on Environmental Cooperation (CEC) to address regional environmental concerns, prevent potential trade and environmental conflicts, and promote effective enforcement of environmental law. One of the CEC's first activities was to develop a program to identify, measure, and mitigate the environmental impacts of the North American Free Trade Agreement (NAFTA) (NAAEC 1999).

In 1995, the CEC established a Sound Management of Chemicals (SMOC) Work group to address the issues of persistent toxic substances and their effects in and transport between the North American countries. The original duties of the work group members included the development of a North American Regional Action Plan (NARAP) for the management and control of PCBs (Commission for Environmental Cooperation 1998). Since 1995, NARAPs have been developed for PCBs, DDT, chlordane, and mercury. The CEC is currently developing a Phase II NARAP for mercury, which represents an amendment to the first mercury NARAP. The CEC is developing NARAPs for dioxins and furans, and hexachlorobenzene. Draft and final NARAPS and additional information related to this effort is available at www.cec.org/programs_projects/pollutants_health/smoc/smoc-rap.cfm?varlan=english.

Recent Developments on the Mercury North American Regional Action Plan

In October 1998, the North American Implementation Task Force on Mercury held a Science Experts Workshop that focused on mercury source identification, fate, transport, and monitoring and the identification of research needs in these areas. The workshop resulted in a *Strategy for the Development of a Trinational North American Mercury Baseline* for mercury concentrations and fluxes (Draft Agenda, October 6-8, 1998, Science Experts Workshop on Mercury, Las Vegas, NV).

U.S.-CANADA AIR QUALITY AGREEMENT

The U.S.-Canada Air Quality Agreement, which was signed in March 1991, addresses transboundary air pollution between the two countries. The agreement focuses on acid rain and ozone transport issues, prevention of deterioration of air quality and visibility, development of emissions monitoring systems, notification and assessment of major projects which could affect transboundary air quality, and coordinated research activities. The two countries established the Air Quality Committee (AQC) to help implement the agreement. In the past, the AQC focused primarily on acid rain and notification issues and is currently expanding the focus to address transboundary ground-level ozone and fine particles (U.S. EPA 1998s).

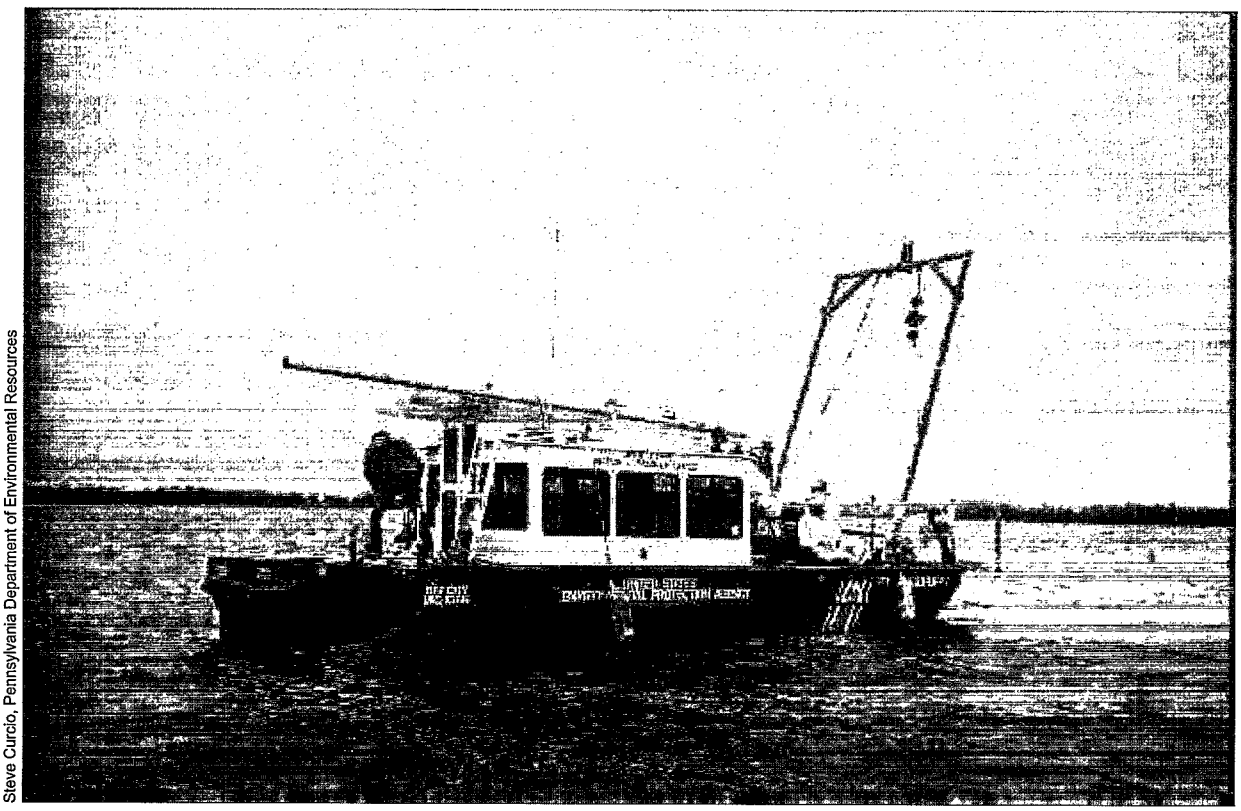
The agreement includes commitments by the U.S. and Canada to reduce SO_2 and NO_x emissions. Specifically, for NO_x, the U.S. and Canada agreed to reduction goals amounting to about 10 percent of the national NO_x emissions for both countries by 2000. This is equal to approximately two million tons in the U.S. and 100,000 tons in Canada. The U.S. expects to meet this goal through mobile and stationary source NO_x emission measures, a large part of which will be realized through Acid Rain Program reductions of emissions from coal-fired electric power plants. After 2000, the U.S. expects to achieve additional reductions in NO_x from implementation of the ozone NAAQS and the NO_x SIP call (see page III-27). Canada has measures in place to reduce NO_x emissions from stationary sources by 100,000 tons by 2000 through national emissions limits for new fossil-fueled power plants, retrofits at several existing power plants, new source standards for boilers, process heaters, and cement kilns, and reconstruction of a metals smelter. In addition, by 2010, Canada anticipates a 10 percent decline in NO_x emissions from 1990 levels as a result of improved emission standards for vehicles.

In April 1997, the Canadian Minister of the Environment and the EPA Administrator reiterated their commitment to addressing transboundary air pollution by signing a Joint Plan of Action for Addressing Transboundary Air Pollution. The commitment focuses on the common concern of both countries for ground-level ozone and fine particles and for protecting public health on both sides of the U.S. - Canada border. Currently, the U.S. and Canada are negotiating an ozone annex to the U.S. - Canada Air Quality Agreement which is expected to be completed by December 2000. This annex will fulfill the two countries' obligations under the LRTAP Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (see discussion above).

CHAPTER IV
SCIENCE AND TOOLS

In addition to the environmental progress discussed in Chapter II and the program progress discussed in Chapter III, there has been significant advancement since the *Second Great Waters Report to Congress* in the development and use of new scientific methods and tools needed to understand the problem associated with Great Waters pollutants of concern. This includes improved understanding and assessment capabilities in some areas and a clearer picture of remaining uncertainties and future research needs in other areas.

This chapter highlights recent advancements in scientific research, as well as new and improved monitoring and modeling capabilities and databases that are improving our understanding of and abilities to address public health and environmental risks posed by the pollutants of concern in the Great Waters. Unlike Chapter II, which focuses on the results of new research and monitoring for the purpose of defining the overall problem, the discussion in this chapter focuses on developments and new findings that improve our understanding of more narrow issues related to pollutant emissions, transport, loadings, and effects. It also discusses the status and directions of research and information sources that will be useful in future assessments of the Great Waters. Finally, the chapter identifies additional research and tools that are necessary to address remaining uncertainties related to atmospheric deposition in the Great Waters. This discussion is not intended to be comprehensive, but rather to highlight some of the key scientific advancements since the *Second Report to Congress*.



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WHAT MAJOR ADVANCEMENTS HAVE OCCURRED SINCE THE SECOND REPORT?

This section summarizes a number of major advancements in scientific research, models, and databases that are and will be useful in better understanding atmospheric deposition of the Great Waters pollutants of concern. Recent developments in emissions inventories are presented first, followed by a discussion of changes in programmatic direction and progress of several ambient air and deposition monitoring networks and environmental monitoring networks. The last two main subsections provide a discussion of recent scientific advancements in environmental fate and transport modeling, and exposure and effects research and modeling.

EMISSION INVENTORIES

National Toxics Inventory

Developing estimates of how much of a pollutant is being emitted is an important key to characterizing the extent of the air toxics problem in the U.S., including persistent, bioaccumulative toxics that are of concern to the Great Waters program. For about 20 years, EPA has routinely collected emissions inventory data for criteria pollutants, such as lead, carbon monoxide, and particulate matter. For air toxics, however, scientists have had to rely on indirect estimates of emissions that are based on emission factors and other imprecise methods. With the air toxics program in the CAA amendments of 1990 came a new focus on air toxics emissions inventories. This information can be used to estimate and characterize risk and develop strategies to reduce risks from air toxics. These pollutants are emitted by major, area, and mobile sources, and emission estimates from these sources can vary from the national-level to regional- and county-level estimates, and to facility- and process-specific emissions data that can be used in air dispersion models.

The dispersion and exposure modeling used to estimate and characterize risks from air toxics requires a model-ready emissions inventory. As a result, EPA has compiled and continues to update and refine the National Toxics Inventory (NTI). To date, EPA has compiled an inventory data set for the 1990 to 1993 period (called the 1993 NTI) and another for 1996. The 1993 NTI data, where available for Great Waters pollutants of concern, are presented in Chapter II to characterize emissions and sources on a national level. The NTI does not include emissions of air toxics from natural sources (e.g., plants, volcanoes).

The 1996 NTI was completed in early 2000, but was not available in time to be included in this report. It represents a substantial improvement over the 1993 NTI in that it is more complete in its coverage of pollutants and their sources, and it carries a higher degree of spatial resolution owing to its development from source-specific information (i.e., the "bottom-up" approach to inventory development) and contains HAP emission estimates for major (facility-specific), area, and mobile on-road and non-road sources. In these 1996 NTI efforts, a majority of the State and local air pollution control agencies have provided direct emission information or a critical review of EPA-developed emissions in a coordinated effort to develop the best inventory in the available timeframe. This approach provides estimates of emissions at the county-level of resolution (or better, in many cases), which subsequently allows the 1996 NTI to be used to develop screening-level modeling assessments of ambient concentrations and inhalation exposures down to the county-level. As such, the 1996 NTI represents the most recently verified and complete emissions inventory available for national assessments.

The EPA has been compiling the 1996 NTI using five primary sources of data:

- State and local toxic air pollutant inventories (developed by State and local air pollution control agencies);
- Existing databases related to EPA's air toxics regulatory program;
- The EPA's Toxic Release Inventory (TRI) database;
- Estimates developed using mobile source methodology (developed by EPA's Office of Transportation and Air Quality); and,
- Emission estimates generated from emission factors and activity data. (NOTE: While major sources were not estimated using emission factors and activity data, 30 area sources were in the 1996 NTI.)

Preference is given to State- and locally-generated information, where available. Where such data are not available, existing data from EPA's regulatory development databases are utilized. If neither of these data sources contains information for a known stationary source, EPA uses data from the TRI. The EPA also gives preference in inventory development to emissions data resulting from direct measurements over those generated from emissions factors and activity data.

The TRI database contains national inventory data submitted by individual facilities that meet certain reporting criteria. The TRI does not account for smaller sources within a source category that do not meet the reporting criteria. Although the TRI data are limited in source category coverage, in many cases, TRI data are used because they are the only available means to estimate emissions from certain source categories. For the missing States and for sources not included in the State inventories, MACT data, or the TRI, EPA estimated air toxic emissions using air toxic emission factors and corresponding activity data.

The compilation of such a large data set presents a significant challenge to EPA and introduces several limitations. In terms of consistency, the NTI is a composite of emissions estimates generated by State and local regulatory agencies, industry, and EPA. Because the estimates originated from a variety of sources and estimation methods, as well as for differing purposes, they vary in quality, included pollutants, level of detail, and geographic coverage. Also, the accuracy of emissions estimation techniques varies with pollutants and source categories. In some cases, an estimate may be based on few (or only one) emissions measurements at a similar source. The techniques used and quality of the estimates will vary between source categories (e.g., some have been better studied than others) and between major, area, and mobile source sectors. Another limitation of the NTI is that emissions from reservoir sources, such as volatilization from soils where chemicals were previously spilled or applied (in the case of pesticides), are either missing or poorly quantified. If such sources are a major component of the total emissions for a given chemical, NTI could under report total emissions for that chemical.

Future updates of the NTI are scheduled every 3 years. However, similar to other inventories, the NTI is dynamic and is subject to change (via periodic updates) as new, more reliable data become available for the year it represents. Further, EPA continues to work with the State and local agencies to promote consistency and accuracy in estimating and reporting emissions information for future years.

The Great Lakes Regional Air Toxics Emissions Inventory

Under the auspices of the Great Lakes Commission, the eight U.S. Great Lakes States and the Canadian province of Ontario have been engaged since the early 1990s in creating and updating a regional air toxics emissions inventory of the Great Lakes pollutants of concern – the Great Lakes Regional Air Toxics Emissions Inventory. The inventory covers 86 pollutants from point, area, and mobile sources. The emissions estimates are at the process level, and the data are model-ready. The inventory is compiled according to a regional protocol designed to provide data of consistent quality and format throughout the Great Lakes region. The inventory data are supplied to the NTI; data for the 1993 reporting year were released in the summer of 1998.

AMBIENT AIR AND DEPOSITION MONITORING NETWORKS

Figure IV-1 displays the monitoring sites for all ambient air and deposition monitoring networks that measure Great Waters pollutants of concern in the regions of the Great Waters and throughout the U.S. and parts of Canada.

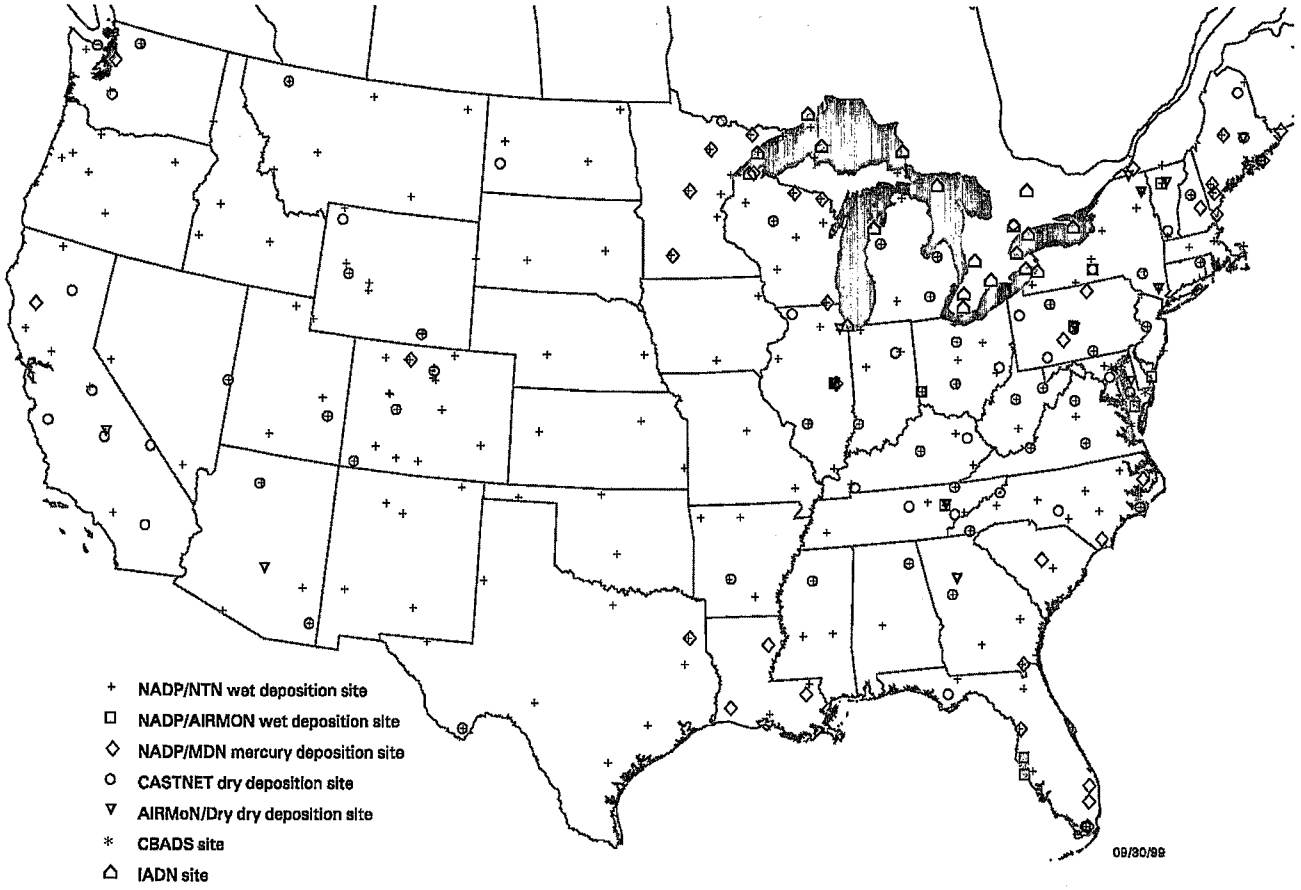
National Atmospheric Deposition Program/National Trends Network

The National Atmospheric Deposition Program (NADP) began in 1978 as a cooperative program between Federal and State agencies, universities, electrical utilities, and other industries to determine geographical patterns and trends in wet deposition of sulfate, nitrate, hydrogen ion, ammonium, chloride, calcium, magnesium, and potassium. The NADP was renamed as NADP/NTN (National Trends Network) in the mid-1980s when the program had grown to almost 200 monitoring sites (Figure IV-1). The monitoring sites are located in rural areas, and data are collected on a weekly basis. The collected data provide insight into natural background levels of pollutants. The network of NADP/NTN monitoring sites allows for the development of concentration and wet deposition maps to describe the trends and spatial patterns in the constituents of acid precipitation. The Mercury Deposition Network (MDN), which is another component of the NADP, measures mercury levels in wet deposition at over 40 NADP sites (U.S. EPA 1998l). The data that are currently available from NADP/NTN and NADP/MDN are presented in Chapter II to characterize the deposition of nitrogen and mercury, respectively.

Clean Air Status and Trends Network

The Clean Air Status and Trends Network (CASTNet) was initiated in 1987 to estimate dry acidic deposition, to provide data on rural ozone levels, and to determine the effectiveness of national emission control programs. The CASTNet is comprised of about 70 monitoring stations across the U.S. (Figure IV-1). Data on atmospheric concentrations of sulfate, nitrate, ammonium, sulfur dioxide, and nitric acid are collected weekly, and ambient ozone concentrations are collected hourly. Most of the stations are operated by EPA's Office of Air and Radiation; 19 stations are operated by the National Park Service in cooperation with EPA (U.S. EPA 1998l).

Figure IV-1
Monitoring Sites of Ambient Air and Deposition Monitoring Networks Measuring Pollutants of Concern



Does not display NADP/MDN mercury deposition monitoring site at St. Anicet in Canada.

Atmospheric Integrated Research Monitoring Network

Established in 1992, the Atmospheric Integrated Research Monitoring Network (AIRMoN), which is sponsored by the NOAA Air Resources Laboratory, conducts research on both wet and dry deposition based on measurements taken at a subset of NADP and CASTNet sites (Figure IV-1). The primary AIRMoN objectives are determining the effectiveness of emission controls mandated by the CAA, evaluating potential impacts of new sources of emissions on protected areas, and identifying source/receptor relationships in atmospheric models. One notable distinction of the AIRMoN-wet network is that sampling is conducted on a daily basis, whereas NADP/NTN data are collected on a weekly basis. Daily sampling allows for the application of source-receptor models to individual storm systems and reduces the storage artifact for ammonium. The AIRMoN-dry network has yielded direct measurements of dry deposition that have been used to support inferred dry deposition measurements at other NOAA and CASTNet stations. In the future, AIRMoN data will also be used to characterize spatial variability around current deposition stations, which will improve NOAA's ability to construct regional deposition loadings estimates.

Integrated Atmospheric Deposition Network

The Integrated Atmospheric Deposition Network (IADN) is a joint U.S.-Canada program begun in 1990 under a formal 6-year implementation plan. The IADN's mandate, which is derived from Annex 15 of the Canada-U.S. Great Lakes Water Quality Agreement of 1987, is to determine atmospheric loadings of toxic substances, including Great Waters pollutants of concern, to the Great Lakes and their temporal and spatial trends. The IADN collects data that can be useful in assessing the relative importance of atmospheric deposition. Under IADN, trends in pollutant concentrations in air and precipitation are assessed and loading estimates of atmospheric deposition and volatilization of pollutants are made every 2 years. The IADN network currently consists of one master station per Great Lake and 14 satellite stations (Figure IV-1). Stations are located in remote areas and do not assess urban sources of pollution. The satellite stations do not meet the original siting criteria but are subject to quality assurance and quality control requirements (U.S./Canada IADN Scientific Steering Committee 1998).

Detailed results from IADN are presented in Chapter II by pollutant group. General conclusions include the following:

- Levels in air and precipitation appear stable for current-use pesticides such as endosulphan, but levels for most other pesticides, PCBs, and lead are decreasing;

- Gas absorption appears to be the dominant deposition process for delivering semi-volatile organic compounds (SVOCs), including PCBs and PAHs, to lake surfaces, while wet and dry deposition dominate for the trace elements and higher molecular weight PAHs;

- For some IADN substances, like dieldrin and PCBs, the surface waters are behaving like a source since the amount that is volatilizing from the water is greater than the amount being deposited to the water;

- The lakes are sensitive to the atmospheric concentration of IADN chemicals, and this points out the fragility of these resources given that long-range transport from other regions may be a significant source of toxic pollutants; and,

- Air trajectory analyses indicate that many SVOCs are potentially originating from outside the Great Lakes basin, whereas trace metals and PAHs may be associated with local sources (U.S./Canada IADN Scientific Steering Committee 1998).

In 1998, the Second Implementation Plan for 1998 to 2004 was developed based on a review of the program from 1990 to 1996. No major changes are anticipated under the Second Implementation Plan. The IADN will continue surveillance and monitoring activities, related research, and provision of information for intergovernmental commitments and agreements. Additional work to be completed under the Second Implementation Plan is the development of a database for all U.S. and Canadian data. Potential modifications will be discussed in relation to the placement of satellite stations to assess urban inputs and air-water gas exchange, criteria for changes to the IADN chemical list, coordination with other research activities, quality assurance and control of IADN operations, and communication of IADN results (U.S./Canada IADN Scientific Steering Committee 1998, U.S. EPA 1998m).

Chesapeake Bay Atmospheric Deposition Study

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. In June 1990, CBADS began collecting data from three rural (i.e., at least 50 km from urban areas) shoreline monitoring sites at Wye Institute and Elms Institute, Maryland, and Haven Beach, VA. Measured parameters included wet and dry deposition and gas exchange of elements (including lead and cadmium), individual PAHs, and total PCBs. Results of data collected from June/July 1990 to the end of 1991 were reported in the *1994 Chesapeake Bay Basin Toxics Loading and Release Inventory* (1994 TLRI) (Chesapeake Bay Program 1994) and in the *Second Report to Congress* (U.S. EPA 1997b). The 1994 TLRI results did not include any data on mercury or current-use agrichemicals, nor did it provide information on wet deposition to urban areas in the Chesapeake Bay watershed. An additional 21 months of CBADS wet deposition data were collected and reported in the 1999 TLRI (making the total study period from June/July 1990-September 1993). This new information on Great Waters pollutants of concern is presented in Chapter II by pollutant group.

Other atmospheric deposition studies were also reported in the 1999 TLRI, including data collected for wet deposition and loadings of mercury and agrichemicals; however, only initial data were available at the time of the report generation to estimate wet deposition loadings to urban areas. For the 1999 TLRI, urban wet deposition fluxes of metals, PAHs, PCBs, and mercury were assumed to be enriched two-, four-, ten-, and two-fold over regional background levels, respectively. Analysis of monitoring data collected by CBADS and other studies not included in the 1999 TLRI will be available for the next report.

Air Toxics Monitoring

The EPA is currently developing a concept for an air toxics monitoring network and beginning implementation. Ambient air toxics data would be useful to characterize ambient concentrations and deposition in representative areas, to provide data to support and evaluate dispersion and deposition models, and to establish trends and evaluate the effectiveness of strategies to reduce air toxics emissions. The goal is to build on monitoring already in place in State, local, and tribal programs as well as other national networks. For example, in the near future, fine particulate matter speciation monitors will provide ambient air measurements of several air toxic metals (including lead and cadmium compounds) at over 50 urban locations in the country. Rural and remote monitoring of these metals takes place as part of EPA's efforts to assess regional haze. As the air toxics network is phased in, the pollutants to be monitored are expected to include several of the compounds of concern to the Great Waters, such as

mercury, POM, and metals. Information about the monitoring strategy can be found at www.epa.gov/ttn/amtic/airtxfil.html.

The EPA has also begun a research effort for ambient air monitoring of dioxin and dioxin-like compounds called the National Dioxin Air Monitoring Network (NDAMN). It is designed with several objectives:

- To provide data useful for calibrating regional-scale, long-range transport models used in estimating air concentrations of dioxin as a function of dioxin source emissions;
- To provide air monitoring capability for the occurrences and levels of dioxin-like compounds in areas where animal feeds (used to feed domestic livestock) are primarily grown;
- To provide for the long-term monitoring of dioxin-like compounds in different regions of the U.S. and over different seasons; and,
- To provide data on potential transboundary import of dioxins and furans into the U.S.

The network is being developed in phases. Phase 1 consists of up to 20 air monitoring stations; Phase 2 is expected to consist of about 30-40 stations. Additional information about the network can be found at www.epa.gov/nceawww1/1page.htm.

OTHER ENVIRONMENTAL MONITORING NETWORKS AND DATABASES

The EPA's Environmental Monitoring and Assessment Program (EMAP)

In 1988, the EPA Science Advisory Board (SAB) recognized a deficiency in the documentation of the status of the Nation's natural environment and recommended that EPA develop a program to monitor ecological status and trends in the U.S. The EPA responded with the initiation of the Environmental Monitoring and Assessment Program (EMAP) in 1989. The EMAP program also collaborates with other agencies, including NOAA.

The primary goal of EMAP is to "monitor the condition of the Nation's ecological resources, to evaluate the cumulative success of current policies and programs, and to identify emerging problems before they become widespread or irreversible" (U.S. EPA 1997i, j, k). Knowledge from the EMAP process will give decision makers the ability to make informed environmental management decisions, set rational priorities, and make known to the public the costs, benefits, and risks of

Condition of the Mid-Atlantic Estuaries

The EPA's Office of Research and Development and the Environmental Monitoring and Assessment Program (EMAP) issued a report, *The Condition of the Mid-Atlantic Estuaries Report*, that is the first in a series of State-of-the-Region Reports for the Mid-Atlantic. The report discusses the adequacy of current protective measures, the present condition of natural resources, and the extent and possible causes of ecological problems. These issues are addressed through the evaluation of the best available scientific information, including data from large sampling programs throughout the estuaries, and use of the latest scientific tools. Although the Mid-Atlantic estuaries are being impacted by human activities, active management by the States and EPA has had positive results. Future State-of-the-Region Reports for the Mid-Atlantic will address streams, forests, and other resources (U.S. EPA 1998b).

proceeding or refraining from implementing specific regulatory actions. Four operational objectives guide EMAP:

1. Estimate the current status, trends, and changes in selected indicators of the Nation's ecological resources on a regional basis with known confidence;

2. Estimate the geographic coverage and extent of the Nation's ecological resources with known confidence;

3. Seek associations between selected indicators of natural and anthropogenic stresses and indicators of ecological resources; and,

4. Provide annual statistical summaries and periodic assessments of the Nation's ecological resources.

These four objectives create an innovative approach for EMAP, which adopts a comprehensive, multimedia perspective of the Nation's natural resources rather than the traditional single-pollutant or single-location approach to environmental assessment.

The EMAP's strategy also includes the development of a Regional Environmental Monitoring and Assessment Program (R-EMAP) to test the effectiveness of the EMAP approach on answering questions about regional and local ecological conditions. The R-EMAP has three main objectives:

- To evaluate and improve EMAP concepts for State and local use;

- To assess the applicability of EMAP indicators at differing spatial scales; and,

- To demonstrate the utility of EMAP for resolving issues of importance to EPA Regions and States.

Since 1990, EPA has funded at least two R-EMAP projects in each of the ten EPA Regions. One of these studies addressed mercury deposition levels and geographic trends in the Great Lakes region, as mentioned in Chapter II.

National Sediment Inventory

The EPA established the National Sediment Inventory (NSI) to prepare the first biennial National Sediment Quality Survey (NSQS) Report to Congress (U.S. EPA 1997i, j, k) (see also page III-8). National-level results from the NSQS on sediment contamination are presented in Chapter II. The NSI is intended to have applications far beyond supporting subsequent NSQS biennial reports. The NSI is the largest set of sediment chemistry and related biological data ever compiled by EPA. In addition to sediment chemistry data, the NSI includes tissue residue and sediment toxicity data.

The NSI includes data from ten existing Federal databases (see sidebar). Minimal data requirements for the NSI include monitoring program, sampling date, latitude and longitude coordinates, and measured units. Additional data fields such as sampling method and other quality assurance/quality control information are included when available. At present, the NSI contains approximately 2 million records for more than 21,000 monitoring stations across the country. More than 230 chemicals and chemical groups are included. Only data from 1980 to 1993 were used for the first NSQS. However, older data are available in the NSI.

NSI Federal Databases

1. EPA's Storage and Retrieval System (STORET)
2. NOAA's Coastal Sediment Inventory (COSED)
3. EPA's Ocean Data Evaluation System (ODES)
4. EPA Region IV Sediment Quality Inventory
5. The Gulf of Mexico Program's Contaminated Sediment Inventory
6. EPA Region X/USACE Seattle District's Sediment Inventory
7. EPA Region IX Dredged Material Tracking System (DMATS)
8. EPA's Great Lakes Sediment Inventory
9. EPA's Environmental Monitoring and Assessment Program (EMAP)
10. USGS's Massachusetts Bay Data

Although the NSI provides broad geographic coverage, the 21,000 monitoring stations currently represented in the data are not randomly distributed. Most of the monitoring programs used to compile the NSI are located in areas with known or suspected contaminant impacts.

National Contaminated Sediment Point Source and Non-point Source Inventories

The EPA's mandate to investigate sediment contamination in the Nation's water included a directive to identify potential pollutant sources. With future biennial NSQS Reports to Congress, EPA will inventory point and non-point sources and estimate loadings. The first NSQS Report to Congress evaluated point sources (i.e., direct discharges to waterbodies) only (see also page III-8). The National Contaminated Sediment Point Source Inventory (U.S. EPA 1997k) provided a relative ranking of industrial categories and discharged chemicals for their potential contribution to sediment contamination. The EPA developed the "load score," a unitless index of the magnitude of potential sediment contamination based on chemical/facility-specific releases, physical and chemical properties, and potential environmental risks. A screening analysis of the load scores indicated that the point sources most likely to contribute to sediment contamination were sewerage systems, metals products and finishing, primary metals industries, industrial organic chemicals, public utilities, petroleum refining, and other chemical products. Metals were associated with higher load scores than other contaminant groups because point source releases are more prevalent. Additional analyses are needed to assess the bioavailability and toxicity of metals in sediments.

Although the NSQS did not inventory specific non-point sources, it examined the land uses in watersheds containing areas of probable concern (APCs) to identify potential relationships between sediment contaminants and human activities. In general, EPA found that diversified land uses were associated with diversified pollutants. However, a high percentage of agricultural land use corresponded with markedly higher contamination from pesticides. Most chemical classes increased with higher percentages of urban land use. The analysis suggested that mercury and PAHs in urban sediments may be attributable to atmospheric deposition from local sources.

Although the National Contaminated Sediment Point Source Inventory identified metals as the leading contaminants based on load scores, the NSQS identified PCBs, mercury, pesticides, and PAHs as most often responsible for contamination levels of concern in sediment (see also page II-73). This

suggests that non-point sources and historical releases (including atmospheric deposition) contribute largely to contamination in the Nation's waters.

NOAA's National Status and Trends Bioeffects Assessments

The National Status and Trends (NS&T) Program under NOAA (see page II-70) conducts bioeffects assessment studies, which include sediment toxicity surveys and the development of effect-based numerical guidelines for use in evaluating the toxicological relevance of sediment contamination, among other factors. The sediment toxicity surveys are conducted in coastal areas where data from NS&T's Mussel Watch and Benthic Surveillance Projects indicate the potential for substantial environmental degradation and biological effects. Bioeffects assessments have been or are being conducted in 16 coastal areas. Reports and data sets are available on the Internet for several of these locations at http://seaserver.nos/noaa.gov/projects/bioeffects/page1.html.

ENVIRONMENTAL TRANSPORT AND FATE

Available Environmental Transport and Fate Models

Models-3 Community Multi-Scale Air Quality (CMAQ) Modeling System

The Models-3 community multi-scale air quality (CMAQ) modeling system is a flexible software system designed to simplify the development and use of environmental assessment and decision support tools for applications ranging from regulatory and policy analysis to understanding the interactions of atmospheric chemistry and physics. This newest generation of environmental modeling software has been under development for the past 7 years. Models-3, in combination with CMAQ, form a third generation air quality modeling and assessment system. First generation air quality models dealt with tropospheric air quality with simple chemistry at local scales using Gaussian plume formulation (i.e., the model assumes that the plume spreads from an emission source laterally and vertically in accordance with a Gaussian distribution) as the basis for prediction. Second generation models covered a broader range of scales (i.e., local, urban, regional) and pollutants, addressing each scale with a separate model and often focusing on a single pollutant. Third generation models treat multiple pollutants simultaneously up to continental scales and incorporate feedback between chemical and meteorological components. Models-3 has a unique framework and science design that enables scientists and regulators to build their own modeling systems to suit their needs. The CMAQ system has capabilities for urban to regional-scale air quality simulation of tropospheric ozone, acid deposition, visibility, toxics, and fine particles. The Models-3 framework contains components that assist the model developer with creating, testing, and performing comparative analysis of new versions of air quality models and enables the user to execute air quality simulation models and visualize the results. The overall goal of Models-3 is to simplify and integrate the development and use of complex environmental models, beginning with air quality and deposition models (U.S. EPA 1998e). It is expected that the Models-3 framework and CMAQ will be useful tools for research related to the Great Waters program.

The initial public release of the Models-3 framework occurred in June 1998. This framework provides a unifying foundation for continued evolution of environmental modeling and assessment tools with the ability to extend the capabilities beyond the current air quality implementations. One area of investigation has been a basic integration of the Chesapeake Bay Water Quality Model (CBWQM) into the Models-3 framework to explore multimedia model linkages. Future development toward a fourth generation system will extend linkages and process feedback to include air, water, land, and biota to

provide a more holistic approach to simulation of transport and fate of chemicals and nutrients throughout an ecosystem (U.S. EPA 1998e).

Total Risk Integrated Methodology (TRIM)

The Total Risk Integrated Methodology (TRIM) is a comprehensive framework for characterizing human health and ecological risk. It is being designed by EPA's Office of Air Quality Planning and Standards (OAQPS) for use in CAA programs (e.g., the Residual Risk Program) as a way to consistently estimate the impacts of air pollutants. The TRIM will have the following characteristics: (1) ability to perform multimedia assessments; (2) ability to perform human health and ecosystem risk assessments; (3) ability to perform multipollutant assessments; (4) capability to address uncertainty/variability; and, (5) ability to provide a readily available, user-friendly tool that will assist in risk management decisions (U.S. EPA 1999e).

The EPA is attempting to create a system that is scientifically defensible, flexible, and user friendly. The format of input data sets to TRIM are consistent with Models-3. When complete, TRIM will contain three separate modules, including (1) fate and transport, (2) exposure, and (3) risk characterization. The most complex module is the multimedia fate and transport module, which is entitled TRIM.FaTE. This is the module on which EPA has been concentrating most of its effort (U.S. EPA 1999e).

The TRIM.FaTE module is a multimedia, chemically mass-balanced model. In the model, the ecosystem scale of interest (landscape) is divided into compartments. The model tracks the mass of the pollutant transported between compartments (rather than a set of one-way fate algorithms). The model is being designed so that a wide variety of pollutants can be addressed. The TRIM.FaTE module will have the capability to (1) simulate steps in a time series and (2) resolve mass distribution spatially. A prototype version of TRIM.FaTE has been developed by EPA; however, significant work remains on this module, including comparisons of output to monitoring data for case studies and other model evaluation exercises. A version of TRIM, including TRIM.FaTE, is expected to be released in 2000 (U.S. EPA 1999e).

Models for Atmospheric Mercury

Because of the need to identify the emission reductions required to meet water quality and other criteria, systems are being developed that use emissions data with atmospheric chemistry and transport models and meteorological models. A report, *A Computer-Based Framework to Model Acceptable Loadings of Mercury to the Atmosphere to Protect Water Quality*, was prepared for EPA Region IV by Nicola Pirrone and Gerald J. Keeler, at the University of Michigan (Pirrone and Keeler 1997). This report discusses the coordination of available computer models for atmospheric mercury – its transport, chemical-physical dynamics, and deposition (wet and dry) to water surfaces and terrestrial receptors. The analysis of model capabilities and critical data inputs is applicable nationwide. In addition, some specific discussions are provided for the South Florida Everglades in comparison to forested watershed and lake systems.

This report presents a framework linking models that relate data on mercury emissions into the air to subsequent mathematical modeling of atmospheric transport, chemistry, and deposition processes (Pirrone and Keeler 1997). The modeling framework is designed so that regulatory agencies can define target load reductions from local and regional sources that are needed to meet water quality goals. In order to correctly relate water quality criteria and bioaccumulation of mercury in the food chain to the atmospheric and other inputs, this modeling framework needs to be coupled to an aquatic modeling system utilizing mercury transport and fate processes in similar timeframes. The set of models in the

framework will need inputs including watershed-specific information or default assumptions and chemical species released from the major pollutant sources, along with meteorological data and mesoscale models (such as RAMS from Colorado State University or MM5 from Pennsylvania State University). This conceptual framework of models is being used as part of the design and analyses in a State/EPA pilot project to develop a TMDL for mercury in a specified section of the Florida Everglades, as discussed in Chapter III.

The Electric Power Research Institute (EPRI) has developed and continues to develop environmental mercury cycling models (MCMs) to assess the transport and fate of atmospheric mercury (Watras and Huckabee 1994, Leonard et al. 1995, Logan 1998, Pai et al. 1999). The MCMs have been used to predict the fate of mercury in the Great Lakes (Leonard et al. 1995). The Maryland Department of Natural Resources reviewed two EPRI models, the Lake MCM (L-MCM) and the Regional MCM (R-MCM), and concluded that they had the potential for use in Maryland to investigate cycling of environmental mercury and bioaccumulation in fish (Logan 1998). The L-MCM was developed for predicting the biogeochemical behavior of mercury in the Mercury in Temperate Lakes Study of seepage lakes in Wisconsin. The L-MCM is a dynamic, mechanistic simulation model that tracks processes and changes in concentrations through time. The R-MCM is a steady-state, mechanistic simulation model.

Recent Environmental Transport And Fate Modeling Results

Atmospheric Exchange Over Lakes and Oceans Surfaces (AEOLOS)

The Atmospheric Exchange Over Lakes and Oceans Surfaces (AEOLOS) project, administered through the EPA Great Lakes National Program Office and the Office of Research and Development, was designed to study atmospheric deposition in the Great Waters. Begun in 1993 by EPA and scientists from the Universities of Minnesota, Michigan, Maryland, Delaware, and the Illinois Institute of Technology, the project's objectives were to determine (1) the dry depositional fluxes of critical urban contaminants to the northern Chesapeake Bay near Baltimore and southern Lake Michigan near the Chicago urban area, (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 (U.S. EPA 1997b). The AEOLOS project proceeded under the hypothesis that emissions of HAPs into the coastal urban atmosphere enhance atmospheric depositional fluxes to the adjacent Great Waters such as Lake Michigan in the vicinity of Chicago and Gary, Indiana, and the Chesapeake Bay near Baltimore (Simcik et al. 1997). Results of some AEOLOS studies are reported in Chapter II for PAHs and PCBs as well as below.

A study under the AEOLOS project found that urban air emissions have an effect on the average coastal atmospheric concentrations of PAHs and PCBs above continental background, increasing them by factors of 12 and 4, respectively. As part of AEOLOS, air concentrations of PCBs and PAHs were measured in the urban/industrial complex of Chicago, over the southern portion of Lake Michigan, and in a non-urban location, in May and July 1994 and January 1995. Gas-phase PAH and PCB concentrations over the lake were much lower than urban air concentrations. The highest concentrations were associated with winds that first crossed-over the urban/industrial area from Evanston, Illinois to Gary, Indiana. Concentrations were near regional background for winds from any other direction. Seasonal variation of PCB occurred, with volatilization being higher during the summer (Simcik et al. 1997).

In another AEOLOS study in the Chicago area, PCB concentrations in wet precipitation collected over southern Lake Michigan ranged from two to as high as 400 times greater than the measured regional background concentration, indicating that the "urban plume" of Chicago increases atmospheric deposition of contaminants to Lake Michigan over tens of kilometers. Concentrations of PCBs in urban

precipitation were dominated by particle-bound congeners, indicating PCB enrichment in rainwater due to efficient scavenging of contaminated particulate matter in the urban atmosphere (Offenberg and Baker 1997).

Simcik et al. (1998), under the AEOLOS project, investigated gas-particle partitioning of PCBs and PAHs in the Chicago urban area and over Lake Michigan and found that PCBs and PAHs in the Chicago/Lake Michigan atmosphere were at equilibrium between the gas and particle phases. Understanding gas-particle partitioning is important because it can be used to help determine the mode of atmospheric deposition (i.e., wet deposition, dry deposition, air-water exchange).

Chesapeake Bay NO_x Modeling with RADM

The Regional Acid Deposition Model (RADM) has been used to develop estimates of the primary airshed of NO_x emissions that are contributing nitrogen deposition to the Chesapeake Bay watershed. The RADM was originally developed to address policy and technical issues associated with acidic deposition. The model is designed to provide a scientific basis for predicting changes in deposition resulting from changes in precursor emissions, to predict the influence of sources in one region on acidic deposition in other sensitive receptor regions, and to predict the levels of acidic deposition in certain sensitive receptor regions (Dennis 1997).

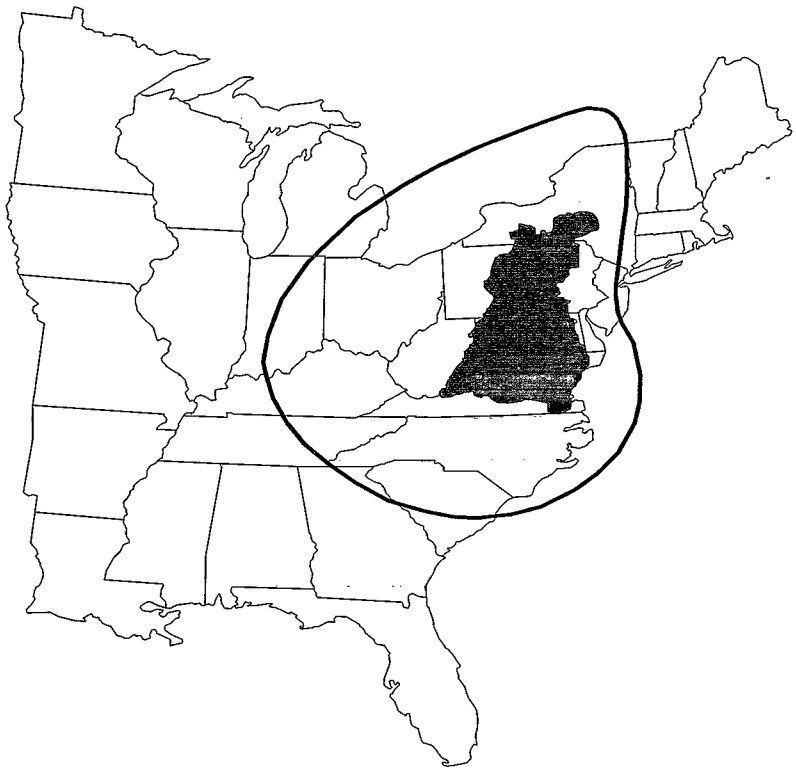
In the RADM, the concentrations of gaseous and particulate species are calculated for specific fixed positions in space as a function of time. The geographic extent, or domain, of the model is 2,800 by 3,040 km and extends south from James Bay in Canada to the southern tip of Florida and westward to central Texas. This domain is partitioned into 80 km-by-80 km grid cells. The space is three-dimensional, having 15 logarithmically-spaced vertical layers, covering the distance from ground level to 16 km in altitude. The RADM has a chemistry component that consists of 140 reactions among 60 species. Chemical decomposition by solar radiation and aqueous-phase reactions that occur in clouds are both included in the model's chemistry. For each grid cell, predictions are generated at dynamically determined time steps of seconds to minutes that are output hourly by the model. The species predicted by RADM include ambient concentrations (SO_2, NO, NO_2, HNO_3, O_3, H_2O_2, NH_3, PAN, HCHO, CO, and $SO_4^=$), wet deposition ($SO_4^=$, NO_3^- as HNO_3, NH_4^+ as NH_3, and H^+), and dry deposition (SO_2, $SO_4^=$, HNO_3, O_3, and NO_2). The meteorological fields that are used to determine the turbulent motion of the atmosphere, which in turn affects the transport of species by wind into and out of the grid in three dimensions, are from the Pennsylvania State University, National Center for Atmospheric Research Mesoscale Model (MM4). The MM4 is a weather model used to recreate historical meteorology.

Recent analysis with RADM shows that the range of influence of nitrogen emissions is similar to the range for sulfur emissions – on the order of 600 - 800 km. However, results suggest that the nitrogen range given is too short (Dennis 1997). This implies the range for nitrogen should be longer than that for sulfur. In other words, the RADM results and analysis used to define the airshed for the Chesapeake Bay watershed is producing an estimate of the boundary of the airshed that is conservative, that is, one that is too small.

In the original 1995 study (Dennis 1997), the extent of the Chesapeake Bay airshed was approximately 900,000 km^2, or more than 5½ times larger than the watershed. The NO_x emissions in the airshed account for approximately 75 percent of the anthropogenic nitrogen deposition to the Chesapeake Bay watershed. More recently, using the same methodology, Dennis conducted a more refined analysis of the subregions within the RADM domain, resulting in a revised estimate of the Chesapeake Bay airshed. This revised airshed, presented in Figure IV-2, is approximately 1,081,600 km^2, making it about 6½ times larger than the watershed (see http://www.chesapeakebay.net/data/model/mod_gis/air_dom.gif). This larger airshed estimate results in only a slight increase (76 percent) of the amount of

NO_x emissions from the airshed which are deposited to the watershed. The contribution from utility and mobile sources are roughly equal and make up the majority of the emissions which are eventually deposited. However, when model simulations are used to compare these emission sources to nitrate deposition a significant pattern emerges. Utilities, which are heavily concentrated to the west of the Bay, tend to contribute a majority of the nitrate that deposits on the western side of the watershed. Moving across the watershed from west to east, nitrate deposition from utility emissions shows a decreasing trend. In contrast, a large amount of the mobile source emissions take place in cities and heavily developed areas relatively close to the Bay. Thus, mobile emissions tend to contribute a majority of the nitrate that deposits along the Delmarva Peninsula, the Chesapeake Bay itself, and the lower portions of the western shore tidal tributaries. Nitrate deposition from mobile emissions shows a decreasing trend moving from east to west across the watershed (Dennis 1997).

Figure IV-2
Revised Principal Airshed for the Chesapeake Bay



(Developed by R. Dennis, Atmospheric Sciences Modeling Division: ARL, NOAA, and NERL U.S. EPA)

Lake Michigan Mass Balance Study

The Lake Michigan Mass Balance Study (LMMBS) was developed and is being implemented by the Great Lakes National Program Office in cooperation with five U.S. Federal agencies (i.e., Department of Energy, NOAA, U.S. Geological Survey, U.S. Fish and Wildlife Service, U.S. Army Corps of Engineers), one foreign agency (i.e., Environment Canada), two State agencies, and eight academic institutions. The LMMBS was initiated to address the objectives of the Lake Michigan Monitoring Program, as well as to assist EPA in implementing section 112(m) of the CAA by characterizing the loadings, transport, and fate of selected pollutants through monitoring and modeling. The *Second Great Waters Report to Congress* describes the project in more detail and describes the

atmospheric monitoring efforts, a system of ten stations situated around Lake Michigan. Additional information can be found at www.epa.gov/glnpo/lmmb. The ultimate goal is to develop a predictive model that forecasts impacts on lakes based on inputs of toxics to the system from a variety of sources (sediments, atmospheric deposition, tributaries). Preliminary modeling results for mercury are presented in Chapter II. Additional results are expected in 2000 that will support, in addition to section 112(m), such efforts as the Binational Toxics Strategy and LaMPs.

The LMMBS model is constructed for a limited group of pollutants: PCBs, chlordane, total mercury, and atrazine. For mercury and atrazine, an emissions inventory either existed or could be developed, so comprehensive modeling is being conducted for these two pollutants. For PCBs and chlordane, a comprehensive modeling approach was not attempted because a comprehensive emissions inventory did not exist. In addition, monitoring data were collected for an extended suite of chemicals, including additional pesticides, PAHs, and heavy metals. Currently, a multimedia database is being developed to house all the data and quality assurance information for use by the modelers. In the future, the data will be available upon request.

Atmospheric loadings of the LMMBS pollutants are being calculated for input into the models. A major finding of the LMMBS to date is that traditional atmospheric loading estimation techniques are not adequate to describe the variability and source influence on deposition to the lake. New techniques have been developed to better characterize the impact of atmospheric deposition on large lake systems. Efforts have also been made to coordinate with ongoing fate and transport modeling activities and to support the most advanced developments. The LMMBS is contributing a major amount of resources to the CMAQ model system (page IV-11) for use in estimating emissions and transport of atrazine and mercury to Lake Michigan. In addition, this project has assisted in the development of an atrazine soil emissions model that could also be used to develop emissions inventories in other regions.

Recent REMSAD Applications

The Regulatory Modeling System for Aerosols and Deposition (REMSAD) was originally developed in part to address the issue of atmospheric deposition of HAPs to the Great Waters. The REMSAD is a grid model designed for use on workstation-class computers. As currently configured, REMSAD (version 4.0) tracks the transport, transformation, and deposition (both wet and dry) of nitrogen, mercury, cadmium, dioxin, polycyclic organic matter (POM), atrazine, speciated primary and secondary fine particles, photochemical oxidants, acids, and ammonia. All of these substances are treated simultaneously for a single integrated emission inventory, so that the effects of proposed policy options on multiple pollutants with common sources may be analyzed at the same time. The model extends vertically all the way to the tropopause, so that long-range transport of pollutants in the upper troposphere can be captured.

The model is capable of nesting finer (i.e., higher resolution) grids within an overall coarse grid, which makes it possible to focus on deposition to a particular watershed within the context of a broad regional distribution of emission sources. The EPA recently sponsored the 1990 Base Case Evaluation Study (Guthrie et al. 1999a, b) in which the National Emission Trends 1990 inventory was used, along with a calendar 1990 meteorological data set produced by the Interagency Working Group on Air Quality Models (IWAQM). This model run was conducted for a full year (less 5 days) for the contiguous U.S. with adjacent portions of Canada and Mexico. The run was carried out on a coarse grid of approximately 60 km resolution, but included seven nested sub-grids at approximately 20 km resolution. These nested sub-grids focused on the eastern seaboard (especially around the Chesapeake Bay), the Lake Michigan-Lake Superior region, the northern Gulf of Mexico, Puget Sound, southern Florida, and the Houston-Galveston region of Texas.

The annual deposition pattern of total wet and dry nitrogen deposition (as nitrate) as simulated in the 1990 Base Case is very similar to that reported by Dennis (1997) for the eastern U.S., given differences in emissions. The REMSAD results indicate a significant seasonal variation, however, especially in the wet deposition fraction, as might be expected. The pattern of mercury deposition from the same model run is similar overall to that seen in the RELMAP simulations reported in the *Mercury Study Report to Congress* (U.S. EPA 1997e), but the localized "hot spots" are more prominent.

In another context, REMSAD has been used in a study to assess the most efficient combination of NO_x controls designed to reach specified reduction levels in nitrogen reaching the Chesapeake Bay via atmospheric deposition. In this application, the total reduction desired is held fixed, but the combination of air quality health benefits attributable to NO_x in various locations is combined with specific costs of NO_x reductions in those locations. This forms the basis for both a least-net-cost analysis and for a potential emission trading approach based on spatially differentiated values for NO_x emission reductions (Krupnick et al. 1998).

Recent Environmental Transport and Fate Research

The Fate of Mercury in the Lake Superior Basin

This project, which began in 1998, is designed to examine mercury sources and deposition in the Lake Superior basin, particularly the contribution of a coal-fired power plant to the mercury loading of local and regional ecosystems. The research is being conducted at the Minnesota Power and Light's Clay Boswell Station and at the Wisconsin Electric Power Company's Presque Isle Power Plant and is expected to answer questions about what forms of mercury are emitted, and how much mercury from the power plant is deposited locally compared to regionally. The overall goal of the project is to determine the fate of anthropogenic mercury in the Lake Superior region, thereby allowing the prediction of the effects of mercury reduction strategies on the bioaccumulation of methylmercury in fish. The project is expected to be completed at the end of 2000.

Sources and Impacts of Nutrient Enrichment in the Gulf of Mexico

Nutrient enrichment in the northern Gulf of Mexico is responsible for one of the largest zones of oxygen-deficient bottom waters in the western Atlantic Ocean. To address this problem, the White House's Committee on Environment and Natural Resources (CENR) established a multiagency scientific team to review the extent and causes (including air pollution sources) of hypoxic waters, the ecological and economic impacts of hypoxia on the gulf, and potential solutions. Specifically, the multiagency team identified six topics of study:

1. Distribution, dynamics, and causes of hypoxia in the gulf;
2. Ecological and economic consequences of hypoxia in the gulf;
3. Sources and loads of nutrients to the gulf from the Mississippi River;
4. Effects of reducing nutrient loading to the Mississippi River and the gulf;
5. Methods to reduce nutrient loads; and,
6. Social and economic costs and benefits of nutrient reduction strategies.

The scientific assessments for the above six topics of study are complete and available at www.nos.noaa.gov/products/pubs_hypox.html. The integrated assessment, which will address all six topics together, is currently under review and will be available later in 2000. These assessments will be used by the Gulf of Mexico Program, State agencies, and other involved agencies to combat nutrient enrichment and hypoxia and measure progress toward the goal of a restored Gulf of Mexico ecosystem.

Pollutant Exchange Mechanisms

Many pollutants, depending on their specific chemical and physical properties, can be transferred between environmental media. In recent years, research has focused primarily on the exchange of pollutants between air and water. The major air-water transfer processes include wet and dry atmospheric deposition, gaseous exchange, bubble stripping and bursting, and spray transfer (Gustafson and Dickhut 1997). In 1996, Hoff et al. reported on revisions to the estimates of atmospheric inputs of 11 organochlorine (OC) chemicals, five trace elements, and four PAHs to the Great Lakes based on IADN data. Calculations include the flux for wet deposition, dry deposition, and vapor transfer across each of the lakes, with highest confidence in the estimates for wet deposition and lowest confidence in the estimates for gas transfer components. Polychlorinated biphenyls, dieldrin, HCB, DDE, phenanthrene, and pyrene all showed net losses from the lakes to the atmosphere via volatilization, while p,p'-DDT was loaded into the lakes from the atmosphere. Alpha- and γ-HCH were near equilibrium with the waterbodies and the atmosphere, with seasonal changes for α-HCH. These results show that the waters of the Great Lakes are close to long-term equilibrium with the atmosphere for most of these chemicals, but that equilibrium is in a constant state of short-term seasonal displacement and adjustment (Mackay and Bentzen 1997). There is a need, however, for integrated assessments of air-water transfer (i.e., measuring both water and air simultaneously and over widely varying conditions) for each lake to obtain reliable estimates of distribution coefficients, deposition velocities, and loadings from other sources.

More recently, Gustafson and Dickhut (1997) quantified the gaseous exchange fluxes for PAHs across the air-water interface of the southern Chesapeake Bay and demonstrated that, for individual PAHs, different particle characteristics influenced particle-gas distributions at the urban and rural sites. Atmospheric PAH concentrations were measured at four sites that were characterized as rural, semi-urban, urban, and industrialized. Exponential increases in gaseous PAH concentrations with temperature were observed at the non-rural sites, which the authors suggested was volatilization from contaminated surfaces during warmer weather. The PAH gas concentrations at the rural site exhibited little seasonal variability. Aerosol particle-associated PAH levels were similar at all sites and increased in winter. This was attributed to the temperature dependence of particle-gas partitioning, including the cold condensation of gases to background aerosols as air masses were dispersed from source areas to remote regions, and also increased emissions from combustion of fossil fuel and wood for home heating during the wintertime. Indications were that either PAH partitioning is not at equilibrium or that different distribution processes were operating in rural areas of southern Chesapeake Bay.

Nelson et al. (1998) measured the dissolved- and gas-phase concentrations of nine PAHs and 46 PCB congeners at eight sites on the Chesapeake Bay at four different times of the year to estimate the diffusive exchange of gaseous PAHs and PCBs across the air-water interface. They found that PAH fluxes varied both temporally and spatially in the Chesapeake Bay. Fluxes were usually larger in the northern bay as a result of higher gaseous concentrations. Gaseous PAHs were absorbed into the bay's surface waters during the spring, and lighter compounds revolatilized in the late summer and early fall due to seasonal changes in surface water temperature and atmospheric PAH levels. On an annual basis, Nelson et al. found that the atmosphere is a net source of volatile PAHs to the bay and that gas absorption may be the largest external source of fluorene and phenanthrene, providing up to three times the combined loadings from wet and dry aerosol deposition and from the tributaries. In contrast to PAHs, PCBs volatilized from Chesapeake Bay throughout the year, with the largest fluxes occurring in

September due to high dissolved concentrations and warmer water, while the smallest fluxes occurred during stratification in June when both dissolved phase concentrations and wind speeds were low.

A study by Ridal et al. (1997) found that as much as 60 percent of the α-HCH in the air above Lake Ontario was derived from the lake itself. Another study of air and water samples from the Bering and Chukchi Seas and on a transect across the polar ice cap to the Greenland Sea concluded that soils and surface waters containing HCH will be a diffuse, non-point source to the atmosphere that will likely maintain detectable atmospheric concentrations for some time into the future (Jantunen and Bidleman 1996, Li et al. 1998).

Hurley et al. (1998a) investigated partitioning and transport of total mercury and methylmercury in the lower Fox River in Wisconsin and found that resuspended sediments were the predominant source of mercury from the Fox River into the Green Bay. The researchers coupled time series data of total mercury at the river mouth with transect sampling in the Lower Fox River. The researchers reported that concentrations of unfiltered total mercury were significantly elevated compared with other large tributaries to Lake Michigan. The transect sampling revealed progressively increasing water column total mercury concentrations and total mercury particulate enrichment downstream, which the authors suggest were consistent with trends in sediment total mercury levels in the river. Despite elevated total mercury concentrations, Hurley et al. (1998a) reported that methylmercury concentrations were relatively low, suggesting limited bioavailability of total mercury associated with sediments.

The processes that take place in the water surface microlayer, particularly those related to pollutant exchange mechanisms at the air-water interface, are receiving increasing attention in the research literature as one of the most important regions of large surface waterbodies. The surface microlayer is the top 30 to 300 μm of a waterbody where atmospheric pollutants first deposit. Hydrophobic contaminants, such as PCBs, can adsorb at the air-water interface even without previous organic accumulations present due to the lower energy state of the interface. The accumulation of organic matter in the surface microlayer also causes a lowering of surface tension values, resulting in further enrichment of the organic matter in the surface microlayer as compared to the subsurface water. As a result of its organic nature, many pollutants concentrate in the surface microlayer, especially hydrophobic organic contaminants such as PAHs, PCBs, and other pollutants that adhere to particles or exhibit increased solubility with elevated dissolved organic matter. Yet, despite its importance, the surface microlayer is possibly the least understood and poorly characterized region of the aquatic environment.

Liu and Dickhut (1997) compared PAH enrichment in the surface microlayer at an urban site (Elizabeth River) and a semiurban site (York River) in the southern Chesapeake Bay watershed and found particulate PAH concentrations in the surface microlayer of the Elizabeth River to be an order of magnitude higher than in the York River. In comparing the enrichment of PAHs in the surface microlayer relative to subsurface water, total PAH concentrations in the surface microlayer were found to be 1-4,900 times higher. The authors speculated that the difference in PAH concentrations in the surface microlayers of the two rivers was due to a greater contribution of atmospheric deposition of soot-like aerosols to the surface microlayer in the urban Elizabeth River.

Liu and Dickhut (1998) concluded that wind-driven mixing is the principal mechanism that distributes suspended particles collected at the air-water interface into the surface microlayer. The researchers found a significant relationship ($p < 0.05$) between wind speed and the surface microlayer thickness at the York River site. Thicker surface microlayers result in greater enrichments of organic material. Liu and Dickhut (1998) also concluded that enrichment of suspended particles and organic carbon in the surface microlayer is related to the sources of these materials in aquatic ecosystems. In the York River, where *in situ* production of organic matter plays a greater role than runoff and atmospheric

deposition, enrichment of the surface microlayer appeared to be related to the buoyancy and density of the particle types. In contrast, in the Elizabeth River, where productivity is lower, small, dense particles presumably derived from runoff and atmospheric deposition appeared to accumulate in the surface microlayer.

MEASURING AND MONITORING TECHNIQUES

Ambient Mercury

Some of the most important new developments made during the last few years of atmospheric mercury research were reliable methods developed to measure reactive (divalent) gaseous mercury species (Hg^{+2}). The importance of developing a reliable method for measuring ambient Hg^{+2} (in the picogram per cubic meter range) was identified as the highest priority research topic at the expert panel on Mercury Atmospheric Processes convened on March 16-18, 1994 in Tampa, Florida. Researchers from the U.S. and Europe reported observing significant spatial gradients in mercury deposition around urban and industrial areas, indicating local anthropogenic influences. The panel considered improved characterization of mercury species from sources and at impacted receptor locations to be vital to elucidate source-receptor relationships.

Over the past several years, three different methods to measure ambient Hg^{+2} have been proposed, including impregnated filters (Gill et al. 1996), refluxing mist chambers (Lindberg and Stratton 1998), and thermal annular denuders (Stevens et al. 1998a). An instrument utilizing thermal denuder technology recently has been introduced by Tekran Inc. that is capable of continuously measuring both Hg^0 and Hg^{+2} (Stevens et. al. 1998b). Determining the applicability of this new Tekran instrument in establishing a national automated mercury monitoring network was recommended at the October 1998 Tri-lateral Commission for Environmental Cooperation meeting in Las Vegas, Nevada. The EPA's National Exposure Research Laboratory is presently evaluating the accuracy and precision of the available ambient Hg^{+2} measurement technologies in order to determine which may be better for routine monitoring purposes.

A method for collection and analysis of total particulate mercury (Hg_p) was recently published in the EPA *Compendium of Methods for the Determination of Inorganic Compounds in Ambient Air (Method IO-5)*. Air is pulled through a pre-fired glass filter, which is subsequently microwave digested in nitric acid. The filter extract is then analyzed using cold vapor atomic fluorescence spectrometry (CVAFS). A semi-continuous method for Hg_p determination was developed by Lu et al. (1998). Ambient samples are collected onto a quartz filter housed in a quartz chamber. The quartz chamber is then heated to 900EC releasing the Hg_p as elemental Hg^0, which is quantified using CVAFS. Research is currently under way to merge the thermal quartz filter Hg_p technique with the Tekran automated mercury instrument so that the three most prevalent forms of mercury can be measured at ambient concentration levels.

Source-Receptor Linkages

A combination of certain analytical techniques with appropriate modeling approaches has been used on a number of occasions to ascertain sources contributing certain contaminants in receptor matrices. Some of these studies have been conducted in geographic locations outside the Great Waters; however, the methods may prove useful in linking sources and receptors in the Great Waters.

Using a combination of statistical and radioisotope dating techniques, Huntley et al. (1998) found that the majority of dioxin and furan loads in Newark Bay Estuary sediments were attributable to

combustion sources and sewage sludge sources. Fifty sediment cores were collected throughout Newark Bay and analyzed for dioxins and furans. In addition, the cores were dated using radioisotopic techniques. The samples were grouped into three categories based on their estimated dates of deposition, and polytopic vector analysis was performed separately on each group to determine the congener fingerprint patterns. The congener fingerprint patterns were used to identify the source of contamination.

Biegalski et al. (1998) used source-receptor modeling to attribute metal concentrations in the air at three sites near Lake Ontario to sources including oil and coal combustion units, mines, incinerators, and smelting operations. Air samples were collected at the three sites and analyzed for trace elements by neutron activation analysis. Factor analysis, elemental ratios, and enrichment factor analysis were used to determine source-receptor relationships at the three sites.

Another new technique used to trace persistent organic pollutants to their sources is the measurement of the enantiomeric ratios of chiral pollutants in the environment (Ridal et al. 1997, Jantunen and Bidleman 1996). For example, information about the enantioselective degradation organochlorine pesticides in soils (e.g., Aigner et al. 1998) could be coupled with data on the enantiomeric composition of the compounds at the point of deposition to help determine pollutant sources.

Using the technique of chemical mass balance, Su and Christensen (1997) determined that coal-fired power plants, municipal waste incinerators, and pentachlorophenol use areas were major sources of dioxins and furans in the Housatonic River (CT), Lake Huron, and the Baltic Sea. Other studies reported in recent years examining source-receptor linkages include an investigation of source-receptor relationships for mercury in South Florida (Dvonch et al. 1998) and stable isotope analysis for characterization of pollutants at high elevation alpine sites (Pichlmayer et al. 1998).

EXPOSURE AND EFFECTS RESEARCH

Endocrine Disruptors

Endocrine disruptors cause adverse effects by interfering with the normal operation of the endocrine system. These chemicals can act in a variety of ways, such as by mimicking natural hormones or by blocking natural hormones. For example, p,p'-DDE (a breakdown product of DDT) has been shown to inhibit the binding of androgen, a male hormone, to receptors (U.S. EPA 1997b). By interfering with the endocrine system, endocrine disruptors may cause changes in homeostasis, reproduction, and development. Also, since the neural and immune systems are closely linked to the endocrine systems, endocrine disruptors may also act as immunosuppressant and neurotoxins. The *Second Great Waters Report to Congress* (U.S. EPA 1997b) provides more detail about the possible mechanisms of endocrine disruption.

The *Second Report to Congress* acknowledged that a growing body of animal and human data suggests that a number of the Great Waters pollutants of concern potentially act as endocrine disruptors. The report specifically identified 11 of the 15 Great Waters pollutants of concern as possible endocrine disruptors in wildlife: chlordane, dieldrin, DDT/DDE, hexachlorobenzene, lead, lindane, mercury (in the form of dimethylmercury), PCBs, dioxins, furans, and toxaphene. These 11 chemicals, as well as polycyclic organic matter, were also listed as potentially affecting the endocrine system in humans. Since the *Second Report to Congress*, scientific research on endocrine disruptors has continued to provide evidence of their adverse effects in wildlife and humans. In addition, EPA recently developed a draft screening and testing approach for systematically identifying endocrine disruptors and quantifying their effects.

Recent Research

Workshop on Perinatal Exposure to Dioxin-like Compounds

The workshop, which was held June 13-15, 1993 in Berkeley, California, considered the effects of perinatal exposure to chemicals such as dioxins, furans, and PCBs on the reproductive, endocrine, neurodevelopmental, and immune systems. Lindström et al. (1995) concluded that many of the observed effects of these compounds suggest they act as endocrine disruptors. They further proposed that neurobehavioral effects (e.g., spatial learning/memory and motor deficits) may be caused by complex interactions between neuroendocrine and neurophysiological systems.

Some research on endocrine disruptors is focused on the role of the endocrine system in prenatal and perinatal development (see sidebar). Development is under hormonal control, and a precise integration of multiple endocrine systems is required in all stages of development. Developmental effects traditionally have been thought of as physical birth defects; however, this view has been expanded to consider the proper functioning of the individual throughout its life cycle. Environmental pollutants that mimic, block, or modulate the chemical messengers of the endocrine system can cause a variety of functional developmental deficits (e.g., impaired learning, memory, motor skills). For example, the *Second Report to Congress* describes several findings of a study of children whose mothers ate PCB-contaminated fish from Lake Michigan. In this study, children born to mothers consuming the greatest amount of contaminated fish exhibited impaired neurobehavioral ability (e.g., reflexes and response to stimulation) as infants and impaired intellectual function (e.g., IQ, reading comprehension) at school age (U.S. EPA 1997b). The interim findings of two new epidemiology studies support the results of the Lake Michigan study. These effects are important to the Great Waters program because the endocrine disrupting pollutants found in Great Waters fish tissue may be entering the waterbody through atmospheric deposition, among other pathways.

- The Oswego Newborn and Infant Development Project was begun to examine the behavioral effects in human newborns, infants, and children of maternal consumption of Lake Ontario fish that were contaminated with a wide range of persistent toxic chemicals, including several Great Waters pollutants of concern such as PCBs, hexachlorobenzene, dioxins, dieldrin, lindane, chlordane, cadmium, and mercury. Interim study results indicate that newborns of women who consumed high levels of fish from Lake Ontario performed lower on tests of neurobehavioral ability (e.g., reflexes, physiologic responses to stress, and reactivity to stimulation) (Lonky et al. 1996).

- A group of researchers in the Netherlands studied the effects of prenatal exposure to PCBs (estimated from levels in mother during pregnancy) and perinatal exposure to PCBs and dioxins (measured in breast milk) on the psychomotor and mental development of infants (Koopman-Esseboom et al. 1996). At 3 months of age, infants with higher prenatal exposure to PCBs had slightly lower psychomotor scores. At 7 months of age, psychomotor development was negatively influenced by perinatal PCB and dioxin exposure. There was no significant influence of the perinatal PCBs and dioxin exposure on mental development at 3 and 7 months of age.

Endocrine Disruptor Screening and Testing Advisory Committee

The 1996 Food Quality Protection Act and the 1996 Safe Drinking Water Act Amendments mandated that EPA "develop a screening program, using appropriate validated test systems and other specifically relevant information, to determine whether certain substances may have an effect in humans that is similar to an effect produced by a naturally occurring estrogen, or other such endocrine effect as the Administrator may designate." According to these mandates, EPA was required to develop this screening program by August 1998, implement the program by August 1999, and report to Congress on

the program's progress by August 2000. In response, EPA formed the Endocrine Disruptor Screening and Testing Advisory Committee (EDSTAC) and charged the committee with designing a screening and testing program for endocrine disrupting chemicals. The EDSTAC was composed of representatives from EPA, other Federal agencies, State agencies, various sectors of industry, water providers, worker protection organizations, national environmental groups, environmental justice groups, public health groups, and research scientists.

The EDSTAC's final report was released in August 1998 (U.S. EPA 1998c). The report outlines a tiered approach for detecting endocrine disrupting chemicals and quantifying their effects. Under this system, chemicals may be subjected to high throughput pre-screening, tier 1 screening, tier 2 testing, and/or hazard assessment. In the report, EDSTAC recommended specific assays that would be conducted at each step; however, at present, none of the assays are fully validated. The recommended assays address the need to consider multiple species and endpoints because historic reliance on existing test species and endpoints was insufficient to appropriately screen, test, and characterize the risks from endocrine disruption.

The EDSTAC estimated that approximately 87,000 chemicals need to be considered for endocrine disruptor screening and testing, including pesticides, commodity chemicals, naturally occurring non-steroidal estrogens, food additives, cosmetics, nutritional supplements, and representative mixtures. Because simultaneous screening, testing, and evaluation of so many chemicals is far beyond the capabilities of available facilities and resources, EDSTAC suggested that the universe of chemicals undergo an initial sorting into four categories with recommended action as follows:

1. Chemicals that are unlikely to have endocrine disrupting effects would be placed on hold initially;

2. Chemicals with insufficient data would initially undergo high throughput pre-screening and tier 1 screening;

3. Chemicals with sufficient data to bypass tier 1 screening would go directly to tier 2 testing; and,

4. Chemicals with sufficient data would go directly to hazard assessment.

The EDSTAC estimated that following the sorting exercise, approximately 62,000 chemicals would still require screening and/or testing. Therefore, EDSTAC recommended these chemicals should be prioritized for further evaluation and that the Endocrine Disruptor Screening and Testing Program should be implemented in a phased manner (i.e., high priority chemicals screened and tested during Phase 1). The core priority setting process recommended by EDSTAC focuses on giving high priority to chemicals with widespread exposure at the national level. The EDSTAC also recommended a nomination process to accommodate chemicals for which exposure is disproportionately high for specific groups, communities, or ecosystems.

As noted previously, some of the Great Waters pollutants of concern (e.g., PCBs, dioxins) are already known to possess endocrine disrupting capabilities. These chemicals would most likely be categorized into group 3 or group 4 using the sorting scheme recommended by EDSTAC. Nevertheless, at this time, it is uncertain what priority would be given to individual Great Waters pollutants of concern for further evaluation under the Endocrine Disruptor Screening and Testing Program.

Chesapeake Bay Toxicity Testing and Biological Community Assessments

A recent pilot project attempted to integrate an environmental ambient toxicity testing approach with a biological community assessment approach to determine to what extent toxic pollutants affect fish populations in Chesapeake Bay. The results are presented as tributary-specific ambient toxicity testing in the framework of a toxicity risk ranking model developed specifically for contrasting complex toxicological results with biological indicators of community health. The two tools that were combined in this project include an ambient toxicity approach which provides a picture of biologically significant environmental contamination and a toxicological risk ranking method which integrates an array of toxicological data results into a site-specific "risk score" (Hartwell et al. 1995).

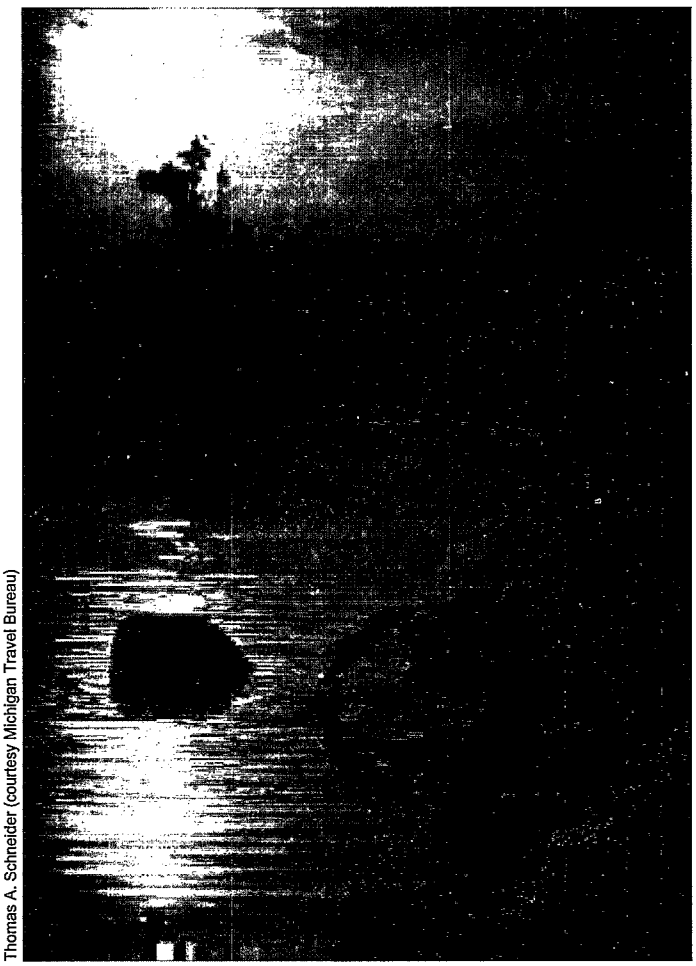
Four tributaries of the Chesapeake Bay were chosen as test sites for this project. Each tributary represents a watershed impacted by different land uses. The Curtis Creek watershed is dominated by urban and commercial development and was selected as an example of a polluted area. The Rock Creek watershed is dominated by urban development but does not have any major industrial areas. Fishing Bay is located in a lightly developed area and is over 70 percent forest and wetlands. This area is considered a relatively uncontaminated environment. The Wicomico River watershed is dominated by forest and agriculture and represents a clean reference area with no direct point source pollution. Fish community and water column sampling was performed between May to September 1993. For the ambient toxicity program, a ranking scheme of five components (end-point severity, response proportion, test variability, site consistency, and number of measured end-points) was developed to evaluate the toxicological results on a site-by-site basis (Hartwell et al. 1995).

The results indicate that the assays are sensitive enough to identify biologically significant contamination. Trends between the Index of Biotic Integrity (IBI) scores, which are an expression of the overall condition of the structure and function of the fish community, and the toxicological risk ranking scheme exist; however, stronger statistical associations are observed between the risk scores and specific metrics in the fish community database. As other studies advance, additional sites can be included in the analyses. As more information is included in the toxicological database, correlations with a variety of community databases will be possible (Hartwell et al. 1995).

ATSDR PCB/PAH Great Lakes Sensitive Population Studies

The Great Lakes Critical Programs Act, enacted in 1990, required EPA in consultation with the Agency for Toxic Substances and Disease Registry (ATSDR) to assess the adverse affects of water pollutants in the Great Lakes. In 1996, EPA and ATSDR published a Report to Congress that summarized existing research on the human health effects of Great Lakes pollutants. In addition, ATSDR developed a Great Lakes Health Effects Research Strategy that the agencies used to guide a suite of new epidemiological studies. In particular, EPA through ATSDR made ten grants to support research related to potential adverse human health effects from consumption of contaminated Great Lakes fish. Eight of the grants supported investigations focusing on susceptible populations (i.e., Native Americans, sports anglers, the urban poor, pregnant women, and fetuses and nursing infants of mothers who eat contaminated fish). The ninth and tenth grants supported the development of an interlaboratory quality assurance and quality control program and the development of more sensitive methods for detecting contaminants in biological samples (U.S. EPA 1999f).

CHAPTER V
NEXT STEPS FOR THE GREAT WATERS PROGRAM



Thomas A. Schneider (courtesy Michigan Travel Bureau)

This report describes significant new scientific and programmatic developments relevant to the Great Waters program since the *Second Report to Congress* (June 1997). These developments support and build on the three broad conclusions presented in the *First* and *Second Reports to Congress*:

- Atmospheric deposition can be a significant contributor of toxic chemicals and nitrogen compounds to the Great Waters. The relative importance of atmospheric loading for a particular chemical in a given waterbody depends on many factors, including characteristics of the waterbody, properties of the chemical, and the kind and amount of atmospheric or water discharges.

- A plausible link exists between emissions into the air of Great Waters toxic pollutants of concern, the atmospheric deposition of these pollutants (and their transformation products), and the concentrations of these pollutants found in water, sediments and biota, especially fish and shellfish. For mercury, fate and transport modeling and exposure assessments predict that the anthropogenic contribution to the total amount of methylmercury in fish is, in part, the result of anthropogenic mercury releases from industrial and combustion sources increasing mercury body burdens (i.e., concentrations) in fish. Furthermore, the consumption of fish is the dominant pathway of exposure to methylmercury for fish-consuming humans and wildlife. However, what is known about each stage of this process varies with each pollutant (for instance, the chemical species of the emissions and its transformation in the atmosphere).

- Airborne emissions from local as well as distant sources, both within and outside the U.S., contribute pollutant loadings to waters through atmospheric deposition. Determining the relative roles of particular sources contributing to specific waterbodies is complex, requiring careful monitoring, atmospheric modeling, and other analytical techniques.

As Chapter II showed, there has been significant progress in environmental research since the *Second Report to Congress*, particularly in monitoring and modeling pollutant transport and loadings at the regional and national scale. As a result, spatial and temporal trends in pollutant emissions, loadings, and effects are becoming clearer as the identified research gaps are being addressed.

Strategic Themes of EPA's Great Waters Program

(1) The EPA will continue ongoing efforts to implement section 112 and other sections of the CAA and use results from this report in the development of policy that will reduce emissions of the Great Waters pollutants of concern.

(2) The EPA recognizes the need for an integrated multimedia approach to the problem of atmospheric deposition of pollutants to waterbodies and will continue to pursue implementation of programs available under various Federal laws to reduce the human and environmental exposure to pollutants of concern.

(3) The EPA is committed to supporting research activities that address the goals of section 112(m) of the CAA.

Chapter III described more than 60 relevant programs and activities, ranging in scale from local to international, many of which directly or indirectly contribute to reducing loadings and exposures for many of the pollutants of concern. Collectively, these programs and activities show a high level of interagency, intergovernmental, and public-private cooperation. In addition, those programs led or supported by EPA have advanced the strategic themes and recommended actions developed in the *First* and *Second Reports to Congress* (see text box), including pursuing integrated multimedia approaches to environmental protection. These programs have also furthered the Agency's *Clean Air, Clean Water, International, Pollution Prevention,* and *Sound Science* goals under the Government Performance and Results Act.

Chapter IV summarized notable developments in science and tools since the *Second Report to Congress*. Because of the large scale and technical complexities of the problems, considerable time, effort, and resources have been expended to develop tools to understand and resolve them. In part because of the efforts of the Great Waters program, there is now a greater level of coordination among research agencies and institutions to target areas of critical uncertainty and suspected threats to human health and the environment. Some of the recently-developed tools and research programs will generate important new data and findings that will be available for future *Great Waters Reports to Congress* and will enable EPA to make further progress in reducing the harmful effects of atmospheric deposition to the Great Waters.

Like the *Second Report to Congress*, the key conclusions of this report and recommendations for continued or further action are organized by the subject areas EPA is required to assess under the Great Waters provisions of the CAA, namely the sources and loadings of the pollutants of concern to the Great Waters, and their environmental and public health effects. In addition, this chapter addresses the description of any revisions to CAA and other Federal requirements, standards, and limitations, (if such revisions are necessary), as well as whether the pollutants of concern have caused any exceedances of water quality or drinking water standards.

In general, deposition rates of the air pollutants of concern to the Great Waters are decreasing or remaining steady. Also, while we observe a trend toward reduced concentrations of these pollutants in water and other media for many atmospherically deposited pollutants of concern, this is not the case for all of them. Potential human health risks are greatest for those individuals who consume fish from contaminated waterbodies. Some pollutants continue to contribute to significant and widespread ecological problems in the Great Waters.

Substantial uncertainties remain about the extent, sources, fate and effects of atmospheric deposition. This report supports continued research targeted toward addressing remaining uncertainties and providing decisionmakers with the tools and information they need to assess potential risks and determine whether further action is necessary to reduce them. The Agency believes that considerable continued effort is justified to address critical remaining uncertainties, improve the characterization of the contribution of atmospherically deposited pollution to the Great Waters, and to further reduce this pollution.

V.A POLLUTANT SOURCES

In the past 2 years, new models and model enhancements, tools, and emissions inventories, including long range transport atmospheric modeling (e.g., RELMAP), source "fingerprinting," back trajectory analysis, EPA's National Toxics Inventory, and the Great Lakes Regional Air Toxics Emission Inventory, have provided new information to help identify sources of pollutants which are deposited to waterbodies. Emissions of some Great Waters pollutants of concern have decreased, while others remained constant or varied. Additional emission reductions are expected to result from continued implementation of current regulations and nonregulatory programs, as well as development of future regulations, pollution prevention initiatives, and voluntary actions.

FINDINGS AND CONCLUSIONS

- **Emissions and numbers of U.S. anthropogenic sources have declined for mercury, lead, dioxins and furans, and the banned and restricted use substances.** For example, lead emissions in the Great Lakes region declined at a rate of 6.4 percent per year from 1982 to 1993 reflecting the national decline in lead emissions resulting from the phase-out of leaded gasoline in automobiles.

- **Emissions from U.S. anthropogenic sources for NO_x have remained relatively constant. For cadmium, emissions in the Great Lakes region have not shown a trend since the 1980s. Trends for POM/PAHs are not known.** For example, nationwide NO_x emissions have fluctuated around 21 to 23 million metric tons per year from 1988 to 1997.

- **The sources of atmospheric deposition vary, depending on the pollutant.** As an example, sources of atmospherically-deposited mercury include emissions from industrial and combustion sources, emissions from natural sources such as volcanoes, and re-emission from mercury-contaminated soils and water. These sources can be in the U.S. or other countries, and the mercury emissions can be deposited near the source or transported long distances across international borders. Some point sources emit significant amounts of reactive chemical forms of mercury which are deposited locally, near the source of emissions. Determining a more complete picture of atmospheric deposition to the Great Waters requires ascertaining the contributions of each of the relevant sources.

- **Local sources, including urban areas, can have a large impact on local pollutant deposition rates.** Recent research under the AEOLOS[1] project continues to show that the diffuse emissions of urban areas (i.e., urban plumes) can significantly affect nearby deposition rates. For example, deposition rates of PCBs and polycyclic aromatic hydrocarbons (PAHs) have been found to be elevated over southern Lake Michigan near the Chicago urban area. Therefore, estimates of pollutant loadings and net flux at the waterbody-scale (e.g., for Lake Michigan) may be sensitive to the placement of monitoring sites.

- **Loadings of banned pesticides to the Great Waters are primarily from sources that are difficult or may not be practical for EPA, States, or tribes in the U.S. to further regulate.** Although there are no major sources of banned pesticides in the U.S., loadings continue from remaining consumer stocks, evaporation from soils, resuspension of contaminated sediments, and

[1] Atmospheric Exchange Over Lakes and Oceans Surfaces (see page IV-13)

airborne transport from other countries. Future reductions must come from clean up of existing stockpiles and contaminated sites and reductions in airborne pollutants transported from other countries. Research continues at EPA and other institutions on soil remediation methods that will reduce emissions of these pollutants from contaminated sites.

- **Implementation of existing EPA regulations is expected to further reduce emissions of mercury, NO_x, POMs, dioxins and furans, cadmium, lead, and hexachlorobenzene, and contribute to declines in deposition of these pollutants.** The EPA regulations issued over the last few years will significantly reduce known sources of these Great Waters pollutants of concern. For instance, the emission standards and guidelines for municipal waste combustors and for hazardous/medical/infectious waste incinerators will reduce mercury and dioxin emissions from these sources by greater than 90 percent from 1990 when fully implemented in 2000 and 2002, respectively. Emission standards for hazardous waste-fired combustors were issued in 1999 and are expected to be implemented in 2002. This rule will reduce dioxins and furans by 70 percent, as well as other pollutants such as mercury. In addition EPA's NO_x SIP call, when implemented, is expected to reduce NO_x emissions by about one million metric tons during the summer ozone season, contributing to a projected decline in NO_x emissions through 2005.

- **Developments in pollution models and source information are improving our ability to identify and quantify sources and deposition of pollutants.** In recent years, EPA and NOAA have continued to develop and refine models and source information in order to improve emissions estimates and understanding of the relative importance of various sources to atmospheric deposition. For example, the long range atmospheric transport model RELMAP[2] was used to estimate domestic and global mercury deposition and RADM[3] was used to estimate coastal NO_x deposition. However, there is still a critical need for key inventory information on the pollutants of concern (e.g., ammonia, speciated data for metals, more accurate data for pollutants emitted in small quantities, locational data for area and mobile sources) in order to generate more effective model estimates that can be used in control strategy decisions.

RECOMMENDATIONS FOR CONTINUED AND FURTHER ACTION

- *Within any limitations imposed by court rulings, EPA will ensure the timely implementation of NO_x control strategies already put in place, such as the NO_x SIP call and emission standards and guidelines for municipal waste combustors, as well as the Tier 1I tailpipe standards, which are expected to significantly reduce on-road mobile source NO_x emissions throughout the year. The EPA will proceed with full implementation of the section 126 rule to achieve the required NO_x reductions by May 1, 2003. Finally, EPA will encourage innovative, nonregulatory approaches to reducing NO_x emissions such as "Smart Growth" planning in urban areas and energy conservation programs.*

- *The EPA will continue to develop and implement technology-based standards and guidelines under sections 111, 112, and 129 of the CAA that will achieve further reductions of Great Waters pollutants of concern. For instance, EPA expects to complete MACT standards mandated by section 112 for source categories which emit pollutants of concern (e.g., chlorine manufacturing*

[2] Regional Langragian Model of Air Pollution

[3] Regional Acid Deposition Model

[chlor-alkali plants], coke ovens [pushing, quenching and battery stacks], industrial boilers, institutional and commercial boilers, iron and steel foundries, and refractory manufacturing) by 2002. Also, EPA expects to complete regulations under section 111 and 129 for commercial and industrial waste incinerators by November 15, 2000, and for small municipal waste combustors by 2001. In addition, EPA expects to finalize the list of area source categories by 2003 and expects to complete regulations by 2004 for the area source categories published in the integrated urban air toxics strategy on July 19, 1999.

- *The EPA expects to complete collection and analysis of information on mercury emissions and controls for coal-fired utility boilers and issue the regulatory determination for air toxics from electric utilities by December 15, 2000.*

- *Under the residual risk program, EPA will assess risks associated with exposures from emissions of Great Waters toxic pollutants of concern, using risk, exposure, and other relevant public health and environmental information developed by EPA offices and its partners. These assessments will focus on the risk remaining from categories of air toxics sources (or individual sources within the categories) for which MACT standards have been applied, and include assessing the public health risk from eating fish contaminated by these sources and risks to ecosystems. Where analyses indicate unacceptable adverse effects to public health or the environment from these sources of pollutants, EPA will pursue appropriate actions to reduce risks.*

- *The EPA will continue to explore ways to integrate the authorities within single media statutes and their programs (i.e., CAA, Clean Water Act) in order to support multimedia strategies to reduce pollutants of concern to the Great Waters. This includes building on recent progress to increase communication and program coordination between EPA's Air and Water programs (e.g., pulp and paper "cluster" rule). For example, EPA's Office of Water and Office of Air and Radiation (OAR) will consider how to involve the air program in review of TMDLs that list air sources.*

- *The EPA will encourage innovation among States to address local air sources of persistent, bioaccumulative toxics and promote voluntary pollution prevention actions, where appropriate. For instance, EPA will document the results of projects under way nationwide to reduce, collect, and recycle mercury-containing products and disseminate the information broadly.*

- *The EPA will continue to encourage phase-out and safe disposal of banned and restricted Great Waters substances (e.g., "clean sweeps" for banned pesticides). The EPA, working with NOAA and other Federal and State partners, will continue to monitor levels in the environment (e.g., fish tissue levels) to verify trends in loadings.*

- *The EPA expects to complete development of a Mercury Research Strategy in 2000. The Strategy will describe the key research questions for mercury that EPA plans to address over the coming 5 years and will identify other technical and scientific issues that are important to the Agency's efforts in addressing mercury.*

- *The EPA expects to finalize in 2000 the Dioxin Reassessment following peer review. This report will update our scientific understanding of the health risks resulting from exposure to dioxins.*

- *The EPA will improve the public's "Right to Know" about certain toxic compounds. The EPA will implement the rulemaking which added dioxin and other persistent, bioaccumulative, toxic*

chemicals to the Toxic Release Inventory and lowered the reporting threshold for these and other listed chemicals under the TRI. The EPA also expects to issue to the public by 2001 the first TRI reports that include these substances.

- *The EPA will continue to lead efforts to research viable controls for sources of pollutants of concern. For example, research will continue on mercury from combustion processes, including modifying the mixtures and inputs of fuel, and more effective control devices on emitted gases and particles. This research will also evaluate costs and relative effectiveness of control options.*

- *The EPA will continue to provide leadership in reducing transboundary transport of mercury, DDT, PCBs, chlordane, and other toxic pollutants of concern by achieving assessment and reduction goals contained in international agreements (e.g., Great Lakes Binational Strategy, CEC[4] North American Regional Action Plans, UN/ECE LRTAP[5] Heavy Metals Protocol). The EPA will support efforts to share technology, information and expertise with other countries on reducing releases to the environment and on cost-effective alternatives. The EPA will also continue to provide support to the international negotiations on persistent organic pollutants under the United Nations Environmental Programme.*

- *The EPA will continue to pursue international agreements to reduce transboundary NO_y including negotiating an ozone annex to the US/Canada Air Quality Agreement.*

- *The EPA will continue to support joint work with States and industry to fill gaps in source categories and further refine emission measurement methods and inventories for Great Waters pollutants of concern (e.g., speciated data on pollutants such as metals and PAHs for the Great Lakes RAPIDS inventory and for the National Toxics Inventory) so that they can be used effectively in models being developed.*

[4] Commission for Environmental Cooperation

[5] United Nations Economic Commission for Europe Long Range Transboundary Air Pollution Convention

V.B CONTRIBUTION OF ATMOSPHERIC DEPOSITION TO POLLUTANT LOADINGS IN THE GREAT WATERS

Research, monitoring, and modeling of pollutant inputs to the Great Waters has continued to provide important information about loadings. Recent results from available data indicate that atmospheric inputs of some pollutants of concern are decreasing while others remain constant. However, there is considerable uncertainty in the data because of limited monitoring, technological barriers, and variable collection and analytical methods, making it difficult to adequately characterize historical as well as current conditions and to discern pollution trends. In addition, not all Great Waters waterbodies or all pollutants of concern have been studied.

FINDINGS AND CONCLUSIONS

- **Based on available data, atmospheric deposition of lead, cadmium, POM/PAHs, PCBs, and some pesticides (e.g., DDT, hexachlorocyclohexanes, dieldrin) to the Great Lakes has continued to decline in recent years.** Similar trends are seen in some other Great Waters.

- **Based on available data, atmospheric deposition of nitrogen compounds has remained relatively unchanged in the Great Waters in recent years.** This correlates with a relatively constant trend in NO_x emissions during the same period. Considerable uncertainty exists, however, concerning dry deposition of nitrogen compounds and wet organic nitrogen deposition.

- **Despite recent declines, atmospheric deposition continues to be a significant contributor of certain pollutants to some Great Waters.** For example, one researcher has reported that atmospheric deposition contributes 70 to 90 percent of the direct lead inputs to the Long Island Sound. In the Great Lakes, according to another study, the relative contribution of dioxins and furans from the atmosphere ranges from 5 to 100 percent. Various studies of nitrogen deposition to Atlantic and Gulf Coast bays and estuaries indicate that 2 to 38 percent of total nitrogen inputs are attributable to atmospheric deposition.

- **While run-off from fertilizer application, farm animal operations, waste treatment, industrial effluents, and crop residues continues to be the largest contributor to total nitrogen loadings, atmospheric inputs of nitrogen compounds to the Great Waters are much higher than natural rates.** Although atmospheric loading rates of inorganic nitrogen appear to have leveled off, the rates remain many times greater than natural rates and have the potential, when combined with other loadings such as runoff, to overwhelm the assimilative capacities of surface waters. Research is ongoing into the lesser known but apparently important forms of nitrogen deposition (e.g., dry deposition, organic nitrogen). In addition, nitrogen saturation of upland forests, grasslands, and agricultural fields may occur in some areas and could significantly increase the rate of nitrogen transport from upland watersheds to coastal waters in the future, even if atmospheric loading rates remain constant.

- **Although considerable uncertainty remains, EPA's 1997 *Mercury Study Report to Congress* estimates, based on a modeling analysis, that one-third (52 tons) of the anthropogenic mercury emitted annually in the U.S. is deposited in the continental U.S., along with an estimated 35 tons from the global reservoir (which includes U.S. anthropogenic as well as natural and re-emitted mercury emissions).** These results to date suggest that the remaining two-thirds (~ 107 tons) of annual U.S. anthropogenic emissions are transported beyond U.S. borders, where they diffuse into the global reservoir. As U.S. anthropogenic emissions are

reduced, the relative contribution from the global pool will be even larger, particularly if similar control actions are not undertaken in other countries. Studies of mercury deposition to certain Great Waters indicate that atmospheric deposition contributes between 10 and 85 percent of the total mercury loadings to these waterbodies. Because a deposition monitoring network for mercury has only recently been established, there are not yet sufficient data to determine monitored trends in mercury deposition levels.

- **Certain banned and restricted use pesticides are projected to be the first pollutants of concern to be reduced to concentrations below current atmospheric detection limits in the Great Lakes atmosphere, indicating that risk management strategies are having the intended impact.** Based on recent academic research on atmospheric pollutant concentrations in the Great Lakes region, DDT and DDE, followed by dieldrin and chlordane, are estimated to fall below current detection limits in the atmosphere between 2010 and 2020. Hexachlorocyclohexane and hexachlorobenzene are projected to fall below current detection limits in the atmosphere over the Great Lakes by 2030 and 2060, respectively. These estimates assume current rates of long-range transport of these pollutants into the region. Because of their persistence, it should be noted that elimination of these pollutants in the atmosphere does not mean that concentrations would be eliminated in deposited media (e.g., sediments) by these dates. However, these estimates indicate that reduction strategies in the Great Lakes, along with the original bans or restrictions on the use of these substances, are having the intended effect.

- **Significant progress has been made involving monitoring and modeling research focused on the factors that affect pollutant fate and transport in regional-scale air masses and in watersheds.** Since the *Second Report to Congress*, there have been significant advances in monitoring and modeling of pollutant transport and loadings at both regional and national scales. Recent accomplishments include data collection and analysis of results from the AEOLOS project, establishment of the Mercury Deposition Network (part of the National Atmospheric Deposition Program), applications of the HYSPLIT/TRANSCO[6] computer program, development of the Models-3 community multiscale air quality modeling system, monitoring established by the South Florida Mercury Science Program, and progress on the Lake Michigan Mass Balance Study. The Integrated Atmospheric Deposition Network, an international effort to monitor air toxics deposition, has produced a long-term data set on loadings trends in the Great Lakes. As in past years, much of the research on modeling of pollutant loading rates has focused on mechanisms of direct pollutant deposition to surface waters (e.g., pollutant exchange mechanisms, surface microlayers). However, a growing area of research addresses factors, such as the effects of land cover, weather events, that affect the fate and transport of pollutants in watersheds and tributaries. For example, a variety of factors influence how much deposited mercury will be methylated and incorporated into the food chain, including lake and watershed characteristics, water chemistry, and the length of the food chain.

- **Long-range transport from other U.S. regions or other countries is estimated to contribute significantly to atmospheric loadings of pollutants of concern to the Great Waters.** As stated above, EPA estimates that 35 tons of mercury from the global reservoir (derived from both foreign and U.S. sources) are deposited annually in the U.S., representing 40 percent of the total mercury deposition to the U.S. In a modeling study conducted in the northeastern U.S., sources from outside the study region contributed 30 percent of the mercury loadings within the study region and the global reservoir contributed approximately 20 percent. Another modeling study

[6] Hybrid single particle Lagrangian integrated trajectory/transfer coefficient (see page II-31)

conducted to assess sources of dioxin to the Great Lakes basin shows that sources as far as central Canada may influence deposition of dioxin to the lakes.

RECOMMENDATIONS FOR CONTINUED AND FURTHER ACTION

- *By summer 2000, EPA expects to develop a work plan to assess atmospheric deposition on a regional basis. This work plan would include: targeting State-identified impaired waterbodies; examining what rules or activities are in place that address impairment caused by atmospheric deposition; and, determining what, if any, additional actions are necessary to address impairment caused by atmospheric deposition. In addition, EPA expects to revise this work plan every two years based on updated scientific information and stakeholder input.*

- *The EPA will work with national deposition monitoring programs, States, National Estuary Programs, NOAA, and other research institutions to establish monitoring sites in coastal areas where they do not already exist, as appropriate. The EPA will encourage expansion of the National Atmospheric Deposition Program's Mercury Deposition Network, the continuation of other toxics deposition monitoring networks, such as the Integrated Atmospheric Deposition Network, and the development of a national air toxics monitoring network to support assessment of the contribution of long-range transport to deposition of Great Waters pollutants of concern in the U.S. and to evaluate the impact of urban sources.*

- *The EPA will encourage the use of standard monitoring methods to enable comparison of data and trends analysis. Where methods for measuring atmospheric deposition for pollutants of concern do not exist, EPA, working with NOAA and other Federal colleagues and key institutions such as the National Atmospheric Deposition Program, will continue to lead and support efforts to develop technology and establish standard methods (e.g., POM and PAH, mercury species, dissolved organic nitrogen, and dry deposition of pollutants).*

- *The EPA will continue to work with NOAA and other scientific partners to develop, refine, and validate modeling tools that help us to better understand the pathways of the pollutants of concern -- from emissions to transport to deposition to fate to environmental and human health effects.*

- *The EPA will work with NOAA and other agencies to better quantify the indirect loadings of atmospheric deposition to the Great Waters through the development of tools that can quantify watershed transport of pollutants of concern.*

- *The EPA will continue to support international efforts to quantify the transboundary contributions of pollutants of concern, such as the Commission for Environmental Cooperation efforts to assess continental pollutant pathways and the Binational Strategy challenge to assess global sources of pollutants to the Great Lakes.*

V.C ENVIRONMENTAL AND PUBLIC HEALTH EFFECTS

Because the *Second Report to Congress* discussed potential adverse human health and environmental effects from exposure to the Great Waters pollutants of concern in considerable detail, this report focused on observed changes in exposure levels and trends in observed effects, to the extent that data were available. Scientific research has continued to improve our understanding of the toxic effects of the pollutants of concern, including endocrine disruption. Concentrations of some of the pollutants of concern in the environment have decreased in recent years, while others have remained constant or are variable. At some locations, mercury concentrations are at levels sufficient to produce adverse health effects for certain groups consuming large amounts of contaminated fish (e.g., young children, pregnant women and their developing fetuses, women of child-bearing age, and populations that subsist on fish), as well as ecological risks.

FINDINGS AND CONCLUSIONS

- **Available monitoring data indicate that concentrations of dioxin/furans and PCBs, in the sediment, water, and biota of several of the Great Waters appear to be declining, while concentrations of lead, cadmium, mercury, and POM/PAH are too variable for a trend to be discerned.** For example, concentrations of PCBs in biota have continued to decline in the Great Lakes, with PCB concentrations in St. Lawrence River fish decreasing by a factor of 30 since 1975. Trends in concentrations of lead, cadmium, mercury, and POM/PAH were more variable or could not be discerned from the available data. Although recent research did not address contamination levels of all pollutants of concern, no identified studies indicated increasing levels of pollutants of concern in the Great Waters.

- **Available monitoring data indicate that various pollutant concentrations in sediments, water, and/or biota of the Great Waters have been detected at levels resulting in exposures that are high enough to cause adverse environmental effects.** As was reported in the *Second Report to Congress*, a number of regional- and national-scale assessment and monitoring programs (e.g., the National Sediment Quality Survey, Benthic Surveillance Project) suggest that the ecological health of many Great Waters are impaired by pollution that is partially attributable to atmospheric deposition. Further evidence of ecological impairment includes research results indicating that up to 30 percent of the loons in the northeastern U.S. have mercury levels sufficiently high to cause adverse effects.

- **Excess nitrogen loadings can cause algal blooms (including harmful algal blooms), shifts in aquatic vegetation, fish declines and kills, and shellfish bed losses.** The NOAA Estuarine Eutrophication Surveys indicate that the adverse effects of excess nitrogen loadings are currently evident to some degree in approximately 89 percent of the U.S. coastal Great Waters. Numerous studies on individual estuaries have estimated that 20 to 40 percent of total loading of nitrogen compounds can come from atmospheric deposition, with the rest of the loading originating from water discharges or land-use activities.

- **Fish consumption is the primary pathway of human exposure to mercury, is an important pathway for dioxin and PCBs, and may be an important pathway for various other Great Waters toxic pollutants. While such exposures may not be of concern for most of the general population, for certain pollutants and contamination situations, certain groups such as young children, developing fetuses, subsistence fish-eating populations, and others who consume large amounts of contaminated fish, may be at risk.** For mercury specifically,

exposures do not appear to pose a health risk to people consuming average amounts of fish , but sensitive sub-populations (e.g., young children, and pregnant women and their developing fetuses) with higher than typical fish consumption are at risk. Also at risk are subsistence fish-eating populations who consume large amounts of fish. The extent of risk for these groups depends on the amount of fish consumed and the mercury concentrations present in the fish.

- **The role of transformation processes on certain pollutants once they are emitted is an important phenomenon which can increase their toxicity and persistence in the environment.** For instance, mercury emitted by industrial or combustion processes becomes much more toxic and biologically available after it is deposited in the environment and transformed to methylmercury. Another example is the transformation of alpha- and gamma-hexachlorocyclohexane to beta-hexachlorocyclohexane (HCH). More research on these transformation processes is needed to better understand their role and to develop appropriate responses.

- **Evidence for adverse effects from endocrine disrupting chemicals continues to be found.** Recent research provides additional evidence of the adverse effects of endocrine disruptors on both wildlife and humans, and at least two Great Waters pollutants of concern (i.e., PCBs and dioxins) are known to possess endocrine disrupting capabilities. The EPA has worked with various governmental and non-governmental interests to develop a screening and testing approach for systematically identifying endocrine disruptors and quantifying their effects.

RECOMMENDATIONS FOR CONTINUED AND FURTHER ACTION

- *The EPA will continue to support the development and validation of modeling tools which address the transport and fate of pollutants in ecosystems and characterize risk, such as the Total Risk Integrated Methodology. The EPA will also continue to support the improvements in inventories, monitoring data, and human and environmental effects information necessary to effectively apply these tools.*

- *In conjunction with its related efforts on persistent, bioaccumulative, toxic pollutants (e.g., the Great Lakes Binational Strategy and the Persistent, Bioaccumulative Toxics Initiative), EPA will identify and evaluate additional pollutants which may be of concern to the Great Waters.*

- *The EPA expects to explore one or more community-based pilot projects to develop and examine methodologies for characterizing local risks and to work with stakeholders on risk reduction strategies. In conducting such pilot studies, the EPA will consider cumulative risks presented by exposures to air toxics emissions in the aggregate, including exposures through noninhalation pathways, such as consumption of contaminated fish from waterbodies affected by deposition of air toxics.*

- *The EPA will continue to support work to quantify the ecological effects of atmospheric deposition. For instance, EPA will support research to answer critical questions related to the assessment of pollutant exposures for varying life stages and for species at various levels of the food chain.*

- *The EPA expects to complete, by December 2000, the new Water Quality Criterion Methodology for Human Health to better reflect pollutants that bioaccumulate and the Nation's changing fish consumption patterns.*

- *The EPA will work with NOAA, US Fish and Wildlife Service, other Federal agencies, States, and tribes to expand the geographic coverage and consistency of waterbody monitoring to enable more accurate characterizations of the extent of contamination and ecosystem effects due to atmospheric deposition of Great Waters pollutants of concern. In addition, EPA will increase efforts to implement nationally-consistent methods and protocols for assessing contaminants in fish and wildlife and establishing consumption advisories.*

- *The EPA, working with States, tribes, and other relevant partners, will continue to support efforts to improve awareness and understanding of fish consumption advisories among populations most at risk to exposure to the Great Water pollutants of concern.*

- *The EPA will provide leadership for efforts to examine the mechanisms of action and resulting effects (e.g., reproductive failure, death, species diversity, ecological sustainability) of exposures to realistic concentrations of common contaminants, alone and in combination. The EPA will also lead the development of predictive models to assess likely effects of single and multiple stressors and alternative environmental conditions on individual aquatic plants and animals, populations, communities, and ecosystems.*

- *The EPA will continue to pursue as a priority its research plan for endocrine disruptors, covering ten broad categories of research needs: basic research, biomarkers, database development, exposure determination, exposure follow-up, mixtures, multidisciplinary studies, risk assessment methods, hazard identification, and sentinel species.*

- *The EPA will accelerate work to better quantify the water quality benefits of air pollution controls in order to provide decisionmakers with critical information on environmental strategies to reduce the extent of contamination by Great Waters pollutants of concern. By 2001, EPA expects to conclude a pilot study of the economic benefits of reducing nitrogen deposition in a particular estuary, developing methodologies that could be applied to other waterbodies.*

V.D EXCEEDANCES OF WATER QUALITY OR DRINKING WATER STANDARDS

Atmospheric deposition and other inputs of pollutants of concern to the Great Waters can result in exceedances of drinking water standards and surface water quality guidance and criteria, posing threats to human health and the environment. Recent data indicate that water quality standards in place for drinking water supplies in the Great Waters are not being exceeded for the pollutants of concern; however, surface water quality guidance and criteria are being exceeded in some of the Great Waters.

FINDINGS AND CONCLUSIONS

- **The pollutants of concern cause no exceedances of water quality standards in place for drinking water supplies in the Great Waters.** Pollutant levels in both the Great Lakes and Lake Champlain are below primary drinking water standards and other thresholds of water quality. Data indicate that most of the Great Lakes nearshore waters can be used as a source of drinking water with normal treatment. Similarly, Lake Champlain was not associated with any violations of standards in place for drinking water supplies due to Great Waters pollutants of concern from 1986 to 1995. Further reductions in pollutant concentrations may reduce the cost of drinking water treatment in some areas.

- **Primarily because of contamination with metals and nutrients from a variety of sources, some of which are atmospheric, approximately 40 percent of the Nation's surveyed rivers, lakes, and estuaries have been found to have contaminant levels exceeding water quality criteria and, therefore, discourage basic uses, such as fishing and swimming.** In lakes and rivers, nutrients and metals are the most widespread pollutants, and agriculture is the most common source; however, atmospheric deposition has also been identified as a source. In estuaries, nutrients are the most widespread pollutants with industrial point dischargers, urban runoff, and storm sewers identified as the most common pollutant sources. Atmospheric deposition has also been identified as a significant source of nitrogen loadings, directly to the water surface and indirectly via runoff, to most coastal waters where measurements have been made.

- **While the Great Lakes generally are safe for swimming and other recreation, virtually all of the surveyed shoreline area shows unfavorable conditions for supporting aquatic life and is impacted by toxic organic chemicals that appear in fish tissue samples at much higher concentrations than in water samples.** This is partially due to persistent toxic pollutant burdens, such as PCBs and PAHs, in the food web. Several of the Great Lakes States identified air pollution among other sources as contributing to impaired water quality.

RECOMMENDATIONS FOR CONTINUED AND FURTHER ACTION

- *The EPA expects to complete the development of national water quality criteria for nutrients for lakes, reservoirs, rivers, streams, estuaries, and coastal waters. This includes developing waterbody-specific guidance manuals to help State and tribal agencies: (1) classify and assess their waters in terms of nutrient condition; (2) develop region-specific water quality standards; and (3) plan management responses to nutrient pollution. Where sufficient data are available, EPA also expects to develop specific target ranges for total nutrient loads that States can use in the development of their standards.*

- *The EPA expects to complete a pilot project to demonstrate total maximum daily load allocations for two waterbodies receiving mercury from atmospheric deposition. This pilot project will evaluate the integration of air and water program technical tools and authorities, and will examine emission reduction options. The EPA will also work with States that have identified waterbodies whose impairment may be the result of atmospheric deposition to develop tools to assist in establishing TMDLs that account for air sources. For example, based on the outcome of the pilot project, EPA will explore the possibility of providing States with modeled regional baseline and projected deposition estimates for several Great Waters pollutants of concern.*

- *The EPA, in partnership with the Chesapeake Bay watershed States, will account for nitrogen deposition as a part of an effort to improve water quality in the Bay and its tributaries. The Chesapeake Bay partners have committed to working to integrate a cooperative, statutory program so that these waters could be removed from the list of impaired waters by 2010.*

- *The EPA, working with the Great Lakes States, will ensure implementation of the Great Lakes Water Quality Guidance.*

- *The EPA will continue to work with its Federal agency partners such as NOAA, and with States, tribes, and local agencies to improve understanding of the relationship and relative contribution of atmospheric deposition and water pollution. The EPA expects to develop guidance to assist National Estuary Programs, States, tribes and local organizations, among others, to evaluate the role of air deposition in assessing water quality.*

V.E SUMMARY AND KEY RECOMMENDATIONS

The EPA expects that the many ongoing and scheduled future regulatory and voluntary programs and activities, many of which are described in this report, will further reduce the impact of air deposition of pollutants of concern on the Great Waters. These include, but are not limited to:

- Establishing remaining MACT and section 112(c)(6) standards for sources emitting Great Waters pollutants of concern;

- Implementing EPA's strategy for the Residual Risk program, which assesses the risk remaining from MACT source categories (including those emitting Great Waters pollutants of concern), and issuing additional standards, as appropriate, within the required 8 years of the MACT standard being promulgated for the source category;

- Implementing programs to control NO_x, such as regional strategies to reduce NO_x emissions (e.g., the NO_x SIP call and the rulemaking by EPA in response to States' petitions under CAA section 126), Tier II/Gasoline sulfur rules for cars, and emission standards and guidelines for municipal waste combustors, as well as encouraging voluntary approaches to reduce these emissions;

- With NOAA and other Federal agency partners, completing the strategy described in the Clean Water Action Plan issued in 1998, which addresses remaining obstacles to establishing "fishable and swimmable waters for all Americans"; and,

- Completing and implementing national multimedia action plans for persistent, bioaccumulative toxics (PBT) under the Agency's PBT Initiative.

Development and implementation of these and other programs and initiatives described in this report should not require revisions to requirements, standards, and limitations pursuant to the CAA and other Federal laws. Consequently, EPA expects that these programs should help to assure protection of human health and the environment from atmospheric deposition to the Great Waters. However, in order to ensure continued progress in reducing sources and loadings of atmospheric deposition to the Great Waters, and to further reduce the environmental and public health effects, EPA will:

- Continue to support the maintenance and expansion of efforts to monitor Great Waters pollutants of concern in order to evaluate the relative contributions of local, regional, and long-range transport to deposition in the U.S., as well as natural versus human-made sources;

- Continue to develop and implement regulations and pollution prevention programs regionally and nationally, including multimedia programs, in order to reduce the release and impact of sources of Great Waters pollutants of concern within the U.S.;

- For Great Waters pollutants not emitted by U.S. sources, work within international frameworks to reduce sources of these pollutants;

- Support model development and research that establish and, if possible, quantify the linkages from emissions to atmospheric deposition to waterbody loadings to adverse public health and the environmental effects of Great Waters pollutants of concern in order to enable effective risk management decisions;

- Encourage and support the establishment of common baselines and measures of progress in order to better assess trends and health of Great Waters and other waterbodies affected by atmospheric deposition; and,

- Work to increase public awareness of risks of exposure to Great Waters pollutants of concern.

The EPA is committed to continuing to address air deposition of pollutants into the Nation's waters as a priority matter. To that end, and to assure continued coordination of the many related tasks involved and outlined in this report, EPA will develop a detailed biennial work plan for implementation actions beginning this year and updated every two years. As EPA develops and implements plans, programs and initiatives with NOAA and its other Federal, State, tribal, industry and community partners, we expect to make significant, measurable progress toward our goal of assuring the protection of human health and the environment from adverse effects attributable to atmospheric deposition of pollution to the Great Waters.

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Major Sources of Information: Publications and Internet Sites

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http://www.cciw.ca/glimr/program.html

Great Lakes Information Network
http://www.great-lakes.net

Great Lakes National Program Office
http://www.epa.gov/glnpo

Gulf of Mexico Program Office
http://pelican.gmpo.gov

Integrated Atmospheric Deposition Network - Environment Canada
http://airquality.tor.ec.gc.ca/IADN

Integrated Atmospheric Deposition Network - U.S. EPA
http://www.epa.gov/grtlakes/air

International Joint Commission
http://www.ijc.org

Lake Champlain Basin Program
http://www.anr.state.vt.us/champ

Lake Michigan Mass Balance Study (LMMBS)
http://www.epa.gov/glnpo/lmmb

Michigan State Mercury Pollution Prevention
http://www.deq.state.mi.us/ead/p2sect/mercury

National Atmospheric Deposition Program/AIRMoN
http://nadp.sws.uiuc.edu/airmon

National Atmospheric Deposition Program/National Trends Network (NADP/NTN)
http://nadp.sws.uiuc.edu

National Atmospheric Deposition Program/Mercury Deposition Network (NADP/MDN)
http://nadp.sws.uiuc.edu/mdn

National Estuarine Research Reserve System (NERRS)
http://inlet.geol.sc.edu/nerrsintro.html

National Estuary Program (NEP)
http://www.epa.gov/nep

NOAA Air Resources Laboratory, AIRMoN-Dry
http://www.arl.noaa.gov/research/projects/airmon_dry.html

North American Agreement on Environmental Cooperation (NAAEC)
http://www.naaec.gc.ca/english/index.html

Persistent Bioaccumulative Toxics (PBT) Initiative
http://www.epa.gov/pbt

U.S. Environmental Protection Agency (EPA)
http://www.epa.gov

APPENDIX B
Detailed Breakdown of Air Emissions Inventory by Pollutant for Source Categories Emitting Less than 1 Percent of Total U.S. Emissions

This appendix lists the source categories that contribute less than 1 percent to total U.S. emissions for mercury and compounds, lead and compounds, cadmium and compounds, dioxins and furans, polycyclic aromatic hydrocarbons (PAHs), and polychlorinated biphenyls (PCBs). This information is from EPA's *Mercury Study Report to Congress* (U.S. EPA 1997e) for mercury and compounds and from EPA's 1993 National Toxics Inventory (NTI) for the remaining compounds (U.S. EPA 1999a). This list is referenced in Tables II-1, II-4, II-5, II-11, II-14, and II-22, in which the source categories contributing greater than 1 percent to total U.S. emissions are listed along with the percent contribution to total U.S. emissions.

MERCURY AND COMPOUNDS

Batteries
Byproduct Coke
Carbon Black
Crematories
Electrical Apparatus
Flourescent Lamp Recycling
Geothermal Power
Instruments Manufacturing
Lime Manufacturing
Mercury Compounds
Pigments, Oil, etc.
Primary Copper
Primary Lead
Primary Mercury Production
Refineries
Secondary Mercury Production
Sewage Sludge Incinerators
Turf Products
Wood-fired Boilers

LEAD AND COMPOUNDS

Abrasive Products
Adhesives and Sealants
Aerospace Industry (Surface Coating)
Agricultural Production
Agricultural Chemicals and Pesticides
Air and Gas Compressors
Air, Water, & Solid Waste Management
Aircraft Engines and Engine Parts Manufacturing
Aircraft Manufacturing
Aircraft Parts and Equipment Manufacturing
Aircraft And Parts
Airports, Flying Fields, & Services
Aluminum Extruded Products
Aluminum Foundries
Aluminum Foundries (Castings)
Aluminum Sheet, Plate, and Foil manufacturing
Aluminum Die-Castings
Ammunition, Except for Small Arms
Amusement Parks
Amusement And Recreation, Nec
Animal Cremation
Asphalt Paving Production
Asphalt Paving Mixtures And Blocks
Asphalt Production
Asphalt Roofing Production
Automotive stampings
Automotive Dealers, Nec
Aviation Gasoline Distribution: Stage I & II
Beet Sugar
Black Liquor Combustion
Blast Furnaces and Steel Mills
Boat Building and Repairing
Bolts, Nuts, Rivets and Washers Manufacturing
Bookbinding And Related Work
Botanical And Zoological Gardens
Brass, Bronze, Copper, Copper Base Alloy Foundries
Bread, Cake, And Related Products
Brick and Structural Clay Tile
Business services, nec (1987)
Canned specialties
Canned Fruits and Vegetables
Carbon and Graphite Products
Carbon Black Manufacture
Carburetors, Pistons, Rings and Valves Manufacturing
Cathode Ray Television Picture Tubes Manufacturing
Cement, Hydraulic (not subject to Portland Cement Regulation)
Ceramic Wall and Floor Tile Manufacturing
Chemical Manufacturing: Explosives & Blasting Agents

LEAD AND COMPOUNDS (CONTINUED)

Chemical Preparations
Chemical Manufacturing: Cyclic Crude and Intermediate Production
Chemicals and Allied Products Manufacturing
Chromium Plating: Chromic Anodizing Plating
Coated Fabrics, not Rubberized, Manufacturing
Coke Ovens: By-product Recovery Plants
Cold Finishing of Steel Shapes
Commercial Printing, Lithographic
Commercial Printing, nec
Commercial Physical Research
Commercial/Institutional Boilers: Coal Combustion, all types
Commercial/Institutional Heating: Anthracite Coal Combustion
Commercial/Institutional Heating: Bituminous and Lignite Coal Combustion
Commercial/Institutional Heating: Distillate Oil Combustion
Commercial/Institutional Heating: Natural Gas Combustion
Commercial/Institutional Heating: Residual Oil Combustion
Commercial/Institutional Heating: Wood/Wood Residue Combustion
Communications Equipment, nec
Computer Peripheral Equipment, nec
Construction Machinery Manufacturing
Construction (SICs 15-17)
Construction Sand And Gravel
Conveyors and Conveying Equipment Manufacturing
Copper Rolling and Drawing
Copper Foundries
Cordage and Twine
Correctional Institutions
Costume Jewelry
Cotton
Cottonseed Oil Mills
Courts
Crop Preparation Services For Market
Crude petroleum and natural gas
Crude Petroleum Pipelines
Crushed And Broken Limestone
Crushed And Broken Stone, Nec
Crushed And Broken Granite
Current-carrying Wiring Devices
Custom Compound Purchased Resins Manufacturing
Dehydrated fruits, vegetables, and soups
Depository Institutions
Dog and Cat Food
Drum and Barrel Reclamation
Durable Goods, Nec
Electric and other services combined
Electric Lamps
Electric services
Electrical Equipment and Supplies, nec
Electrical Industrial Apparatus, nec
Electrical Apparatus and Equipment
Electromedical Equipment Manufacturing
Electrometallurgical Products
Electron Tubes Manufacturing
Electronic Components, nec
Electronic Computers
Electronic Computing Equipment
Electronic Connectors
Electronic Capacitors Manufacturing
Electronic Resistors
Elevators and Moving Stairways Manufacturing
Engine Electric Equipment
Engineering Services
Environmental Controls Manufacturing
Fabricated Metal Products, nec
Fabricated Plate Work (Boiler Shops)
Fabricated Rubber Products, nec
Fabricated Structural Metal Manufacturing
Fabricated Structural Metal Products
Farm Machinery and Equipment Manufacturing
Ferroalloy Ores, Except Vanadium
Fertilizers, Mixing only
Flat Glass
Fluid power Valves and Hose Fittings Manufacturing
Fluid Meters and Counting Devices
Food and Agricultural Products: Cotton Ginning
Food Preparations Production
Food Products Machinery Manufacturing
Frozen fruits, fruit juices and vegetables
Funeral Service And Crematories
Furniture and Fixtures Manufacturing
Gas And Other Services Combined
Gaskets, Packing and Sealing Devices Manufacturing
Gasoline Distribution Stage II
Gasoline Distribution Stage I
General Industrial Machinery Manufacturing
General Automotive Repair Shops
General Medical & Surgical Hospitals
Glass Containers
Gold Ores
Grain And Field Beans
Gray and Ductile Iron Foundries
Greeting Cards
Grocery Stores
Guided Missiles and Space Vehicles Manufacturing
Halogenated Solvent Cleaners
Hand and Edge Tools Manufacturing
Hardware Manufacturing
Heating Equipment, Except Electric
Heavy Construction, Nec
Hospitals
Hotels And Motels
Household Cooking Equipment
Household Audio and Video Equipment
Hunting, Trapping, Game Propagation
Industrial Boilers: Residual Oil Combustion
Industrial Boilers: Bituminous and Lignite Coal Combustion
Industrial Boilers: Coal, all types
Industrial Boilers: Distillate Oil Combustion
Industrial Boilers: Natural Gas Combustion
Industrial Boilers: Anthracite Coal Combustion
Industrial Boilers: Waste Oil Combustion
Industrial Boilers: Wood/Wood Residue Combustion
Industrial Gases Manufacturing
Industrial Inorganic Chemical Manufacturing
Industrial machinery and equipment
Industrial Organic Chemicals Manufacturing
Industrial Sand
Industrial Supplies
Industrial Trucks and Tractors Manufacturing
Industrial/Utility Dist. Oil/Diesel Turbines
Inorganic Pigments Manufacturing
Instruments to Measure Electricity
Integrated Iron and Steel Mills

LEAD AND COMPOUNDS (CONTINUED)

Intercity & Rural Bus Transportation
Internal Combustion Engine Manufacturing
Iron and Steel Foundries: Steel Foundries
Iron and Steel Foundries: Steel Investment Foundries
Iron and Steel Forging
Laminated Plastics Plate and Sheet
Lead Pencils, Art Goods Manufacturing
Life Insurance
Lighting Equipment
Lightweight Aggregate Kilns
Lime
Lubricating Oils and Greases
Malleable Iron Foundries
Malt Beverages
Manufacturing Industries Manufacturing
Marine Cargo Handling
Measuring and Controlling Devices, nec
Meat Packing Plants
Membership Sports & Recreation Clubs
Metal cans (3411)
Metal coating and allied services (3479)
Metal Coil (Surface Coating)
Metal Barrels, Drums, and Pails Manufacturing
Metal Foil and Leaf
Metal Forgings and Stampings
Metal Heat Treating Manufacturing
Metal Sanitary Ware Manufacturing
Metal Stampings Manufacturing
Metal Valves
Mineral Wool Manufacturing (includes Wool Fiberglass)
Mineral Wool
Minerals, Ground or Treated Production
Mining Machinery Manufacturing
Miscellaneous Plastics Products
Miscellaneous Fabricated Wire Products
Miscellaneous Metal Work
Miscellaneous Nonmetallic Minerals
Miscellaneous Fabricated Metal Products
Miscellaneous Plastics Products, nec
Mobile Sources: Railroads
Mobile Sources: Non-Road Vehicles and Equipment - Commercial Marine Vessels
MON - Continuous Processes
Motion Picture & Video Production
Motor Vehicle Equipment
Motor Vehicle Parts and Accessories Manufacturing
Motor Vehicles and Car Bodies Manufacturing
Motor and Generators Manufacturing
Musical Instruments
National Security
Natural Gas Liquids
Newspapers
Non-road Mobile Vehicles
Noncommercial research organizations (1987)
Noncurrent-Carrying Wiring Devices
Nonferrous Foundries, nec
Nonferrous Rolling and Drawing
Nonferrous Wire Drawing and Insulating
Nonferrous Forgings
Nonferrous Die-castings, Except Aluminum
Nonmetallic Mineral Products Manufacturing
Nursing And Personal Care, Nec
Office Machines
Office Furniture, Except Wood Manufacturing
Oil And Gas Field Services, Nec
Oil and Gas Field Machinery Manufacturing
On-Site Waste Incineration
Open Burning: Forest and Wildfires
Open Burning: Scrap Tires
Ophthalmic Goods
Optical Instruments and Lenses
Ordnance and Accessories Manufacturing
Organic Fibers, Non-cellulosic Manufacturing
Ornamental Nursery Products
Other Structural Clay Products
Paint Application: Medium Shops
Paint Application: Large Shops
Paints and Allied Products Manufacturing
Paper Coated and Laminated, Packaging, nec
Paperboard Mills
Paved Road Dust
Petroleum Refining
Petroleum Refining
Petroleum Refining: Catalytic Cracking Units
Petroleum Refining: Other Petroleum Products
Petroleum Bulk Stations and Terminals
Pharmaceutical Preparations Manufacturing
Phosphatic Fertilizers
Plastics Materials and Resins Manufacturing
Plastics Products Manufacturing
Plastics Foam Products Manufacturing
Plumbing Fixture Fittings and Trim
Porcelain Electrical Supplies
Portland Cement Manufacture: All Fuels
Potash, Soda, And Borate Minerals
Potato Chips and Similar Snacks
Pottery Products, nec
Power Transmission Equipment
Prefabricated Metal Buildings
Prepared Feeds Manufacturing
Primary Aluminum Production
Primary Batteries, Dry and Wet, Manufacturing
Primary Copper
Primary Metal Products Manufacturing
Primary Smelting and Refining of Zinc
Printing Ink
Printing Trades Machinery Manufacturing
Products of Purchased Glass
Pulp mills (2611)
Pulp and Paper: Kraft Recovery Furnaces
Pumps and Pumping Equipment Manufacturing
Radio and Television Communications Equipment (3662)
Radio and Television Communications Equipment (3663)
Railroad Equipment Manufacturing
Railroads, Line-haul Operating
Ready-mixed Concrete
Refrigeration and Heating Equipment Manufacturing
Refuse Systems
Relays and Industrial Controls
Repair services, nec
Residential Bituminous and Lignite Coal Combustion
Residential Care
Residential Distillate Oil Combustion
Residential Anthracite Coal Combustion
Residential Wood/Wood Residue Combustion
Rice Milling
Rubber and Plastic Hose and Belting Manufacturing
Sawmills and Planing Mills, general

LEAD AND COMPOUNDS (CONTINUED)

Schools & Educational Services, Nec
Scrap And Waste Materials
Screw Machine Products, Bolts, etc.
Screw Machine Products Manufacturing
Search and Navigation Equipment
Secondary Aluminum Smelting
Semiconductors and Related Devices
Semivitreous Table & Kitchenware
Service Industry Machinery
Sewage Sludge Incineration
Sewerage Systems
Sheet Metal Work
Ship Building & Repair
Ship Building And Repairing
Signs and Advertising Displays
Silverware and Plated Ware
Skilled Nursing Care Facilities
Small Arms Ammunition
Small Arms
Soap and Other Detergents Manufacturing
Soil Dust
Space Research and Technology
Space Vehicle Parts and Equipment, nec
Space Propulsion Units and Parts Manufacturing
Special Dies, Tools, Jigs and Fixtures
Special Industry Machinery, nec
Special Trade Contractors, nec
Specialty Hospitals Exc. Psychiatric
Speed Changers, Drives, and Gears
Sporting and Athletic Goods Manufacturing
Stainless Steel Manufacture - EAF
Stationary IC Engines - Diesel
Stationary IC Engines - Natural Gas
Steam And Air-conditioning Supply
Steel Pipe and Tubes Manufacturing
Structural Clay Products, nec
Structure Fires
Surface Active Agents Manufacturing
Surface Coating Operations
Surgical and Medical Instruments Manufacturing
Taconite Iron Ore Processing
Telephone and Telegraph Apparatus
Textile Machinery
Tire Cord and Fabric
Tire Manufacturing
Tires and Inner Tubes
Top & body repair and paint shops (1987)
Transformers, Except Electronic
Travel Trailers and Campers Manufacturing
Truck Trailers
Truck and Bus Bodies
Trucking, Except Local
Turbines and Turbine Generator Sets
Turbines - Natural Gas
U.S. Postal Service
Unpaved Road Dust
Unsupported Plastics Profile Shapes
Unsupported Plastics Film & Sheet
Utility Boilers: Coke
Utility Boilers: Natural Gas Combustion
Utility Boilers: Oil Combustion, all types
Valves and Pipe Fittings Manufacturing
Vehicular Lighting Equipment
Vitreous Plumbing Fixtures
Vitreous China Table and Kitchenware
Waste Disposal: Open Burning (all categories)
Water supply
Wood Products
Wool Fiberglass Manufacturing
X-ray Apparatus and Tubes

CADMIUM AND COMPOUNDS

Adhesives and Sealants
Aerospace Industry (Surface Coating)
Agricultural Production
Air, Water, & Solid Waste Management
Aircraft Manufacturing
Aircraft Parts and Equipment Manufacturing
Aircraft And Parts
Airports, Flying Fields, & Services
Aluminum Extruded Products
Aluminum Foundries
Aluminum Foundries (Castings)
Aluminum Die-Castings
Ammunition, Except for Small Arms
Amusement Parks
Animal Cremation
Asphalt Production
Asphalt Roofing Production
Asphalt Paving Mixtures And Blocks
Automotive stampings
Ball and Roller Bearings Manufacturing
Beet Sugar
Black Liquor Combustion
Blast Furnaces and Steel Mills
Boat Building and Repairing
Bolts, Nuts, Rivets and Washers Manufacturing
Brick and Structural Clay Tile
Cadmium Stabilizers for Plastics
Canned specialties
Canned Fruits and Vegetables
Carbon Black Manufacture
Cement, Hydraulic (not subject to Portland Cement Regulation)
Chemical Preparations
Chemical Manufacturing: Cyclic Crude and Intermediate Production
Chromium Plating: Chromic Anodizing Plating
Chromium Metal Plating
Coated Fabrics, not Rubberized, Manufacturing
Commercial Physical Research
Commercial Printing, Letterpress, and Screen
Commercial Printing, Lithographic
Commercial Printing, nec
Commercial/Institutional Boilers: Coal Combustion, all types
Commercial/Institutional Heating: Anthracite Coal Combustion
Commercial/Institutional Heating: Bituminous and Lignite Coal Combustion
Commercial/Institutional Heating: Distillate Oil Combustion
Commercial/Institutional Heating: Natural Gas Combustion
Commercial/Institutional Heating: Residual Oil Combustion
Commercial/Institutional Heating: Wood/Wood Residue Combustion

CADMIUM AND COMPOUNDS (CONTINUED)

Confectionery
Construction Machinery Manufacturing
Construction Sand And Gravel
Construction (SICs 15-17)
Copper Rolling and Drawing
Correctional Institutions
Cotton
Cottonseed Oil Mills
Courts
Crop Services
Crop Preparation Services For Market
Crude petroleum and natural gas
Crude Petroleum Pipelines
Crushed And Broken Limestone
Crushed And Broken Stone, Nec
Crushed And Broken Granite
Custom Compound Purchased Resins Manufacturing
Dehydrated fruits, vegetables, and soups
Depository Institutions
Durable Goods, Nec
Electric and other services combined
Electric services
Electron Tubes Manufacturing
Electronic Computers
Electronic Connectors
Electronic Components, nec
Engine Electric Equipment
Engineering Services
Environmental Controls Manufacturing
Fabricated Structural Metal Manufacturing
Fabricated Metal Products, nec
Ferroalloy Ores, Except Vanadium
Flat Glass
Flour and other grain mill products
Fluid power Valves and Hose Fittings Manufacturing
Food and Agricultural Products: Cotton Ginning
Frozen fruits, fruit juices and vegetables
Funeral Service And Crematories
Gas And Other Services Combined
General Medical & Surgical Hospitals
Glass Containers
Gold Ores
Gray and Ductile Iron Foundries
Grocery Stores
Guided Missiles and Space Vehicles Manufacturing
Halogenated Solvent Cleaners
Hardware Manufacturing
Heating Equipment, Except Electric
Heavy Construction, Nec
Hospitals
Hotels And Motels
Household Cooking Equipment
Human Cremation
Hydrochloric Acid Production
Industrial Boilers: Bituminous and Lignite Coal Combustion
Industrial Boilers: Anthracite Coal Combustion
Industrial Boilers: Coal, all types
Industrial Boilers: Distillate Oil Combustion
Industrial Boilers: Natural Gas Combustion
Industrial Boilers: Residual Oil Combustion
Industrial Boilers: Waste Oil Combustion
Industrial Boilers: Wood/Wood Residue Combustion
Industrial machinery and equipment
Industrial Machinery, nec
Industrial Organic Chemicals Manufacturing
Industrial Sand
Industrial/Utility Dist. Oil/Diesel Turbines
Inorganic Pigments Manufacturing (Cadmium only)
Instruments to Measure Electricity
Intercity & Rural Bus Transportation
Internal Combustion Engine Manufacturing
Iron and Steel Foundries: Steel Foundries
Iron and Steel Forging
Lead Pencils, Art Goods Manufacturing
Life Insurance
Lighting Equipment
Lightweight Aggregate Kilns
Lime
Lubricating Oils and Greases
Malt Beverages
Manufacturing Industries Manufacturing
Marine Cargo Handling
Metal coating and allied services (3479)
Metal Heat Treating Manufacturing
Metal Sanitary Ware Manufacturing
Metal cans (3411)
Mineral Wool
Mineral Wool Manufacturing (includes Wool Fiberglass)
Minerals, Ground or Treated Production
Miscellaneous Nonmetallic Minerals
Miscellaneous Metal Work
Mobile Sources: On- Road Vehicles
Mobile Sources: Non-Road Vehicles and Equipment - Commercial Marine Vessels
MON - Continuous Processes
Motion Picture & Video Production
Motor and Generators Manufacturing
Motor Vehicle Parts and Accessories Manufacturing
National Security
Natural Gas Liquids
Non-road Mobile Vehicles
Non-stainless Steel Manufacture - EAF
Noncommercial research organizations (1987)
Nonferrous Wire Drawing and Insulating
Nonferrous Rolling and Drawing
Nonferrous Foundries, nec
Nonmetallic Mineral Products Manufacturing
Nursing And Personal Care, Nec
Oil And Gas Field Services, Nec
On-Site Waste Incineration
Ordnance and Accessories Manufacturing
Ornamental Nursery Products
Other Cadmium Compound Production
Other Structural Clay Products
Paints and Allied Products Manufacturing
Paved Road Dust
Petroleum Refining
Petroleum Bulk Stations and Terminals
Petroleum Refining
Petroleum Refining: Other Petroleum Products
Pharmaceutical Preparations Manufacturing
Plastics Materials and Resins Manufacturing
Plastics Products Manufacturing
Plastics Foam Products Manufacturing
Portland Cement Manufacture: All Fuels
Prepackaged Software
Prepared Feeds Manufacturing
Pressed and Blown Glass and Glassware Manufacturing

CADMIUM AND COMPOUNDS (CONTINUED)

Primary Copper
Primary Batteries, Dry and Wet, Manufacturing
Primary Metal Products Manufacturing
Pulp mills (2611)
Pulp and Paper: Kraft Recovery Furnaces
Radio and Television Communications Equipment (3663)
Railroads, Line-haul Operating
Raw cane sugar
Ready-mixed Concrete
Refrigeration and Heating Equipment Manufacturing
Refuse Systems
Repair services, nec
Residential Bituminous and Lignite Coal Combustion
Residential Care
Residential Distillate Oil Combustion
Residential lighting fixtures
Residential Anthracite Coal Combustion
Residential Wood/Wood Residue Combustion
Rice Milling
Sawmills and Planing Mills, general
Schools & Educational Services, Nec
Scrap And Waste Materials
Screw Machine Products, Bolts, etc.
Search and Navigation Equipment
Secondary Aluminum Smelting
Secondary Nonferrous Metals Production
Secondary Zinc Production
Semiconductors and Related Devices
Service Industry Machinery
Sewerage Systems
Sheet Metal Work
Ship Building And Repairing
Skilled Nursing Care Facilities
Soap and Other Detergents Manufacturing
Soil Dust
Space Research and Technology
Space Propulsion Units and Parts Manufacturing
Specialty Hospitals Exc. Psychiatric
Stainless Steel Manufacture - EAF
Stationary IC Engines - Diesel
Stationary IC Engines - Natural Gas
Steam And Air-conditioning Supply
Storage Batteries Manufacturing
Structural Clay Products, nec
Telephone and Telegraph Apparatus
Tire Manufacturing
Tires and Inner Tubes
Top & body repair and paint shops (1987)
Travel Trailers and Campers Manufacturing
Trucking, Except Local
Turbines and Turbine Generator Sets
Turbines - Natural Gas
Unpaved Road Dust
Unsupported Plastics Film & Sheet
Utility Boilers: Coal Combustion, all types
Utility Boilers: Coke
Utility Boilers: Natural Gas Combustion
Utility Boilers: Oil Combustion, all types
Valves and Pipe Fittings Manufacturing
Water supply
Wood Products

DIOXINS AND FURANS (MEASURED AS 2,3,7,8-TCDD TEQ)

Carbon Reactivation Furnaces
Commercial/Institutional Heating: Anthracite Coal Combustion
Commercial/Institutional Heating: Bituminous and Lignite Coal Combustion
Commercial/Institutional Heating: Wood/Wood Residue Combustion
Drum and Barrel Reclamation
Hazardous Waste Incineration
Industrial Boilers: Anthracite Coal Combustion
Industrial Boilers: Bituminous and Lignite Coal Combustion
Lightweight Aggregate Kilns
Portland Cement Manufacture: Hazardous Waste-fired
Pulp and Paper: Kraft Recovery Furnaces
Residential Bituminous and Lignite Coal Combustion
Residential Anthracite Coal Combustion
Scrap Tire Combustion
Secondary Copper Smelting
Secondary Lead Smelting
Sewage Sludge Incineration
Utility Boilers: Coke

POLYCYCLIC AROMATIC HYDROCARBONS (MEASURED AS 16-PAH)

Abrasive Products
Adhesives and Sealants
Agricultural Chemicals and Pesticides
Aircraft Parts and Equipment Manufacturing
Aircraft Manufacturing
Aluminum Extruded Products
Aluminum Foundries
Aluminum Sheet, Plate, and Foil manufacturing
Animal Cremation
Asphalt Felts And Coatings
Asphalt Paving Production
Blast Furnaces and Steel Mills
Carbamate Insecticides Production
Carbon and Graphite Products
Cement, Hydraulic (not subject to Portland Cement Regulation)
Chemical Manufacturing: Naphthalene
Chemical Manufacturing: Alkalies and Chlorine
Chemical Manufacturing: Cyclic Crude and Intermediate Production
Chemical Manufacturing: Naphthalene Sulfonates
Chemical Preparations
Cigarette Smoke
Clay Refractories
Coke Ovens: By-product Recovery Plants
Cold Finishing of Steel Shapes
Commercial/Institutional Heating: POTW Digester Gas Combustion
Commercial Physical Research
Commercial Printing, Gravure
Commercial Printing, Letterpress, and Screen

POLYCYCLIC AROMATIC HYDROCARBONS (MEASURED AS 16-PAH) (CONTINUED)

Commercial/Institutional Heating: Anthracite Coal Combustion
Commercial/Institutional Heating: Bituminous and Lignite Coal Combustion
Commercial/Institutional Heating: Distillate Oil Combustion
Commercial/Institutional Heating: Natural Gas Combustion
Commercial/Institutional Heating: Residual Oil Combustion
Commercial/Institutional Heating: Wood/Wood Residue Combustion
Construction Machinery Manufacturing
Drum and Barrel Reclamation
Fabric Dress and Work Gloves
Fabricated Plate Work (Boiler Shops)
Fabricated Metal Products, nec
Fabricated Rubber Products, nec
Fabricated Structural Metal Manufacturing
Ferroalloy Manufacture
Fiber Cans, Drums, and Similar Products
Gray and Ductile Iron Foundries
Gum and Wood Chemical Manufacturing
Hard Surface Floor Coverings Manufacturing
Household Cooking Equipment
Household Appliances Manufacturing
Human Cremation
Hydrochloric Acid Production
Industrial Boilers: Waste Oil Combustion
Industrial Boilers: Anthracite Coal Combustion
Industrial Boilers: Bituminous and Lignite Coal Combustion
Industrial Boilers: Distillate Oil Combustion
Industrial Boilers: Natural Gas Combustion
Industrial Boilers: Residual Oil Combustion
Industrial Boilers: Wood/Wood Residue Combustion
Industrial Gases Manufacturing
Industrial Inorganic Chemical Manufacturing
Integrated Iron and Steel Mills
Internal Combustion Engine Manufacturing
Landfills: Gas Flares
Lubricating Oils and Greases
Manufacturing Industries Manufacturing
Meat Packing Plants
Medical Waste Incineration
Medicinals and Botanicals Manufacturing
Metal Barrels, Drums, and Pails Manufacturing
Metal Coil (Surface Coating)
Metal Doors, Sash, and Trim
Metal Household Furniture
Miscellaneous Plastics Products
Miscellaneous Chemical Products (2890)
Mobile Sources: Non-Road Vehicles and Equipment - Aircraft
Mobile Sources: Non-Road Vehicles and Equipment - Commercial Marine Vessels
Mobile Sources: Non-Road Vehicles and Equipment - Other
Mobile Sources: On- Road Vehicles
Motor Vehicle Equipment
Motor Vehicles and Car Bodies Manufacturing
Municipal Waste Combustion
Naphthalene: Miscellaneous Uses
Needles, Pins, Hooks and Eyes and Similar Notions
Non-stainless Steel Manufacture - EAF
Nonferrous Wire Drawing and Insulating
Nonmetallic Mineral Products Manufacturing
Office Furniture, Except Wood Manufacturing
Oil and Gas Field Machinery Manufacturing
Other Structural Clay Products
Paints and Allied Products Manufacturing
Paper Coated and Laminated, Packaging, nec
Paper Mills
Partitions and Fixtures, Except Wood
Petroleum Bulk Stations and Terminals
Petroleum Refining
Pharmaceutical Preparations Manufacturing
Phthalic Anhydride Production
Plastics Materials and Resins Manufacturing
Plastics Foam Products Manufacturing
Polishes and Sanitation Goods Manufacturing
Potato Chips and Similar Snacks
Primary Nonferrous Metals Production
Public Building and Related Furniture
Publicly Owned Treatment Works (POTWs)
Refrigeration and Heating Equipment Manufacturing
Residential Anthracite Coal Combustion
Residential Bituminous and Lignite Coal Combustion
Residential Distillate Oil Combustion
Residential Natural Gas Combustion
Residential Wood/Wood Residue Combustion
Scrap Tire Combustion
Secondary Lead Smelting
Sewage Sludge Incineration
Sheet Metal Work
Ship Building & Repair
Soap and Other Detergents Manufacturing
Sporting and Athletic Goods Manufacturing
Stainless Steel Manufacture - EAF
Stationary IC Engines - Diesel
Stationary IC Engines - Natural Gas
Surface Active Agents Manufacturing
Tire Manufacturing
Transformers, Except Electronic
Truck and Bus Bodies
Turbines - Natural Gas
Utility Boilers: Coke
Utility Boilers: Coal Combustion, all types
Utility Boilers: Natural Gas Combustion
Utility Boilers: Oil Combustion, all types
Wood Household Furniture Manufacturing
Wood Treatment/Wood Preserving
Yarn Spinning Mills

POLYCHLORINATED BIPHENYLS

Adhesives and Sealants
Air, Water, & Solid Waste Management
Animal Cremation
Commercial Printing, Lithographic
Electric services
Industrial Boilers: Wood/Wood Residue Combustion
Landfills: Gas Flares
Minerals, Ground or Treated Production
Petroleum Refining
Secondary Nonferrous Metals Production
Services, Nec

APPENDIX C
Fish Consumption Advisories Issued for the Great Waters

Waterbody	PCBs	Dioxins	Hg	Cadmium	Chlordane	Toxaphene	Mirex	DDT	Dieldrin	Kepone	Others
Great Lakes											
Lake Superior	•		•		•	•					
Lake Michigan	•		•		•						
Lake Huron	•	•			•						
Lake Erie	•										
Lake Ontario	•	•					•				
Other Great Waters											
Chesapeake Bay											
Potomac River [a]	•	•			•						
James River [a]										•	
Back River [a]					•						
Baltimore Harbor [a]					•						
Anacostia River [a]	•				•						
Lake Champlain, NY	•		•								
NEP and NERR Sites											
Casco Bay, ME (NEP)		•									
Wells, ME (NERR)		•									
Great Bay, NH (NERR)	•										
Great Bay, Little Bay, and Hampton Harbor, NH (NEP)	•										
Massachusetts Bays, MA (NEP)			•								• [b]
Buzzards Bay, MA (NEP)	•		•								

Appendix C
Fish Consumption Advisories Issued for the Great Waters

Waterbody	PCBs	Dioxins	Hg	Cadmium	Chlordane	Toxaphene	Mirex	DDT	Dieldrin	Kepone	Others
Waquoit Bay, MA (NERR)			•								
Narragansett Bay, RI (NEP) (NERR)	•										
Long Island Sound, NY/CT (NEP)	•	•		•							
Peconic Bay, NY (NEP)	•	•		•							
Hudson River, NY (NERR)	•			•							
New York/New Jersey Harbor (NEP)	•	•		•	•						
Barnegat Bay, NJ (NEP)	•										
Jacques Cousteau-Great Bay and Mullica River, NJ (NERR)	•										
Old Woman Creek, OH (NERR)											
Delaware Estuary, DE/NJ/PA (NEP) (NERR)	•	•		•	•						
Delaware Inland Bays, DE (NEP)											
Maryland Coastal Bays, MD (NEP) (includes Assawoman, Isle of Wight, Sinepuxent, Newport, and Chincoteague Bays)											
Chesapeake Bay, MD (NERR)											
Chesapeake Bay, VA (NERR)											
Albemarle-Pamlico Sounds, NC (NEP)		•									
NC Coastal sites (NERR) (4 reserves)											

Waterbody	PCBs	Dioxins	Hg	Cadmium	Chlordane	Toxaphene	Mirex	DDT	Dieldrin	Kepone	Others
North Inlet/Winjah Bay, SC (NERR)											
Ashepoo-Combahee-Edisto Basin, SC (NERR)			•								
Sapelo Island, GA (NERR)											
Indian River Lagoon, FL (NEP)			•								
San Juan Bay, PR (NEP)											
Jobos Bay, PR (NERR)											
Charlotte Harbor, FL (NEP)			•								
Rookery Bay, FL (NERR)			•								
Sarasota Bay, FL (NEP)			•								
Tampa Bay, FL (NEP)			•								
Apalachicola Bay, FL (NERR)			•								
Mobile Bay, AL (NEP)			•								
Weeks Bay, AL (NERR)			•								
Barataria-Terrebonne Estuarine Complex, LA (NEP)			•								
Galveston Bay, TX (NEP)		•	•								
Corpus Christi Bay, TX (NEP)			•								
Kachemak Bay, AK (NERR)											
Padilla Bay, WA (NERR)											
Puget Sound, WA (NEP)	•	•	•								• [c]
Columbia River, OR/WA (NEP)	•	•						•			
Tillamook Bay, OR (NEP)											

Appendix C
Fish Consumption Advisories Issued for the Great Waters

Waterbody	PCBs	Dioxins	Hg	Cadmium	Chlordane	Toxaphene	Mirex	DDT	Dieldrin	Kepone	Others
South Slough, OR (NERR)											
San Francisco Bay, CA (NEP)	•	•	•		•			•	•		• [d]
Elkhorn Slough, CA (NERR)											
Morro Bay, CA (NEP)											
Santa Monica Bay, CA (NEP)	•							•			
Tijuana River, CA (NERR)											

[a] Major tributary rivers to the Chesapeake Bay
[b] Multiple pollutants (unspecified)
[c] Specific embayments of Puget Sound are listed for the following pollutants; creosote, pentachlorophenol, volatile organic compounds (VOCs), tetrachloroethylene, arsenic, metals (unspecified), vinyl chloride, polyaromatic hydrocarbons (PAHs), polynuclear aromatics, and pesticides (unspecified).
[d] Other unspecified pesticides.
Note: Shading is used in rows to designate waters where no advisories were in effect as of December 31, 1997.
Source: U.S. EPA 1998m (Advisories current through December 31, 1997).

APPENDIX D
Names of Numbered Sites from Figure II-17: Locations of Watersheds Designated as Areas of Probable Concern

Map #	Watershed Name	Map #	Watershed Name
1	Charles	49	Watts Bar Lake
2	Cape Cod	50	Lower Clinch
3	Narragansett	51	Middle Tennessee-Chickamauga
4	Hackensack-Passaic	52	Hiwassee
5	Sandy Hook-Staten Island	53	Guntersville Lake
6	Raritan	54	Pickwick Lake
7	Southern Long Island	55	Lower Tennessee-Beech
8	Middle Delaware-Musconetcong	56	Kentucky Lake
9	Lower Delaware	57	Twin Cities
10	Schuylkill	58	Rush-Vermillion
11	Mullica-Toms	59	Buffalo-Whitewater
12	Gunpowder-Patapsco	60	Castle Rock
13	Conococheague-Opequon	61	Copperas-Duck
14	Lower Pee Dee	62	Kishwaukee
15	Seneca	63	Chicago
16	Middle Savannah	64	Des Plaines
17	Lower St. Johns	65	Upper Fox
18	Middle Chattahoochee-Lake Harding	66	Lower Illinois-Senachwine Lake
19	Choctawhatchee Bay	67	Cahokia-Joachim
20	Perdido Bay	68	Big Muddy
21	Mobile Bay	69	Upper Kaskaskia
22	Door-Kewaunee	70	Middle Kaskaskia
23	Menominee	71	Lower Mississippi-Memphis
24	Lower Fox	72	Deer-Steele
25	Little Calumet-Galien	73	Lower Ouachita
26	Pike-Root	74	Lower Calcasieu
27	Milwaukee	75	Lower Mississippi-New Orleans
28	St. Joseph	76	Lower Kansas

Appendix D
Names of Numbered Sites from Figure II-16: Locations of Watershed Designated as Areas of Probable Concern

Map #	Watershed Name	Map #	Watershed Name
29	Manistee	77	Spring
30	Lake St. Clair	78	Lower Neosho
31	Detroit	79	Buffalo-San Jacinto
32	Ottawa-Stony	80	Coeur D'Alene Lake
33	Raisin	81	Lower Yakima
34	Cedar-Portage	82	Lower Willamette
35	Huron-Vermillion	83	Strait of Georgia
36	Black-Rocky	84	Duwamish
37	Ashtabula-Chagrin	85	Puyallup
38	Chautauqua-Conneaut	86	Puget Sound
39	Buffalo-Eighteenmile	87	Tulare-Buena Vista Lakes
40	Niagara	88	Coyote
41	Oak Orchard-Twelvemile	89	San Francisco Bay
42	Upper St. Lawrence	90	Santa Monica Bay
43	Upper Ohio	91	Los Angeles
44	Shenango	92	San Pedro Channel Islands
45	Tuscarawas	93	Seal Beach
46	Vermilion	94	Newport Bay
47	Middle Wabash-Busseron	95	Aliso-San Onofre
48	Holston	96	San Diego

Source: U.S. EPA 1997i