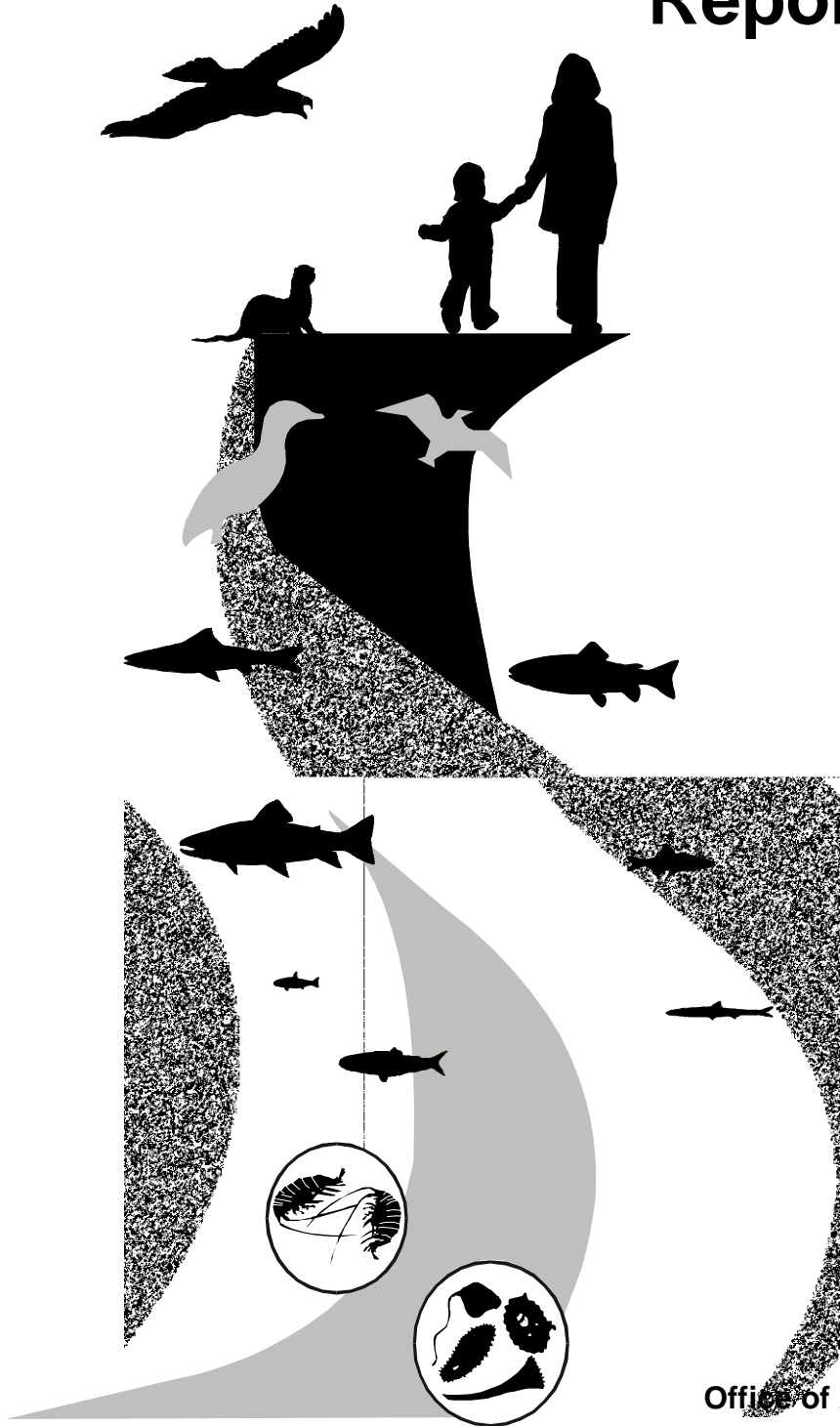


Mercury Study Report to Congress

Volume I:
Executive Summary



Office of Air Quality Planning & Standards
and
Office of Research and Development

MERCURY STUDY REPORT TO CONGRESS

VOLUME I:

EXECUTIVE SUMMARY

December 1997

**Office of Air Quality Planning and Standards
and
Office of Research and Development**

U.S. Environmental Protection Agency

DEDICATION

The U.S. EPA scientists who authored this Report dedicate their efforts to the memory of their colleague, Terry Clark. Terry began his career at the U.S. EPA in 1975, where he became a national, and then an international expert in the atmospheric transport of acid rain and toxic trace gases. Terry designed the initial long-range transport analysis for the Mercury Study. The energy and creativity he brought to his work sustained him even through the final months of his illness when he continued to work daily on this Report. His honesty, intelligence and generosity of spirit are greatly missed. Terry Clark died on January 28, 1994.

TABLE OF CONTENTS

	<u>Page</u>
U.S. EPA AUTHORS	ii
SCIENTIFIC PEER REVIEWERS	iii
WORK GROUP AND U.S. EPA REVIEWERS	vi
LIST OF TABLES AND FIGURES	vii
LIST OF SYMBOLS, UNITS AND ACRONYMS	viii
OVERVIEW	O-1
1. THE MERCURY STUDY REPORT TO CONGRESS	1-1
2. MERCURY IN THE ENVIRONMENT	2-1
3. FINDINGS OF THE MERCURY STUDY REPORT TO CONGRESS	3-1
Sources Contributing to Mercury in the Environment	3-1
Inventory Approach and Uncertainties	3-4
Anthropogenic Emissions Summary	3-5
Trends in Mercury Emissions	3-5
Trends in Mercury Use	3-7
Assessment Approach for Fate and Transport of Mercury	3-8
Assessment of Exposure	3-20
Human Health Effects of Methylmercury	3-22
How Much Methylmercury is Harmful to Humans?	3-25
Levels of Methylmercury Exposure Addressed by the U.S. Food and Drug Administration, World Health Organization and State Recommendations	3-29
Characterization of Risk to Human Populations	3-31
How Much Methylmercury Exposure is Harmful to Wildlife and What Are the Effects? ...	3-43
4. MANAGEMENT ALTERNATIVES	4-1
Possible Control Strategies	4-1
Cost of Controls	4-8
Ongoing U.S. EPA Activities to Reduce Mercury in the Environment	4-10
International Activities	4-14
5. RESEARCH NEEDS	5-1

U.S. EPA AUTHORS

Principal Authors:

Martha H. Keating
Office of Air Quality Planning and Standards
Research Triangle Park, NC

Kathryn R. Mahaffey, Ph.D.
National Center for Environmental Assessment-Washington
Office of Research and Development
Washington, DC

Rita Schoeny, Ph.D.
Office of Water
Washington, DC

Glenn E. Rice
National Center for Environmental Assessment-Cincinnati
Office of Research and Development
Cincinnati, OH

O. Russell Bullock
Atmospheric Sciences Modeling Division
Air Resources Laboratory
National Oceanic and Atmospheric Administration
Research Triangle Park, NC
on assignment to the U.S. EPA National Exposure Research Laboratory

Robert B. Ambrose, Jr., P.E.
Ecosystems Research Division
National Exposure Research Laboratory
Athens, GA

Jeff Swartout
National Center for Environmental Assessment-Cincinnati
Office of Research and Development
Cincinnati, OH

John W. Nichols, Ph.D.
Mid-Continent Ecology Division
Office of Research and Development
Duluth, MN

SCIENTIFIC PEER REVIEWERS

Dr. William J. Adams*
Kennecott Utah Corporation

Dr. Brian J. Allee
Harza Northwest, Incorporated

Dr. Thomas D. Atkeson
Florida Department of Environmental
Protection

Dr. Donald G. Barnes*
U.S. EPA Science Advisory Board

Dr. Steven M. Bartell
SENES Oak Ridge, Inc.

Dr. David Bellinger*
Children's Hospital, Boston

Dr. Nicolas Bloom*
Frontier Geosciences, Inc.

Dr. Mike Bolger
U.S. Food and Drug Administration

Dr. Peter Botros
U.S. Department of Energy
Federal Energy Technology Center

Thomas D. Brown
U.S. Department of Energy
Federal Energy Technology Center

Dr. Dallas Burtraw*
Resources for the Future

Dr. Thomas Burbacher*
University of Washington
Seattle

Dr. James P. Butler
University of Chicago
Argonne National Laboratory

Dr. Elizabeth Campbell
U.S. Department of Energy

Policy Office, Washington, DC

Dr. Rick Canady
Agency for Toxic Substances and Disease
Registry

Dr. Rufus Chaney
U.S. Department of Agriculture

Dr. Joan Daisey*
Lawrence Berkeley National Laboratory

Dr. John A. Dellinger*
Medical College of Wisconsin

Dr. Kim N. Dietrich*
University of Cincinnati

Dr. Tim Eder
Great Lakes Natural Resource Center
National Wildlife Federation for the
States of Michigan and Ohio

Dr. Lawrence J. Fischer*
Michigan State University

Dr. William F. Fitzgerald
University of Connecticut
Avery Point

A. Robert Flaak*
U.S. EPA Science Advisory Board

Dr. Katharine Flegal
National Center for Health Statistics

Dr. Bruce A. Fowler*
University of Maryland at Baltimore

Dr. Steven G. Gilbert*
Biosupport, Inc.

Dr. Cynthia C. Gilmour*
The Academy of Natural Sciences

Dr. Robert Goyer

SCIENTIFIC PEER REVIEWERS (continued)

National Institute of Environmental Health
Sciences

Dr. George Gray
Harvard School of Public Health

Dr. Terry Haines
National Biological Service

Dr. Gary Heinz*
Patuxent Wildlife Research Center

Joann L. Held
New Jersey Department of Environmental
Protection & Energy

Dr. Robert E. Hueter*
Mote Marine Laboratory

Dr. Harold E. B. Humphrey*
Michigan Department of Community Health

Dr. James P. Hurley*
University of Wisconsin
Madison

Dr. Joseph L. Jacobson*
Wayne State University

Dr. Gerald J. Keeler
University of Michigan
Ann Arbor

Dr. Ronald J. Kendall*
Clemson University

Dr. Lynda P. Knobeloch*
Wisconsin Division of Health

Dr. Leonard Levin
Electric Power Research Institute

Dr. Steven E. Lindberg*
Oak Ridge National Laboratory

Dr. Genevieve M. Matanoski*

The Johns Hopkins University

Dr. Margaret McDowell
National Center for Health Statistics

Dr. Thomas McKone*
University of California
Berkeley

Dr. Malcolm Meaburn
National Oceanic and Atmospheric
Administration
U.S. Department of Commerce

Dr. Michael W. Meyer*
Wisconsin Department of Natural Resources

Dr. Maria Morandi*
University of Texas Science Center at Houston

Dr. Paul Mushak
PB Associates

Dr. Christopher Newland*
Auburn University

Harvey Ness
U.S. Department of Energy
Federal Energy Technology Center

Dr. Jerome O. Nriagu*
The University of Michigan
Ann Arbor

William O'Dowd
U.S. Department of Energy
Federal Energy Technology Center

Dr. W. Steven Otwell*
University of Florida
Gainesville

Dr. Jozef M. Pacyna
Norwegian Institute for Air Research

Dr. Ruth Patterson

SCIENTIFIC PEER REVIEWERS (continued)

Cancer Prevention Research Program
Fred Gutchinson Cancer Research Center

Dr. Valerie Thomas*
Princeton University

Dr. Donald Porcella
Electric Power Research Institute

Dr. M. Anthony Verity
University of California
Los Angeles

Dr. Deborah C. Rice*
Toxicology Research Center

Samuel R. Rondberg*
U.S. EPA Science Advisory Board

Charles Schmidt
U.S. Department of Energy

Dr. Pamela Shubat
Minnesota Department of Health

Dr. Ellen K. Silbergeld*
University of Maryland
Baltimore

Dr. Howard A. Simonin*
NYSDEC Aquatic Toxicant Research Unit

Dennis Smith
U.S. Department of Energy
Federal Energy Technology Center

Dr. Ann Spacie*
Purdue University

Dr. Alan H. Stern
New Jersey Department of Environmental
Protection & Energy

Dr. David G. Strimaitis*
Earth Tech

Dr. Edward B. Swain
Minnesota Pollution Control Agency

*With U.S. EPA's Science Advisory Board, Mercury Review Subcommittee

WORK GROUP AND U.S. EPA REVIEWERS

Core Work Group Reviewers:

Dan Axelrad, U.S. EPA
Office of Policy, Planning and Evaluation

Angela Bandemehr, U.S. EPA
Region 5

Jim Darr, U.S. EPA
Office of Pollution Prevention and Toxic
Substances

Thomas Gentile, State of New York
Department of Environmental Conservation

Arnie Kuzmack, U.S. EPA
Office of Water

David Layland, U.S. EPA
Office of Solid Waste and Emergency Response

Karen Levy, U.S. EPA
Office of Policy Analysis and Review

Steve Levy, U.S. EPA
Office of Solid Waste and Emergency Response

Lorraine Randecker, U.S. EPA
Office of Pollution Prevention and Toxic
Substances

Joy Taylor, State of Michigan
Department of Natural Resources

U.S. EPA Reviewers:

Robert Beliles, Ph.D., D.A.B.T.
National Center for Environmental Assessment
Washington, DC

Eletha Brady-Roberts
National Center for Environmental Assessment
Cincinnati, OH

Dianne M. Byrne
Office of Air Quality Planning and Standards
Research Triangle Park, NC

Annie M. Jarabek
National Center for Environmental Assessment
Research Triangle Park, NC

Matthew Lorber
National Center for Environmental Assessment
Washington, DC

Susan Braen Norton
National Center for Environmental Assessment
Washington, DC

Terry Harvey, D.V.M.
National Center for Environmental Assessment
Cincinnati, OH

LIST OF TABLES AND FIGURES

	<u>Page</u>
<u>Tables</u>	
2-1 Range of Mean Mercury Concentrations (ppm) for Major Fish Species in the U.S. in 36 States and DC, 1990-1995	2-7
2-2 Mercury Concentration in Bivalve Mollusks from Mussel Watch Sites (1986-1993)	2-8
2-3 Nationwide Geometric Mean Concentrations of Mercury in Bivalve Mollusks (1986-1993) ..	2-8
2-4 Trends in Mercury Concentrations in Bivalve Mollusks (1986-1993)	2-9
3-1 Best Point Estimates of National Mercury Emission Rates by Category	3-6
3-2 Models Used in the Report to Congress	3-9
3-3 Modeled Mercury Mass Budget in Metric Tons for 1994-1995 Using the Specified Speciation Profiles and 1989 Meteorology	3-14
3-4 WHO Data on Mercury in Hair	3-30
3-5 Body Weights and Fish Consumption Values Used in Exposure Modeling	3-33
3-6 Mercury Concentrations in the Top Ten Types of Fish/Shellfish Consumed by U.S. Residents	3-35
3-7 Wildlife Criteria for Mercury	3-44
4-1 Summary of Mercury Control Techniques for Selected Source Types	4-2
4-2 Potential Mercury Emission Reductions and Costs for Selected Source Categories	4-9
<u>Figures</u>	
2-1 The Global Mercury Cycle	2-2
2-2 Cycling of Mercury in Freshwater Lakes	2-3
2-3 Example Aquatic Food Web	2-4
3-1 Comparison of Estimated Current and Pre-Industrial Mercury Budgets and Fluxes	3-2
3-2 Fate, Transport and Exposure Modeling Conducted in the Combined ISC-3 and RELMAP Local Impact Analysis	3-11
3-3 Total Simulated Wet + Dry Deposition of Mercury in All Forms	3-12
3-4 Distribution of Mercury Concentrations in U.S. EPA-Sampled Fish Tissue Throughout the U.S.	3-34
3-5 Distribution of Fish Consumption Rates of Various Populations	3-38

LIST OF SYMBOLS, UNITS AND ACRONYMS

ATSDR	Agency for Toxic Substances and Disease Registry
BAF	Bioaccumulation factor
bw	Body weight
CAA	Clean Air Act as Amended in 1990
CH ₃ Hg	Monomethylmercury
(CH ₃) ₂ Hg	Dimethylmercury
CSFII	U.S. Department of Agriculture's Continuing Surveys of Individual Food Consumption
H ₂ S	Hydrogen sulfide
Hg ⁰	Elemental mercury
Hg(II)	Mercuric ion (divalent mercury)
HgS	Mercuric sulfide
U.S. EPA	U.S. Environmental Protection Agency
U.S. FDA	U.S. Food and Drug Administration
GACT	Generally available control technology
g	Gram
HAP	Hazardous Air Pollutant
kg	Kilogram (1,000 grams)
km	Kilometer (1,000 meters)
MACT	Maximum achievable control technology
Mg	Megagram (one million grams or one metric ton)
NHANES	National Health and Nutrition Examination Survey
ORD	U.S. EPA Office of Research and Development
PBPK	Physiologically-based pharmacokinetic
pg	Picogram (10 ⁻¹² gram)
ppm	Part per million
RfC	Reference concentration
RfD	Reference dose
µg	Microgram (10 ⁻⁶ gram)
WC	Wildlife criterion
WHO	World Health Organization
WHO/IPCS	World Health Organization's International Programme for Chemical Safety
yr	Year

OVERVIEW

This Mercury Study is a Report to Congress prepared by the U.S. Environmental Protection Agency. It fulfills the requirements of section 112(n)(1)(B) of the Clear Air Act, as amended in 1990. The Report provides an assessment of the magnitude of U.S. mercury emissions by source, the health and environmental implications of those emissions, and the availability and cost of control technologies. As the state-of-the-science for mercury is continuously and rapidly evolving, this Report should be viewed as a “snapshot” of our current understanding of mercury. This Report does not quantify the risk from mercury exposure because of scientific uncertainty in a number of important areas. The Report identifies areas where further research is needed to provide a quantitative risk assessment.

Mercury cycles in the environment as a result of natural and human (anthropogenic) activities. The amount of mercury mobilized and released into the biosphere has increased since the beginning of the industrial age. Most of the mercury in the atmosphere is elemental mercury vapor, which circulates in the atmosphere for up to a year, and hence can be widely dispersed and transported thousands of miles from likely sources of emission. Most of the mercury in water, soil, sediments, or plants and animals is in the form of inorganic mercury salts and organic forms of mercury (e.g., methylmercury). The inorganic form of mercury, when either bound to airborne particles or in a gaseous form, is readily removed from the atmosphere by precipitation and is also dry deposited. Wet deposition is the primary mechanism for transporting mercury from the atmosphere to surface waters and land. Even after it deposits, mercury commonly is emitted back to the atmosphere either as a gas or associated with particles, to be re-deposited elsewhere. As it cycles between the atmosphere, land, and water, mercury undergoes a series of complex chemical and physical transformations, many of which are not completely understood.

Mercury accumulates most efficiently in the aquatic food web. Predatory organisms at the top of the food web generally have higher mercury concentrations. Nearly all of the mercury that accumulates in fish tissue is methylmercury. Inorganic mercury, which is less efficiently absorbed and more readily eliminated from the body than methylmercury, does not tend to bioaccumulate.

Mercury Emissions and Deposition in the U.S.

The best point estimate of annual anthropogenic U.S. emissions of mercury in 1994-1995 is 158 tons. Roughly 87 percent of these emissions are from combustion sources, including waste and fossil fuel combustion. Contemporary anthropogenic emissions are only one part of the mercury cycle. Releases from human activities today are adding to the mercury reservoirs that already exist in land, water, and air, both naturally and as a result of previous human activities. The flux of mercury from the atmosphere to land or water at any one location is comprised of contributions from the natural global cycle including re-emissions from the oceans, regional sources, and local sources. Local sources could also include direct water discharges in addition to air emissions. Past uses of mercury, such as fungicide application to crops are also a component of the present mercury burden in the environment. One estimate of the total annual global input to the atmosphere from all sources including natural, anthropogenic, and oceanic emissions is 5,500 tons. Based on this, U.S. sources are estimated to have contributed about 3 percent of the 5,500 tons in 1995.

A computer simulation of long-range transport of mercury suggests that about one-third (~ 52 tons) of U.S. anthropogenic emissions are deposited, through wet and dry deposition, within the lower 48 States. The remaining two-thirds (~ 107 tons) is transported outside of U.S. borders where it diffuses into the global reservoir. In addition, the computer simulation suggests that another 35 tons of mercury from the global reservoir is deposited for a total deposition of roughly 87 tons. Although this type of

modeling is uncertain, the simulation suggests that about three times as much mercury is being added to the global reservoir from U.S. sources as is being deposited from it. What is not uncertain is that additional emissions to air will contribute to levels in the global reservoir, and concomitant deposition to water bodies.

The highest deposition rates from anthropogenic and global contributions for mercury are predicted to occur in the southern Great Lakes and Ohio River valley, the Northeast and scattered areas in the South, with the most elevated deposition in the Miami and Tampa areas. The location of sources, the chemical species of mercury emitted and the climate and meteorology are key factors in mercury deposition. Humid locations have higher deposition than arid locations.

Public Health Impacts

Epidemics of mercury poisoning following high-dose exposures to methylmercury in Japan and Iraq demonstrated that neurotoxicity is the health effect of greatest concern when methylmercury exposure occurs to the developing fetus. Dietary methylmercury is almost completely absorbed into the blood and distributed to all tissues including the brain; it also readily passes through the placenta to the fetus and fetal brain. The reference dose (RfD) is an amount of methylmercury, which when ingested daily over a lifetime is anticipated to be without adverse health effects to humans, including sensitive subpopulations. At the RfD or below, exposures are expected to be safe. The risk following exposures above the RfD is uncertain, but risk increases as exposures to methylmercury increase.

Extrapolating from the high-dose exposures that occurred in the Iraq incident, the U.S. EPA derived a RfD for methylmercury of 0.1 $\mu\text{g}/\text{kg}$ bw/day. While the U.S. EPA has been advised by scientific reviewers to employ this RfD for this analysis, new data are emerging. Currently ongoing are two large epidemiology studies in the Seychelle Islands and in the Faroe Islands that were designed to evaluate childhood development and neurotoxicity in relation to fetal exposures to methylmercury in fish-consuming populations. Because of various limitations and uncertainties in all of the available data, the U.S. EPA and other Federal agencies intend to participate in an interagency review of the human data on methylmercury, including the most recent studies from the Seychelle Islands and the Faroe Islands. The purposes of this review are to refine the estimates of the level of exposure to mercury associated with subtle neurological endpoints and to further consensus between all of the Federal agencies. After this process, the U.S. EPA will determine if a change in the RfD for methylmercury is warranted.

Fish consumption dominates the pathway for human and wildlife exposure to methylmercury. This study supports a plausible link between anthropogenic releases of mercury from industrial and combustion sources in the United States and methylmercury in fish. However, these fish methylmercury concentrations also result from existing background concentrations of mercury (which may consist of mercury from natural sources, as well as mercury which has been re-emitted from the oceans or soils) and deposition from the global reservoir (which includes mercury emitted by other countries). 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.

Critical elements in estimating methylmercury exposure and risk from fish consumption include the species of fish consumed, the concentrations of methylmercury in the fish, the quantity of fish consumed, and how frequently fish is consumed. The typical U.S. consumer eating fish from restaurants

and grocery stores is not in danger of consuming harmful levels of methylmercury from fish and is not advised to limit fish consumption. The levels of methylmercury found in the most frequently consumed commercial fish are low, especially compared to levels that might be found in some non-commercial fish from fresh water bodies that have been affected by mercury pollution. While most U.S. consumers need not be concerned about their exposure to methylmercury, some exposures may be of concern. Those who regularly and frequently consume large amounts of fish -- either marine species that typically have much higher levels of methylmercury than the rest of seafood, or freshwater fish that have been affected by mercury pollution -- are more highly exposed. Because the developing fetus may be the most sensitive to the effects from methylmercury, women of child-bearing age are regarded as the population of greatest interest. In this Report, an analysis of dietary surveys led the U.S. EPA to conclude that between 1 and 3 percent of women of child-bearing age (i.e., between the ages of 15 and 44) eat sufficient amounts of fish to be at risk from methylmercury exposure, depending on the methylmercury concentrations in the fish. These consumers should be aware of the Food and Drug Administration and State fish advisories that suggest limiting the consumption of contaminated fish. Advisories in the United States have been issued by 39 states and some Tribes, warning against consumption of certain species of fish contaminated with methylmercury.

To the extent that concern is focused on high-end fish and seafood consumers, research is needed on the actual consumption patterns and estimated methylmercury exposure of this subpopulation. In addition, the findings from such research should be validated by analysis of hair samples from a representative sample of members of this subpopulation.

Environmental Impacts

The pattern of mercury deposition nationwide influences which eco-regions and eco-systems will be more highly exposed. Piscivorous (fish-eating) birds and mammals are more highly exposed to mercury than any other known component of aquatic ecosystems. Adverse effects of mercury on fish, birds and mammals include death, reduced reproductive success, impaired growth and development, and behavioral abnormalities.

Mercury contamination has been documented in the endangered Florida panther and the wood stork, as well as populations of loons, eagles, and furbearers such as mink and otter. These species are at high risk of mercury exposure and effects because they either are piscivores or eat piscivores. Concentrations of mercury in the tissues of wildlife species have been reported at levels associated with adverse health effects in laboratory studies with the same species. However, field data are insufficient to conclude whether piscivorous wading birds or mammals have suffered adverse effects due to airborne mercury emissions. Modeling analyses conducted for this Report suggest that it is probable that individuals of some highly exposed wildlife subpopulations are experiencing adverse effects due to airborne mercury emissions.

Mercury Control Technologies

Mercury is widely used in industry because of its diverse properties and serves as a process or product ingredient in several industrial sectors, however, industrial demand for mercury has declined by about 75 percent between 1988 and 1996, due largely to the elimination of mercury additives in paints and pesticides and the reduction of mercury in batteries. Most of the emissions of mercury are produced when waste or fuel containing mercury is burned. The U.S. EPA has already finalized emission limits for municipal waste combustors and medical waste incinerators. As a result, by the year 2000, emissions from these categories will decline at least 90 percent from 1995 levels. In addition, mercury emission limits have been proposed for hazardous waste incinerators.

The largest remaining identified source of mercury emissions are coal-fired utility boilers. Although a number of mercury control technologies are being evaluated for utility boilers, most are still in the research stages, making it difficult to predict final cost-effectiveness as well as the time required to scale-up and commercialize the technologies. Because the chemical species of mercury emitted from boilers varies from plant to plant, there is no single control technology that removes all forms of mercury. There remains a wide variation in the end costs of control measures for utilities and the possible impact of such costs on utilities. Preliminary estimates of national control costs for utility boilers (based on pilot scale data) are in the billions of dollars per year. Ongoing research, as well as research needs related to mercury controls for utilities, are described in the document.

Cost-effective opportunities to deal with mercury during the product life-cycle, rather than just at the point of disposal, need to be pursued. A balanced strategy which integrates end-of-pipe control technologies with material substitution and separation, design-for-environment, and fundamental process change approaches is needed. In addition, international efforts to reduce mercury emissions as well as greenhouse gases will play an important role in reducing inputs to the global reservoir of mercury.

1. THE MERCURY STUDY REPORT TO CONGRESS

The Clean Air Act Amendments of 1990 (CAA) established section 112(n)(1)(B) which requires the United States Environmental Protection Agency (U.S. EPA) to study the impacts of mercury air pollution. In particular, section 112(n)(1)(B) specifies the following:

The Administrator shall conduct, and transmit to the Congress not later than 4 years after the date of enactment of the Clean Air Act Amendments of 1990, a study of mercury emissions from electric utility steam generating units, municipal waste combustion units, and other sources, including area sources. Such study shall consider the rate and mass of such emissions, the health and environmental effects of such emissions, technologies which are available to control such emissions, and the costs of such technologies.

The U.S. EPA designed the Mercury Study to address many different (but linked) types of information:

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

The Report used the above types of information to assess the impact of emissions to air of mercury from a variety of sources. This assessment included judgments as to the potential hazard to humans and wildlife of methylmercury exposure which (as is described in succeeding sections) is largely through the consumption of contaminated fish.

There was no attempt in this Report to do a comparative risk/benefit analysis of fish as an important source of protein and calories in the diet of U.S. populations. Such an analysis would be beyond the scope of the CAA mandate. As emphasized in succeeding sections, the typical U.S. consumer of fish is not in danger of consuming harmful levels of methylmercury and is not being advised to reduce fish consumption.

This Mercury Study Report to Congress fulfills the mandate of section 112(n)(1)(B). The Report is in eight volumes:

- Volume I: Executive Summary
- Volume II: An Inventory of Anthropogenic Mercury Emissions in the United States
- Volume III: Fate and Transport of Mercury in the Environment
- Volume IV: An Assessment of Exposure to Mercury in the United States
- Volume V: Health Effects of Mercury and Mercury Compounds
- Volume VI: An Ecological Assessment for Anthropogenic Mercury Emissions in the United States
- Volume VII: Characterization of Human Health and Wildlife Risks from Mercury Exposure in the United States
- Volume VIII: An Evaluation of Mercury Control Technologies and Costs.

The various analyses documented in this Report were designed and conducted in accordance with accepted guidelines and procedures. For example, the human health risk assessment performed for this Report follows published Guidelines for Risk Assessment (including guidelines on *Exposure Assessment, Developmental Toxicity, Carcinogenicity* and *Germ Cell Mutagenicity*) and uses established methodologies for quantitative assessment of general systemic toxicity (e.g., in the calculation of reference doses (RfDs) and reference concentrations (RfCs)). Moreover, the assessment of ecological effects, presented in Volume VI, follows U.S. EPA's *Framework for Ecological Risk Assessment*. Criteria values for protection of piscivorous wildlife were developed using the methodology developed for the *Great Lakes Water Quality Initiative*.

In 1994, the National Research Council of the National Academy of Sciences, in *Science and Judgment in Risk Assessment*, recommended several areas in which U.S. EPA could improve its risk assessment and risk characterization practices. These recommendations are listed below along with a description of how they were implemented in this Report.

- *Provide an understanding of the type and magnitude of an adverse effect that a specific chemical or emission could cause under particular circumstances.* The Report characterizes both the type and magnitude of health and ecological effects associated with airborne emissions of mercury from anthropogenic sources.
- *Validate methods and models.* All models used for the Report were critiqued by scientific experts and model predictions were compared to measured mercury levels using the most appropriate data available.
- *Describe the basis for default options.* All assumptions are described and justified based on available data. Where appropriate, exposure models were modified to improve assumptions and to focus on areas of prediction where use of model assumptions is most justified.
- *Articulate and prioritize data needs.* The Report includes a section on Research Needs in each volume.
- *Distinguish between variability and uncertainty.* The Report provides discussions that attempt to make these distinctions for the risk results.

- *Perform formal uncertainty analyses.* Uncertainty analyses were formally conducted for the dose-response and exposure assessment steps of the study, and were implicit in weight-of-evidence processes used in the hazard identification step of the human health risk assessment and the problem formulation phase of the ecological risk assessment. Uncertainty also was analyzed quantitatively in other components of the study, such as in the calculation of bioaccumulation factors and the RfD for methylmercury.

2. MERCURY IN THE ENVIRONMENT

As a chemical element, mercury cannot be created or destroyed. The same amount has existed on the planet since the earth was formed. Mercury, however, can cycle in the environment as part of both natural and human (anthropogenic) activities. Measured data and modeling results indicate that the amount of mercury mobilized and released into the biosphere has increased since the beginning of the industrial age.

Several types of emission sources contribute to the total atmospheric loading of mercury. Once in the air, mercury can be widely dispersed and transported thousands of miles from likely emission sources. The distance of this transport and eventual deposition depends on the chemical and physical form of the mercury emitted. Studies indicate that the residence time of elemental mercury in the atmosphere may be on the order of a year, allowing its distribution over long distances, both regionally and globally, before being deposited to the earth. The residence time of oxidized mercury compounds in the atmosphere is uncertain, but is generally believed to be on the order of a few days or less. Even after it deposits, mercury commonly is emitted back to the atmosphere either as a gas or in association with particulates to be re-deposited elsewhere. Mercury undergoes a series of complex chemical and physical transformations as it cycles among the atmosphere, land, and water. Humans, plants and animals are routinely exposed to mercury and accumulate it during this cycle, potentially resulting in a variety of ecological and human health impacts.

Properties and Uses of Mercury

Elemental mercury metal is a heavy, silvery-white liquid at typical ambient temperatures and atmospheric pressures. The vapor pressure of mercury metal is strongly dependent on temperature, and it vaporizes readily under ambient conditions. Most of the mercury encountered in the atmosphere is elemental mercury vapor.

Mercury can exist in three oxidation states: Hg^0 (metallic), Hg_2^{2+} (mercurous) and Hg^{2+} (mercuric). The properties and behavior of mercury depend on the oxidation state. Most of the mercury in water, soil, sediments, or biota (i.e., all environmental media except the atmosphere) is in the form of inorganic mercury salts and organic forms of mercury.

Mercury is widely used because of its diverse properties. In very small quantities, mercury conducts electricity, responds to temperature and pressure changes and forms alloys with almost all other metals. Mercury serves an important role as a process or product ingredient in several industrial sectors.

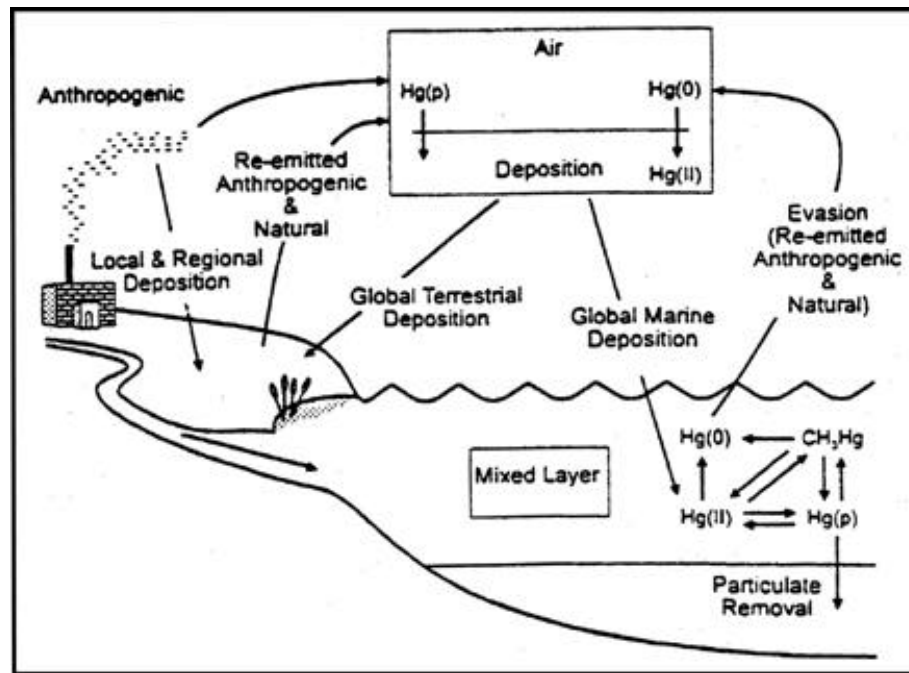
In the electrical industry, mercury is used in components such as fluorescent lamps, wiring devices and switches (e.g., thermostats) and mercuric oxide batteries. Mercury also is used in navigational devices, instruments that measure temperature and pressure and other related uses. It also is a component of dental amalgams used in repairing dental caries (cavities).

In addition to specific products, mercury is used in numerous industrial processes. The largest quantity of mercury used in manufacturing in the U.S. is the production of chlorine and caustic soda by mercury cell chlor-alkali plants. Other processes include amalgamation, use in nuclear reactors, wood processing (as an anti-fungal agent), use as a solvent for reactive and precious metals, and use as a catalyst. Mercury compounds are also frequently added as a preservative to many pharmaceutical products.

The Role of Atmospheric Releases and Processes

A schematic of the most recent conceptualization of the current global mercury cycle is presented in Figure 2-1. As indicated in this figure, mercury is emitted to the atmosphere by a variety of sources, dispersed and transported in the air, deposited to the earth, and stored in or transferred between the land, water, and air.

Figure 2-1
The Global Mercury Cycle



Source: Adapted from Mason, R.P., Fitzgerald, W.F., and Morel, M.M. 1994. The Biogeochemical Cycling of Elemental Mercury: Anthropogenic Influences. *Geochim. Cosmochim. Acta*, 58(15):3191-3198.

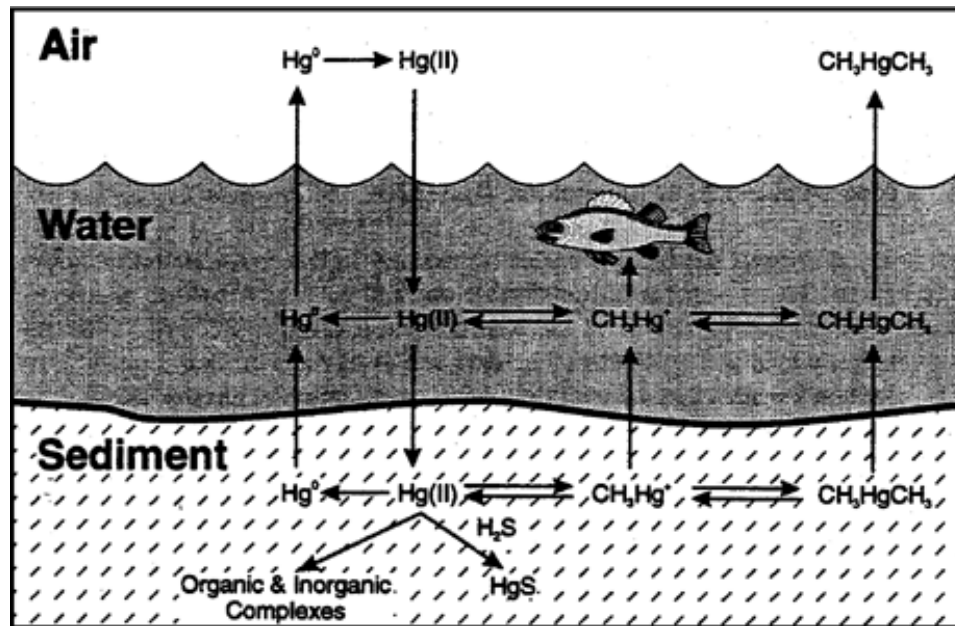
Mercury deposits on the earth in different ways and at different rates, depending on its physical and chemical form. Mercuric species are subject to much faster atmospheric removal than elemental mercury. Mercuric mercury bound to airborne particles and in a gaseous form is readily scavenged by precipitation and is also dry deposited (that is, deposited in the absence of precipitation). In contrast, elemental mercury vapor has a strong tendency to remain airborne and is not susceptible to any major process resulting in direct deposition to the earth's surface. Although much uncertainty still exists, several studies indicate that the relative contribution of mercury loadings to land and water from atmospheric deposition can be substantial.

Numerous studies of elevated mercury levels in remote locations, where atmospheric transport and deposition appears to be the primary mechanism for contamination, provide further evidence of the importance of the atmospheric pathway.

Fate and Transport of Mercury in the Environment

The movement and distribution of mercury in the environment can be confidently described only in general terms. There has been increasing consensus on many, but not all, of the detailed behaviors of mercury in the environment. The depiction of the mercury cycle in Figure 2-2 illustrates the major transfer and transformation processes expected to occur. These processes include a number of infinite and/or indefinite loops.

Figure 2-2
Cycling of Mercury in Freshwater Lakes



Source: Adapted from Winfrey, M.R. and J.W.M. Rudd. 1990. Review -- Environmental Factors Affecting the Formation of Methylmercury in Low pH Lakes. *Environ. Toxicol. Chem.* 9:853-869.

Mercury cycling and partitioning in the environment are complex phenomena that depend on numerous environmental parameters. The following points generally describe the key factors that affect the fate and transport of mercury in the environment.

- The form of mercury in air affects both the rate and mechanism by which it deposits to earth.

- Wet deposition apparently is the primary mechanism for transporting mercury from the atmosphere to surface waters and land.
- Once in aquatic systems, mercury can exist in dissolved or particulate forms and can undergo a number of chemical transformations (see Figure 2-2).
- Contaminated sediments at the bottom of surface waters can serve as an important mercury reservoir, with sediment-bound mercury recycling back into the aquatic ecosystem for decades or longer.
- Mercury has a long retention time in soils. As a result, mercury that has accumulated in soils may continue to be released to surface waters and other media for long periods of time, possibly hundreds of years.

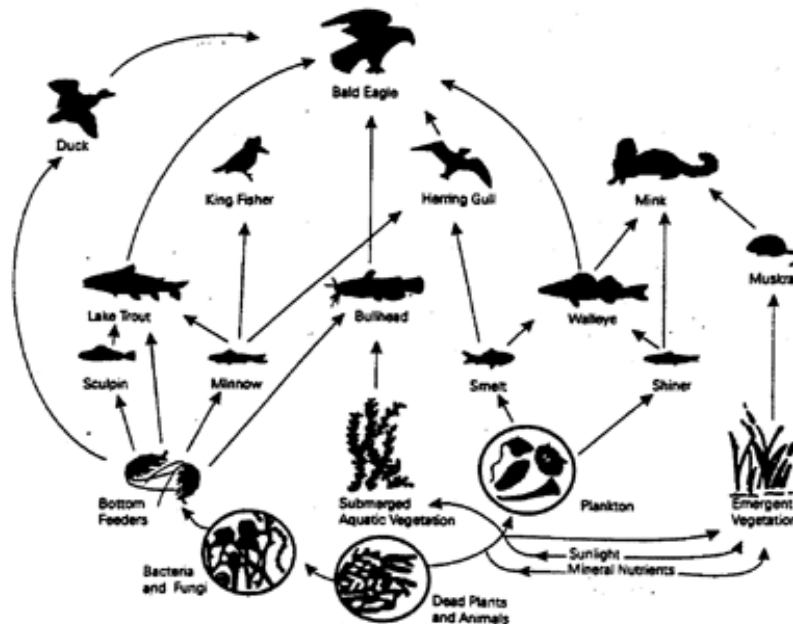
Potential Exposure Pathways

Plants, animals and humans can be exposed to mercury by direct contact with contaminated environmental media or ingestion of mercury-contaminated water and food.

Generally, mercury accumulates up aquatic food chains so that organisms in higher trophic levels have higher mercury concentrations. An example aquatic food web is shown in Figure 2-3. At the top trophic levels are piscivores, such as humans, bald eagles, cormorants, herring gulls and other fish-eating species. The larger wildlife species (e.g., bald eagle, otter) can prey on fish that occupy high trophic levels, such as trout and salmon, which in turn feed on smaller "forage" fish. Smaller piscivorous wildlife (e.g., kingfishers, ospreys) tend to feed on the smaller forage fish, which in turn feed on zooplankton or benthic invertebrates. Zooplankton feed on phytoplankton and the smaller benthic invertebrates feed on algae and detritus. Thus, mercury is transferred and accumulated through several trophic levels.

Figure 2-3

**Example
Aquatic Food
Web**



Mercury Methylation and Bioaccumulation

Methylation of mercury is a key step in the entrance of mercury into food chains. The biotransformation of inorganic mercury species to methylated organic species in water bodies can occur in the sediment and the water column. All mercury compounds entering an aquatic ecosystem, however, are not methylated; demethylation reactions as well as volatilization of dimethylmercury decrease the amount of methylmercury available in the aquatic environment. There is a large degree of scientific uncertainty regarding the rate at which these reactions take place. There is general scientific agreement however that there is significant variability between waterbodies concerning the environmental factors that influence the methylation of mercury.

Nearly 100% of the mercury that bioaccumulates in fish tissue is methylated. Numerous factors in can influence the bioaccumulation of mercury in aquatic biota. These include the acidity of the water (pH), length of the aquatic food chain, temperature and dissolved organic material. Physical and chemical characteristics of a watershed, such as soil type and erosion, affect the amount of mercury that is transported from soils to water bodies. Interrelationships between these factors are poorly understood, however, and there is no single factor (including pH) that has been correlated with mercury bioaccumulation in all cases examined.

Mercury accumulates in an organism when the rate of uptake exceeds the rate of elimination. Although all forms of mercury can accumulate to some degree, methylmercury accumulates to a greater extent than other forms of mercury. Inorganic mercury can also be absorbed but is generally taken up at a slower rate and with lower efficiency than is methylmercury. Elimination of methylmercury takes place very slowly resulting in tissue half-lives (i.e., the time in which half of the mercury in the tissue is eliminated) ranging from months to years. Elimination of methylmercury from fish is so slow that long-term reductions of mercury concentrations in fish are often due mainly to growth of the fish. By comparison, other mercury compounds are eliminated relatively quickly resulting in reduced levels of accumulation.

Methylmercury production and accumulation in the freshwater ecosystem is an efficient process for accumulating mercury which can then be ingested by fish-eating (piscivores) birds, animals and people. In addition, methylmercury generally comprises a relatively greater percentage of the total mercury content at higher trophic levels. Accordingly, mercury exposure and accumulation is of particular concern for animals at the highest trophic levels in aquatic food webs and for animals and humans that feed on these organisms.

Human Exposure Pathways and Health Effects

Humans are most likely to be exposed to methylmercury through fish consumption. Exposure may occur through other routes as well (e.g., the ingestion of methylmercury-contaminated drinking water and food sources other than fish, and dermal uptake through soil and water); however, the fish consumption pathway dominates these other pathways for people who eat fish.

There is a great deal of variability among individuals who eat fish with respect to food sources and fish consumption rates. As a result, there is a great deal of variability in exposure to methylmercury in these populations. The presence of methylmercury in fish is, in part, the result of anthropogenic mercury releases from industrial and combustion sources. As a consequence of human consumption of the affected fish, there is an incremental increase in exposure to methylmercury.

Mercury is a known human toxicant. Clinically observable neurotoxicity has been observed following exposure to high amounts of mercury (for example, "Mad Hatters' Disease"). Consumption of highly contaminated food also has produced overt mercury neurotoxicity. Studies in humans and in experimental animals are described in Volume V of the Mercury Study Report to Congress. Generally, the most subtle indicators of methylmercury toxicity are neurological changes. The neurotoxic effects include subtle decrements in motor skills and sensory ability at comparatively low doses to tremors, inability to walk, convulsions and death at extremely high exposures.

Environmental Impacts

Adverse effects of mercury on fish include death, reduced reproductive success, impaired growth and development and behavioral abnormalities. Exposure to mercury can also cause adverse effects in plants, birds and mammals. Reproductive effects are the primary concern for mercury poisoning and can occur at dietary concentrations well below those which cause overt toxicity. Effects of mercury on birds and mammals include death, reduced reproductive success, impaired growth and development and behavioral abnormalities. Sublethal effects of mercury on birds and mammals include liver damage, kidney damage, and neurobehavioral effects. Effects of mercury on plants include death and sublethal effects. Sublethal effects on aquatic plants can include plant senescence, growth inhibition and decreased chlorophyll content. Sublethal effects on terrestrial plants can include decreased growth, leaf injury, root damage, and inhibited root growth and function.

Concentrations of mercury in the tissues of wildlife species have been reported at levels associated with adverse effects. Toxic effects on piscivorous avian and mammalian wildlife due to the consumption of contaminated fish have been observed in association with point source releases of mercury to the environment. However, field data are insufficient to conclude whether wildlife has suffered adverse effects due to airborne mercury.

Mercury Levels in the United States

Based on 1996 data compiled by U.S. EPA's Office of Water, advisories have been issued in 39 states that warn against the consumption of certain amounts and species of fish that are contaminated with mercury. Ten states have statewide advisories (i.e., advisories posted on every freshwater body in that state). These advisories are based on the results of sampling surveys that measure mercury levels in representative fish species collected from water bodies. The advisories are intended for people who catch and eat fish from those waterbodies.

Table 2-1 presents the range of average mercury concentrations in parts per million (ppm) in major fish species throughout the U.S. (i.e., these are ranges of averages values measured by State agencies across the U.S., not ranges of individual sample values used to calculate the means). This information is based on data which represent the results of fish samples from the District of Columbia and 36 states.

The fish samples were analyzed during the period from 1990 through 1995. The three species of bottom feeders categorized in the table are carp, white sucker and channel catfish. Largemouth bass, smallmouth bass, walleye, brown trout and northern pike represent the major predatory fish species.

Table 2-1
Range of Average Mercury Concentrations (ppm) for Major Fish Species in the U.S. in 36 States and DC, 1990-1995

Carp	0.061 -0.250	White sucker	0.042 - 0.456
Channel catfish	0.010 - 0.890	Largemouth bass	0.101 - 1.369
Smallmouth bass	0.094 - 0.766	Walleye	0.040 - 1.383
Brown trout	0.037 - 0.418	Northern pike	0.084 - 0.531

Fish sold in commerce are under the jurisdiction of the FDA which issues action levels for concentration of mercury in fish and shellfish. The current FDA action level is 1 ppm mercury based on a consideration of health impacts. As illustrated in the table above, freshwater fish can have mercury levels which exceed the U.S. FDA action limit of 1 ppm. The concentration of methylmercury in commercially important marine species is, on the average, close to ten times lower than the FDA action level.

Mercury levels in marine fish have been monitored for at least 20 years by the National Marine Fisheries Service. The data in marine fish have shown mercury levels over this time to be relatively constant in various species. Comparable trends data for freshwater fish do not exist, although there are data for coastal and estuarine sites.

The following information on mercury levels in coastal and estuarine bivalve mollusks (mussels and oysters) is taken from the Mussel Watch Project, which is part of the National Oceanic and Atmospheric Administration's (NOAA's) National Status and Trends (NS&T) Program. The Mussel Watch Project is a large-scale monitoring project that measures concentrations of organic and trace metal contaminants in fresh whole soft-parts of mussels and oysters at over 240 coastal and estuarine sites. These data, which are available for 1986-1993, provide important information about spatial and temporal trends in mercury contamination.

These data are summarized on a regional basis in Table 2-2. Although statistical evaluation has not been conducted, median concentrations along the North Atlantic, Eastern Gulf, and Pacific coasts (0.15, 0.14, and 0.11 ppm dry weight, respectively) appear to be higher relative to those along the Middle Atlantic, South Atlantic, and Western Gulf coasts (0.06, 0.09, and 0.08 ppm dry weight, respectively)¹. The highest mercury concentrations measured exceed 1.0 ppm dry weight at sites along the Western Gulf and Pacific coasts (1.80 and 1.01 ppm dry weight, respectively)¹.

For the purpose of temporal analysis, annual Mussel Watch data on mercury concentrations in bivalve mollusks at specific sites have been aggregated to national geometric means¹. The national means, which are shown in Table 2-3, do not show any temporal trend in mercury concentrations in mussels and oysters for the period 1986-1993.

¹O'Connor, T. P., and B. Beliaeff (1995). Recent Trends in Coastal Environmental Quality: Results from the Mussel Watch Project. 1986 to 1993. U.S. Department of Commerce, National Oceanic and Atmospheric Administration, National Ocean Service, Office of Ocean Resources, Conservation and Assessment, Silver Spring, MD.

Table 2-2
Mercury Concentration in Bivalve Mollusks from Mussel Watch Sites (1986-1993)

Region	States	Concentration Range (ppm-dry weight)	Median Concentration (ppm-dry weight)
North Atlantic	ME, MA , RI, CT, NY, NJ	0.005-0.72	0.15
Middle Atlantic	DE, MD, VA	0.003-0.33	0.06
South Atlantic	NC, SC, GA, FL (east coast)	0.012-0.98	0.09
Eastern Gulf of Mexico	FL (west coast), AL, MS	0.005-0.72	0.14
Western Gulf of Mexico	LA, TX	0.002-1.80	0.08
Pacific	CA, OR, WA, HI, AK	0.002-1.01	0.11

Table 2-3
Nationwide Geometric Mean Concentrations of Mercury in Bivalve Mollusks (1986-1993)

	1986	1987	1988	1989	1990	1991	1992	1993
Mean Mercury Concentration (ppm-dry weight)	0.11	0.11	0.11	0.12	0.09	0.11	0.11	0.12

Temporal trend analysis was also conducted on a site-by-site basis for 154 Mussel Watch sites that had data for at least six years during the period 1986-1993 (O’Conner and Beliaeff 1995). Seven sites exhibited an increasing trend in mercury concentrations, and eight sites exhibited a decreasing trend in mercury concentrations, with 95% statistical confidence. The sites with increasing and decreasing trends are shown in Table 2-4. Many of these sites occur in areas which are heavily industrialized. It is probable that there are point source discharges to these estuaries. The contribution of mercury via air deposition to these sites is unclear.

Table 2-4
Trends in Mercury Concentrations in Bivalve Mollusks (1986-1993)

Site Name	State
Increasing Trend	
Mobile Bay - Hollingers Island Channel	AL
Lake Borgne - Malheureux Point	LA
Galveston Bay - Confederate Reef	TX
Point Loma - Lighthouse	CA
San Francisco Bay - Emeryville	CA
Point Arena - Lighthouse	CA
Crescent - Point St. George	CA
Decreasing Trend	
Charlotte Harbor - Bord Island	FL
Mississippi Sound - Pascagoula Bay	MS
Sabine Lake - Blue Buck Point	TX
Mission Bay - Ventura Bridge	CA
Marina Del Rey - South Jetty	CA
Elliott Bay - Four-Mile Rock	WA
Sinclair Inlet - Waterman Point	WA
Whidbey Island - Possession Point	WA

3. FINDINGS OF THE MERCURY STUDY REPORT TO CONGRESS

Sources Contributing to Mercury in the Environment

In the CAA, Congress directed U.S. EPA to examine sources of mercury emissions, including electric utility steam generating units, municipal waste combustion units and other sources, including area sources. The U.S. EPA interpreted the phrase "... and other sources..." to mean that a comprehensive examination of mercury sources should be made and to the extent data were available, air emissions should be quantified. Volume II of this Report describes in some detail various source categories that emit mercury. In many cases, a particular source category is identified as having the potential to emit mercury, but data are not available to assign a quantitative estimate of emissions. The U.S. EPA's intent was to identify as many sources of mercury emissions to the air as possible and to quantify those emissions where possible.

The mercury emissions data that are available vary considerably in quantity and quality among different source types. Not surprisingly, the best available data are for source categories that U.S. EPA has examined in the past or is currently studying.

Sources of mercury emissions in the United States are ubiquitous. To characterize these emissions, the types are defined in the following way:

- *Natural mercury emissions* -- the mobilization or release of geologically bound mercury by natural processes, with mass transfer of mercury to the atmosphere;
- *Anthropogenic mercury emissions* -- the mobilization or release of geologically bound mercury by human activities, with mass transfer of mercury to the atmosphere; or
- *Re-emitted mercury* -- the mass transfer of mercury to the atmosphere by biologic and geologic processes drawing on a pool of mercury that was deposited to the earth's surface after initial mobilization by either anthropogenic or natural activities.

Contemporary anthropogenic emissions of mercury are only one component of the global mercury cycle. Releases from human activities today are adding to the mercury reservoirs that already exist in land, water, and air, both naturally and as a result of previous human activities. Given the present understanding of the global mercury cycle, the flux of mercury from the atmosphere to land or water at any one location is comprised of contributions from the following:

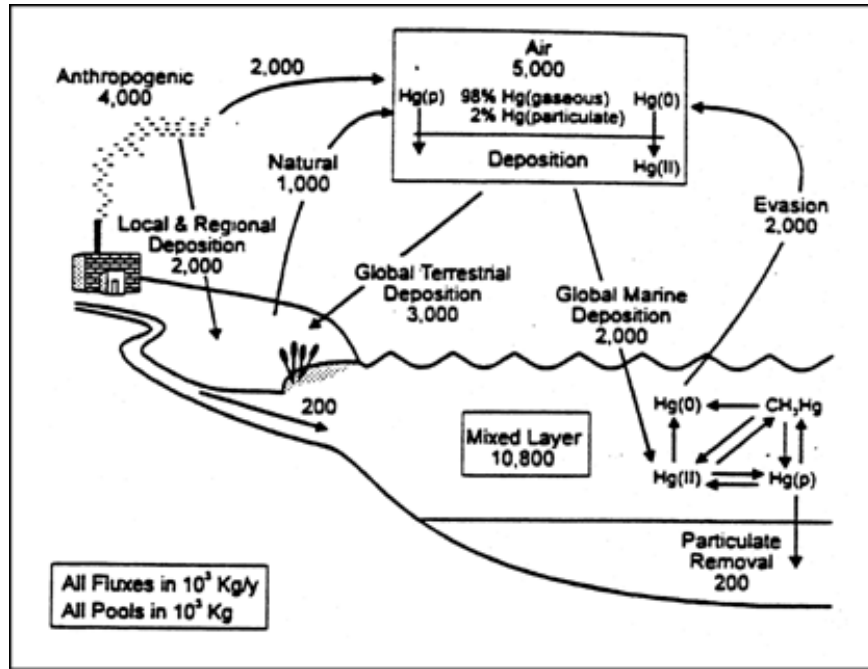
- The natural global cycle,
- The global cycle perturbed by human activities,
- Regional sources, and
- Local sources.

Local sources could also include direct water discharges in addition to air emissions. Past uses of mercury, such as fungicide application to crops are also a component of the present mercury burden in the environment.

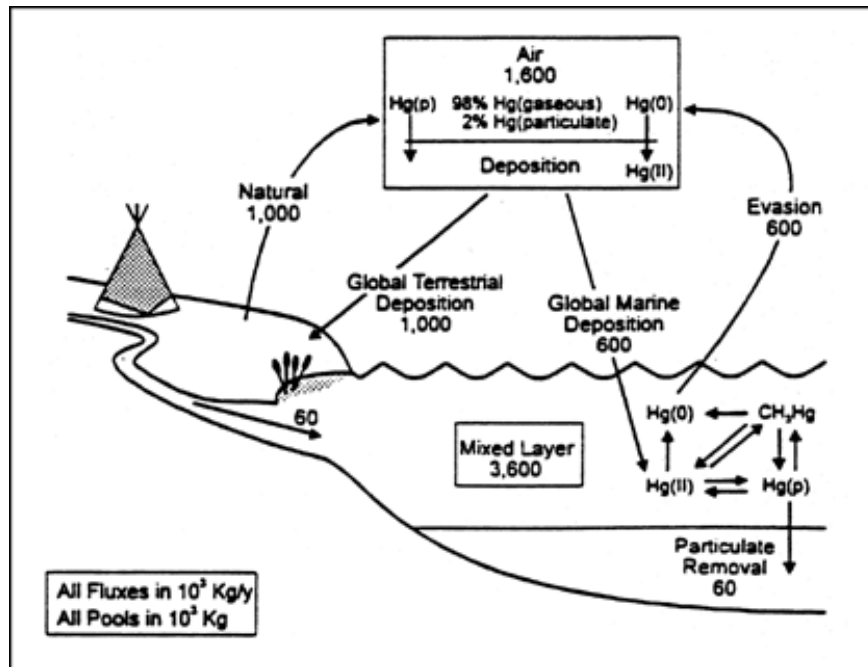
Understanding of the global mercury cycle (shown schematically in Figure 3-1) has improved significantly with continuing study of source emissions, mercury fluxes to the earth's surface, and the

Figure 3-1
 Comparison of Estimated Current and Pre-Industrial Mercury Budgets and Fluxes

Current Mercury Budgets and Fluxes



Pre-Industrial Mercury Budgets and Fluxes



Source: Adapted from Mason, R.P. Fitzgerald, W.F. and Morel, M.M. 1994. The Biogeochemical Cycling of Elemental Mercury: Anthropogenic Influences. *Geochem. Cosmochim. Acta*, 58(15):3191-3198.

magnitude of mercury reservoirs that have accumulated in 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. Recent estimates place the annual amounts of mercury released into the air by human activities at between 50 and 75 percent of the total yearly input to the atmosphere from all sources. Recycling of mercury at the earth's surface, especially from the oceans, extends the influence and active lifetime of anthropogenic mercury releases.

A better understanding of the relative contribution of mercury from anthropogenic sources is also limited by substantial remaining uncertainties regarding the level of natural emissions as well as the amount and original source of mercury that is re-emitted to the atmosphere from soils, watersheds, and ocean waters. Recent estimates indicate that of the approximately 200,000 tons of mercury emitted to the atmosphere since 1890, about 95 percent resides in terrestrial soils, about 3 percent in the ocean surface waters, and 2 percent in the atmosphere. More study is needed before it is possible to accurately differentiate between natural emissions from these soils, watersheds and ocean water and from re-emissions of mercury which originated from anthropogenic sources. For instance, approximately one-third of total current global mercury emissions are thought to cycle from the oceans to the atmosphere and back again to the oceans, but a major fraction of the emissions from oceans consists of recycled anthropogenic mercury. According to the Expert Panel on Mercury Atmospheric Processes² 20 to 30 percent of the oceanic emission is from mercury originally mobilized by natural sources. Similarly, a potentially large fraction of terrestrial and vegetative emissions consists of recycled mercury from previously deposited anthropogenic and natural emissions.

Comparisons of contemporary (within the last 15-20 years) measurements and historical records indicate that the total global atmospheric mercury burden has increased since the beginning of the industrialized period by a factor of between two and five (see Figure 3-1). It is uncertain, however, whether overall atmospheric mercury levels are currently increasing, decreasing or remaining stable. Measurements over remote areas in the Atlantic Ocean show increasing levels up until 1990 and a decrease for the period 1990-1994. At some locations in the upper midwest of the U.S., measurements of deposition rates suggest decreased deposition at some locations. This decrease has been attributed to control of mercury emissions from local or regional sources. However, measurements at remote sites in northern Canada and Alaska show deposition rates that continue to increase. Since these remote sites are subject to global long-range sources rather than regional sources, these measurements may indicate that the global atmospheric burden of mercury is still increasing.

Although the estimated residence time of elemental mercury in the atmosphere is about 1 year, the equilibrium between the atmosphere and ocean waters results in a longer time period needed for overall change to take place in the size of the global reservoir. Therefore, by substantially increasing the size of the oceanic mercury pool, anthropogenic sources have introduced long term perturbations into the global mercury cycle. Fitzgerald and Mason³ estimate that if all anthropogenic emissions were ceased, it would take about 15 years for mercury reservoirs in the oceans and the atmosphere to return to pre-industrial conditions. The Science Advisory Board, in its review of this study concluded that it could take significantly longer. There is scientific agreement however, that the slow release of mercury from

²Expert Panel on Mercury Atmospheric Processes. September 1994. *Mercury Atmospheric Processes: A Synthesis Report*. Electric Power Research Institute. Report No. TR-104214.

³Fitzgerald, W. F., and R. P. Mason. 1996. The Global Mercury Cycle: Oceanic and Anthropogenic Aspects. Pp. 185-108 in Baeyens, W., R. Ebinghaus, and O. Vasiliev, eds., Global and Regional Mercury Cycles: Sources, Fluxes, and Mass Balances.

terrestrial sinks to freshwater and coastal waters will persist for a long time, probably decades, which effectively increases the length of time anthropogenic emissions would impact the environment. This is particularly significant given that the surface soils contain most of the pollution-derived mercury of the industrial period. As a result, it is uncertain at this time how long it would take after reductions in anthropogenic emissions for mercury levels in the global environment, including fish levels, to return to true background levels.

Because of the current scientific understanding of the environmental fate and transport of this pollutant, it is not possible to quantify the contribution of U.S. anthropogenic emissions relative to other sources of mercury, including natural sources and re-emissions from the global pool, on methylmercury levels in seafood and freshwater fish consumed by the U.S. population. Consequently, the U.S. EPA is unable to predict at this time how much, and over what time period, methylmercury concentrations in fish would decline as a result of actions to control U.S. anthropogenic emissions. This is an area of ongoing study.

Inventory Approach and Uncertainties

Given the considerable uncertainties regarding the levels of natural and re-emitted mercury emissions, the emissions inventory focused only on the nature and magnitude of mercury emissions from current anthropogenic sources. The U.S. EPA recognizes, however, that an assessment of the relative public health and environmental impact that can be attributed to current anthropogenic emissions is greatly complicated by both natural mercury emissions, previous emissions of mercury that have subsequently deposited and other sources such as water discharges and other previous uses (e.g., fungicide application). Further study is needed to determine the importance of natural and re-emitted mercury, and the contribution of water discharges relative to atmospheric deposition. Based on estimates of the total annual global input to the atmosphere from all sources (i.e., 5000 Mg from anthropogenic, natural, and oceanic emissions as illustrated by Figure 3-1), U.S. sources are estimated to contribute about 3 percent, based on 1995 emissions estimates as described below.

For most anthropogenic source categories, an emission factor-based approach was used to develop both facility-specific estimates for modeling purposes and nationwide emission estimates. This approach requires an emission factor, which is a ratio of the mass of mercury emitted to a measure of source activity. It also requires an estimate of the annual nationwide source activity level. Examples of measures of source activity include total heat input for fossil fuel combustion and total raw material used or product generated for industrial processes. Emission factors are generated from emission test data, from engineering analyses based on mass balance techniques, or from transfer of information from comparable emission sources. Emission factors reflect the "typical control" achieved by the air pollution control measures applied across the population of sources within a source category.

The emission factor-based approach does not generate exact emission estimates. Uncertainties are introduced in the estimation of emission factors, control efficiencies and the activity level measures. Ideally, emission factors are based on a substantial quantity of data from sources that represent the source category population. For trace pollutants like mercury, however, emission factors are frequently based on limited data that may not have been collected from representative sources. Changes in processes or emission measurement techniques over time may also result in biased emission factors. Emission control estimates are also generally based on limited data; as such, these estimates are imprecise and may be biased. Further uncertainty in the emission estimates is added by the sources of information used on source activity levels, which vary in reliability.

Once emitted to the environment, the fate and transport of mercury is greatly influenced by the chemical form of mercury. The data collected for the emissions inventory was all reported as total mercury with the exception of hazardous waste combustors for which there are site-specific speciated data. For medical waste incinerators and utility boilers there were limited speciated samples from a few facilities. In the exposure analysis described below, estimates were made of speciation profiles for modeling purposes. Speciated data derived from actual monitoring of sources are a critical research need. These data are needed to establish a clear causal link between mercury originating from anthropogenic sources and mercury concentration (projected or actual) in environmental media and/or biota.

To improve the emissions estimates, a variety of other research activities are also needed. These are listed in Chapter 5 of this Volume.

Anthropogenic Emissions Summary

Table 3-1 summarizes the estimated national mercury emission rates by source category. While these emission estimates for anthropogenic sources have limitations, they do provide insight into the relative magnitude of emissions from different groups of sources. All of these emissions estimates should be regarded as best point estimates given available data. Considering the data gaps and other uncertainties in the inventory, the external peer review panel that reviewed this work in January 1995 concluded that missing sources could contribute as much as 20 percent more mercury emissions to the U.S. total. This could affect the relative ranking of the smaller sources.

Of the estimated 144 Megagrams (Mg) (158 tons) of mercury emitted annually into the atmosphere by anthropogenic sources in the United States, approximately 87 percent is from combustion point sources, 10 percent is from manufacturing point sources, 2 percent is from area sources, and 1 percent is from miscellaneous sources. Four specific source categories account for approximately 80 percent of the total anthropogenic emissions--coal-fired utility boilers (33 percent), municipal waste combustion (19 percent), commercial/industrial boilers (18 percent), and medical waste incinerators (10 percent). It should be noted that the U.S. EPA has finalized mercury emission limits for municipal waste combustors and medical waste incinerators. When fully implemented, these emission limits will reduce mercury emissions from these sources by an additional 90 percent over 1995 levels.

All four of the most significant sources represent high temperature waste combustion or fossil fuel processes. For each of these operations, the mercury is present as a trace contaminant in the fuel or feedstock. Because of its relatively low boiling point, mercury is volatilized during high temperature operations and discharged to the atmosphere with the exhaust gas.

Trends in Mercury Emissions

It is difficult to predict with confidence the temporal trends in mercury emissions for the U.S., although there appears to be a trend toward decreasing total mercury emissions from 1990 to 1995. This is particularly true for the waste combustion sources where emissions have declined 50 percent from municipal waste combustors and 75 percent from medical waste incinerators since 1990 (see below). Also, as previously noted, there are a number of source categories where there is insufficient data to estimate current emissions let alone potential future emissions. Based on available information, however, a number of observations can be made regarding mercury emission trends from source categories where some information is available about past activities and projected future activities.

**Table 3-1
Best Point Estimates of National Mercury Emission Rates by Category**

Sources of mercury ^a	1994-1995 Mg/yr ^b	1994-1995 tons/yr ^b	% of Total Inventory ^b
Area sources	3.1	3.4	2.2
Lamp breakage	1.4	1.5	1.0
General laboratory use	1.0	1.1	0.7
Dental preparations	0.6	0.7	0.4
Landfills	<0.1	<0.1	0.0
Mobile sources	c	c	c
Paint use	c	c	c
Agricultural burning	c	c	c
Point Sources	140.9	155.7	97.8
Combustion sources	125.2	137.9	86.9
Utility boilers	47.2	52.0	32.8
Coal	(46.9) ^d	(51.6)	(32.6)
Oil	(0.2)	(0.2)	(0.1)
Natural gas	(<0.1)	(<0.1)	(0.0)
MWCs ^h	26.9	29.6	18.7
Commercial/industrial boilers	25.8	28.4	17.9
Coal	(18.8)	(20.7)	(13.1)
Oil	(7.0)	(7.7)	(4.9)
MWIs ^h	14.6	16.0	10.1
Hazardous waste combustors ^e	6.4	7.1	4.4
Residential boilers	3.3	3.6	2.3
Oil	(2.9)	(3.2)	(2.0)
Coal	(0.4)	(0.5)	(0.3)
SSIs	0.9	1.0	0.6
Wood-fired boilers ^f	0.2	0.2	0.1
Crematories	<0.1	<0.1	0.0
Manufacturing sources	14.4	15.8	10.0
Chlor-alkali	6.5	7.1	4.5
Portland cement ^e	4.4	4.8	3.1
Pulp and paper manufacturing	1.7	1.9	1.2
Instruments manufacturing	0.5	0.5	0.3
Secondary Hg production	0.4	0.4	0.3
Electrical apparatus	0.3	0.3	0.2
Carbon black	0.3	0.3	0.2
Lime manufacturing	0.1	0.1	0.1
Primary lead	0.1	0.1	0.1
Primary copper	<0.1	<0.1	0.0
Fluorescent lamp recycling	<0.1	<0.1	0.0
Batteries	<0.1	<0.1	0.0
Primary Hg production	c	c	c
Mercury compounds	c	c	c
Byproduct coke	c	c	c
Refineries	c	c	c
Miscellaneous sources	1.3	1.4	0.9
Geothermal power	1.3	1.4	0.9
Turf products	g	g	g
Pigments, oil, etc.	g	g	g
TOTAL	144	158	100

^a MWC = Municipal waste combustor; MWI = medical waste incinerator; SSI = sewage sludge incinerator.

^b Numbers do not add exactly because of rounding.

^c Insufficient information to estimate 1994-1995 emissions.

^d Parentheses denote subtotal within larger point source category.

^e For the purpose of this inventory, cement kilns that burn hazardous waste for fuel are counted as hazardous waste combustors.

^f Includes boilers only; does not include residential wood combustion (wood stoves).

^g Mercury has been phased out of use.

^h U.S. EPA has finalized emission guidelines for these source categories which will reduce mercury emissions by at least an additional 90 percent over 1995 levels.

Current emissions of mercury from manufacturing sources are generally low compared to combustion sources (with the exception of chlor-alkali plants using the mercury cell process and portland cement manufacturing plants). The emissions of mercury are more likely to occur when the product (e.g., lamps, thermostats) is broken or discarded. Therefore, in terms of emission trends, one would expect that if the future consumption of mercury remains consistent with the 1996 consumption rate, emissions from most manufacturing sources would remain about the same.

Secondary production of mercury (i.e., recovering mercury from waste products) has increased significantly over the past few years. While 372 Mg of mercury were used in industrial processes in 1996, 446 Mg were produced by secondary mercury producers and an additional 340 Mg were imported. This is a two-fold increase since 1991. The number of secondary mercury producers is expected to increase as more facilities open to recover mercury from fluorescent lamps and other mercury-containing products (e.g., thermostats). As a result there is potential for mercury emissions from this source category to increase.

The largest identified source of mercury emissions during 1994-1995 is fossil fuel combustion by utility boilers, particularly coal combustion. Future trends in mercury emissions from this source category are largely dependent on both the nation's future energy needs and the fuel chosen to meet those needs. Another factor is the nature of actions the utility industry may take in the future to meet other air quality requirements under the Clean Air Act (e.g., national ambient air quality standards for ozone and particulate matter).

Two other significant sources of mercury emissions currently are municipal waste combustors and medical waste incinerators. Emissions from these source categories have declined considerably since 1990 on account of plant closures (for medical waste incinerators) and reduction in the mercury content of the waste stream (municipal waste combustors). Mercury emissions from both of these source types will decline even further by the year 2000 due to regulatory action the U.S. EPA is taking under the statutory authority of section 129 of the CAA. The U.S. EPA has finalized rules for municipal waste combustors and medical waste incinerators that will, when fully implemented, reduce mercury emissions from both of these source categories by an additional 90 percent over 1995 levels. In addition to this federal action, a number of states (including Minnesota, Florida and New Jersey) have implemented mandatory recycling programs to reduce mercury-containing waste, and some states have regulations that impose emission limits that are lower than the federal regulation. These factors will reduce national mercury emissions from these source categories even further.

Trends in Mercury Use

Data on industrial demand for mercury show a general decline in domestic mercury use since demand peaked in 1964. Domestic demand fell by 74 percent between 1980 and 1993, and by more than 75 percent between 1988 and 1996. The rate of decline, however, has slowed since 1990. Further evidence of the declining need for mercury in the U.S. is provided by the general decline in imports since 1988 and the fact that exports have exceeded imports since at least 1989. Federal mercury sales steadily increased from 1988 to 1993, reaching a peak of 97 percent of the domestic demand. However, in July 1994, DLA suspended future sales of mercury from the Department of Defense stockpile until the environmental implications of these sales are addressed. In addition, in past years, DLA sold mercury accumulated and held by the Department of Energy, which is also considered excess to government needs. DLA suspended these mercury sales in July 1993 for an indefinite period in order to concentrate

on selling material from its own mercury stockpile. These suspensions caused federal sales to rapidly decrease to 18 percent of domestic demand in 1994 and to zero since 1995⁴.

For industrial or manufacturing sources that use mercury in products or processes, the overall consumption of mercury is generally declining. Industrial consumption of mercury has declined by about 75 percent between 1988 (1503 Mg) and 1996 (372 Mg). Much of this decline can be attributed to the elimination of mercury as a paint additive and the reduction of mercury in batteries. Use of mercury by other source categories remained about the same between 1988 and 1996.

In general, these data suggest that industrial manufacturers that use mercury are shifting away from mercury except for uses for which mercury is considered essential. This shift is believed to be largely the result of Federal bans on mercury additives in paint and pesticides; industry efforts to reduce mercury in batteries; increasing state regulation of mercury emissions sources and mercury in products; and state-mandated recycling programs. A number of Federal activities are also underway to investigate pollution prevention measures and control techniques for a number of sources categories (see Volume VIII of this Report to Congress).

Assessment Approach for Fate and Transport of Mercury

Study Design of the Fate and Transport Analysis

This analysis relied heavily on computer modeling to describe the environmental fate of emitted mercury because no monitoring data have been identified that conclusively demonstrate or refute a relationship between any of the individual anthropogenic sources in the emissions inventory and increased mercury concentrations in environmental media or biota. To determine if there is a connection between the above sources and increased environmental mercury concentrations, three different models were utilized. Volume III of this Report describes in detail the justification for choices of values for model parameters.

Given the scientific uncertainties associated with environmental mercury, U.S. EPA decided that it was most appropriate to examine the environmental fate of mercury at generalized, rather than specific, sites. A single air model which was capable of modeling both the local as well as regional fate of mercury was not identified. This resulted in the use of two air models: the Regional Lagrangian Model of Air Pollution (RELMAP), for assessing regional scale atmospheric transport, and the Industrial Source Code model (ISC3), for local scale analyses (i.e., within 50 km of a source). To examine the fate of mercury in terrestrial and aquatic environments, U.S. EPA modified an existing generalized watershed and water body fate model. The modified model is identified as IEM-2M. Each of the fate and transport models used in the analysis is summarized in Table 3-2.

⁴Plachy, Jozef, 1997. Mineral Industry Surveys: Mercury Annual Review 1996. Reston, VA. June 1997.

**Table 3-2
Models used in the Report to Congress**

Model	Function
RELMAP	Predict average annual atmospheric mercury concentrations as well as wet and dry deposition flux for 40 Km ² grids across the continental U.S. Model predictions were based on anthropogenic emissions from the sources described in Volume II, Inventory of Anthropogenic Mercury Emissions in the United States.
ISC3	Predict average annual atmospheric mercury concentrations as well as the wet and dry deposition fluxes that result from emissions within 50 Km of a single source.
IEM-2M	Predict environmental media concentrations and the exposures that result from atmospheric mercury concentrations and deposition.

In the first step of this risk assessment, RELMAP was used to simulate the regional-scale transport of anthropogenic mercury emissions over a one-year period. The predicted anthropogenic mercury emissions were added to a uniform elemental mercury background concentration of 1.6 ng/m³ which represented natural and recycled anthropogenic sources of mercury worldwide.

In the second step of the assessment, ISC3 was used to simulate the local-scale transport of anthropogenic mercury emissions. This approach was selected because environmental monitoring studies indicate that measured mercury levels in environmental media and biota may be elevated in areas around stationary industrial and combustion sources known to emit mercury. Rather than use actual facilities for this assessment, a set of model plants was defined to represent typical sources. The source categories evaluated were municipal waste combustors (MWCs), medical waste incinerators (MWIs), coal- and oil-fired utility boilers, and chlor-alkali plants. Two generalized sites where these plants could be located were developed to assess mercury emissions, deposition and subsequent transport through a watershed to a water body. These sites are referred to as the hypothetical western U.S. site and the hypothetical eastern U.S. site. The primary differences between the two hypothetical locations were the assumed erosion characteristics for the watershed and the amount of dilution flow from the water body. Both sites were assumed to have flat terrain for purposes of the atmospheric modeling. The background concentrations in all environmental compartments except for the atmosphere (e.g., soils and sediments) were also assumed to be higher in the eastern U.S. than in the west. The hypothetical eastern and western sites were “placed” at 2.5, 10, and 25 km from the sources (model plants). The ISC3 model predicted mercury air concentrations and deposition rates that resulted from individual model plants at the specified distances.

To estimate the total amount of atmospheric deposition at a site, the 50th or 90th percentile predictions of the RELMAP model for the western or eastern sites were added to the predictions of the local atmospheric model (ISC3) for the individual model plants. These combined model predictions of average atmospheric concentrations and annual-average deposition rates represent the total mercury one might see as a result of both emissions from a single source and impacts from other regional sources.

These estimates were used as inputs to the IEM-2M aquatic and terrestrial fate models at the hypothetical western and eastern U.S. sites.

In the third step of this risk assessment, IEM-2M was utilized to predict the different chemical species of mercury and their concentrations in watershed soils, the water column and sediments of the hypothetical lake, as well as in terrestrial and aquatic organisms. Soil concentrations are used along with vapor concentrations, deposition rates and biotransfer factors to estimate concentrations in various plants. These are used, in turn, along with other biotransfer factors to estimate concentrations in animals. Methylmercury (MHg) concentrations in fish are derived from dissolved MHg water concentrations using bioaccumulation factors (BAF). The BAF accounts for mercury accumulation in organisms that comprise the food web. The BAFs used in this analysis were calculated from existing field data.

A significant input to the IEM-2M model was the estimate of existing mercury concentrations in the environment. To determine existing background concentrations in soil, water, and sediments, U.S. EPA estimated current “background” atmospheric concentrations and deposition rates to the hypothetical western and eastern sites. Each site was then modeled using IEM-2M until equilibrium was achieved with the specified atmospheric background conditions. At both hypothetical sites, the fate of deposited mercury was examined in three different settings: rural (agricultural), lacustrine (around a water body), and urban. The resulting predictions of mercury concentrations in soil, water, and biota were then used to evaluate mercury exposures to humans and wildlife as described in Volumes IV and V of this Report.

Figure 3-2 illustrates the how the various fate models were integrated.

Long-Range Transport Analysis

The long range transport modeling predicts the regional and national deposition of mercury across the continental U.S. Details of several studies which demonstrate the long range transport of mercury are presented in Volume III. In this analysis, the long range transport of mercury was modeled using site-specific, anthropogenic emission source data (presented in Volume II of this Report) to generate average annual atmospheric mercury concentrations and deposition values across the continental U.S. The Regional Lagrangian Model of Air Pollution (RELMAP) was the model selected for this analysis.

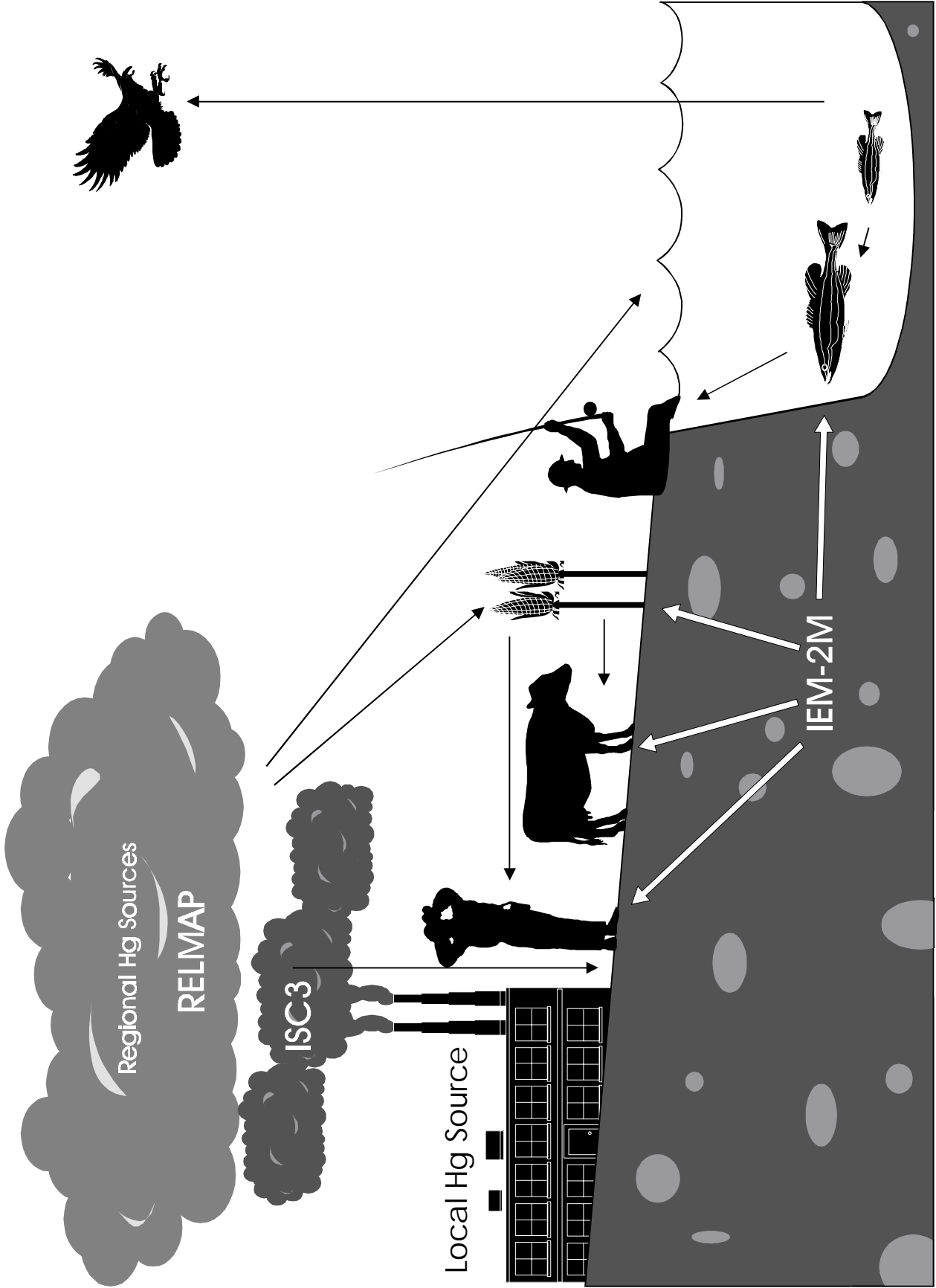
From the RELMAP analysis and a review of field measurement studies, it is concluded that mercury deposition appears to be ubiquitous across the continental U.S., and at, or above, detection limits when measured with current analytic methods. The southern Great Lakes and Ohio River Valley, the Northeast, and scattered areas in the South (particularly in the Miami and Tampa areas) are predicted to have the highest annual rate of deposition of total mercury (above the levels predicted at the 90th percentile). Figure 3-3 illustrates the pattern of mercury deposition across the U.S. This figure also illustrates the boundaries of the RELMAP modeling domain. Measured deposition data are limited, but are available for certain geographic regions. The data that are available corroborate the RELMAP modeling predictions for specific areas. These comparisons are discussed in detail in Volume III.

A wide range of mercury deposition rates is predicted across the continental U.S. The highest predicted rates (i.e., above 90th percentile) are about 20 times higher than the lowest predicted rates (i.e., below the 10th percentile).

The three principal factors that contribute to these modeled and observed deposition patterns are:

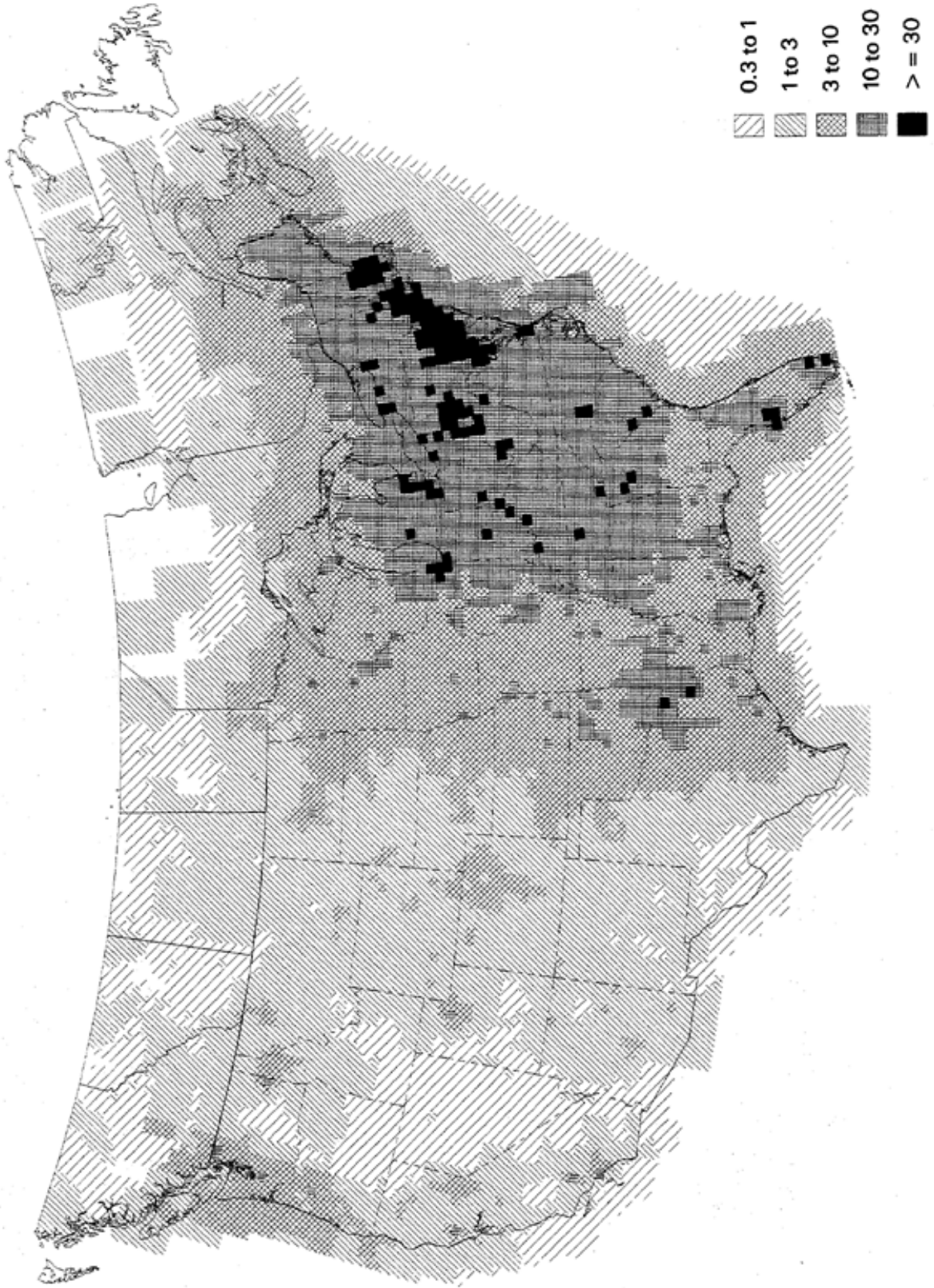
- the emission source locations;

Figure 3-2
Fate, Transport and Exposure Modeling Conducted in the Combined ISC3 and RELMAP Local Impact Analysis



C70032-2

Figure 3-3
Total Simulated Wet + Dry Deposition of Mercury in All Forms
Units: $\mu\text{g}/\text{m}^2$



- the amount of divalent and particulate mercury emitted or formed in the atmosphere; and
- climate and meteorology.

A facility located in a humid climate is predicted to have a higher annual rate of mercury deposition than a facility located in an arid climate.

The critical variables within the model are:

- the estimated washout ratios of elemental and divalent mercury; and
- the annual amount of precipitation.

Precipitation is important because it removes various forms of mercury from the atmosphere and deposits them to the surface of the earth.

Mass Balances of Mercury within the Long-range Model Domain

The chemical form of emitted mercury is a critical factor in its fate, transport and toxicity in the environment. With the exception of hazardous waste incinerators, for which there are site-specific speciated data, mercury emissions are reported as total mercury in all forms. The form distributions, or speciation factors, define the estimated fraction of mercury emitted as elemental mercury (Hg^0), divalent mercury (Hg^{2+}), or mercury associated with particulates (Hg_p). These speciation factors were adopted from Peterson et al.⁵ with adjustments made to reflect the types of air pollution control equipment known to be installed at individual industrial plants. There is considerable uncertainty about the speciation factors for some industrial sources. A wide variety of alternate speciation scenarios have been investigated to measure the sensitivity of the RELMAP results to this uncertainty^{6,7}. The results show that the total simulated wet and dry deposition of mercury to the continental U. S. is strongly and positively correlated to the fraction of mercury emitted as Hg^{2+} and Hg_p for all major source types. The speciation factors used in the RELMAP modeling for this Report are discussed in Volume III.

The results of the RELMAP modeling using these assumed speciation factors are described below. The general mass balance of elemental mercury gas, divalent mercury gas, and particle-bound mercury from the RELMAP simulation results using specified speciation profiles are shown in Table 3-3. Using the meteorologic data from the year 1989, the mass balance shows a total of 141.8 metric tons of mercury emitted to the atmosphere from anthropogenic sources. (This simulated emission total differs from the national totals indicated in Volume II since the states of Alaska and Hawaii are not within the model domain.) The simulation indicates that 47.6 metric tons of anthropogenic mercury emissions are deposited within the model domain and 0.4 metric tons remain in the air within the model domain at the end of the simulation. The remainder, about 93.8 metric tons, is transported outside the model domain and probably diffuses into the global atmospheric reservoir.

⁵Petersen, G., Å. Iverfeldt and J. Munthe. (1995) Atmospheric mercury species over Central and Northern Europe. Model calculations and comparison with observations from the Nordic Air and Precipitation Network for 1987 and 1988. *Atmospheric Environment* 29:47-68.

⁶Bullock, Jr., O. R., W. G. Benjey and M. H. Keating. (1997) Modeling of regional scale atmospheric mercury transport and deposition using RELMAP. *Atmospheric Deposition of Contaminants to the Great Lakes and Coastal Waters: Joel E. Baker, Ed.* pp.323-347. SETAC Press, Pensacola, Florida.

⁷Bullock, Jr., O. R., K. A. Brehme and G. R. Mapp. (1997) Lagrangian modeling of mercury air emission, transport and deposition: An analysis of model sensitivity to emissions uncertainty. *Special Issue on Mercury as a Global Pollutant: Science of the Total Environment*, in press.

The simulation also indicates that 32.0 metric tons of mercury are deposited within the model domain from this global atmospheric reservoir, suggesting that about three times as much mercury is being added to the global reservoir from U.S. emissions as is being deposited from it. The total amount of mercury deposited in the model domain annually from U.S. anthropogenic emissions and from the global background concentration is estimated to be 79.6 metric tons, of which approximately three-fifths is emitted by anthropogenic sources in the lower 48 United States.

Of the total anthropogenic mercury mass deposited to the surface in the model domain, 77% is estimated by the RELMAP to come from Hg^{2+} emissions, 21% from Hg_p emissions and 2% from Hg^0 emissions. When the deposition of Hg^0 from the global background is considered in addition to anthropogenic sources in the lower 48 states, the species fractions of total deposition become 46% Hg^{2+} , 41% Hg^0 and 13% Hg_p . The vast majority of mercury already in the global atmosphere is in the form of Hg^0 and, in general, the anthropogenic Hg^0 emissions do not greatly elevate the Hg^0 air concentration over the global background value. Although Hg^0 is removed from the atmosphere very slowly, the global background reservoir is large and total deposition from it is significant. It should be noted that dry deposition of Hg^0 is thought to be significant only at very elevated concentrations and has not been included in the RELMAP simulations. Wet deposition is the only major pathway for removal of Hg^0 from the atmosphere. This removal pathway simulated by the RELMAP involves oxidation of mercury by ozone in an aqueous solution; thus, the Hg^0 that is extracted from the atmosphere by the modeled precipitation process would actually be deposited primarily in the form of Hg^{2+} .

Table 3-3
Modeled Mercury Mass Budget in Metric Tons for 1994-1995
Using the Specified Speciation Profiles and 1989 Meteorology

Source/Fate	Hg^{0a}	Hg^{2+b}	Hg_p^c	Total Mercury
Total U.S. anthropogenic emissions	63.5	52.3	26.0	141.8
Mass advected from model domain	62.3	15.5	16.0	93.8
Dry deposited anthropogenic emissions	0.0	22.9	0.5	23.4
Wet deposited anthropogenic emissions	0.9	13.8	9.5	24.2
Remaining in air at end of simulation	0.4	<0.1	<0.1	0.4
Total deposited anthropogenic emissions	0.9	36.8	10.0	47.6
Deposited from background Hg^0	32.0	0.0	0.0	32.0
Mercury deposited from all sources	32.9	36.8	10.0	79.6

(All figures rounded to the nearest tenth of a metric ton)

^a Hg^0 = Elemental Mercury

^b Hg^{2+} = Divalent Vapor-phase Mercury

^c Hg_p = Particle-Bound/Mercury

Of the 63.5 metric tons of anthropogenic Hg^0 emitted in the lower 48 states, only 0.9 tons (1.4%) is deposited within the model domain, while of the 52.3 metric tons of Hg^{2+} emitted, about 36.8 tons (70.4%) is deposited. Ninety-eight percent of the deposited anthropogenic mercury was emitted in the form of Hg^{2+} or Hg_p . Thus, a strong argument can be made that the combined Hg^{2+} and Hg_p component of anthropogenic mercury emissions can be used as an indicator of eventual deposition of those emissions to the lower 48 states and surrounding areas. The emission inventory and chemical/physical

speciations profiles indicate that of all combined Hg^{2+} and Hg_p emissions in 1994-1995, about 29% is from electric utility boilers, 25% is from municipal waste combustion, 18% is from medical waste incineration, 16% is from commercial and industrial boilers, and 12% is from all other modeled sources.

Limitations of the Long-Range Transport (RELMAP) Analysis

There are a number of uncertainties with the RELMAP analysis. These have to do to a large degree with the current state-of-the-science concerning atmospheric chemistry and speciation profiles of mercury emissions. Some of the most important limitations are listed below.

- Comprehensive emissions data for a number anthropogenic and natural sources are not available. This reflects the current developmental nature of emission speciation methods, resulting in few data on the various species of mercury and proportions of vapor and solid forms emitted. Both elemental and divalent mercury species as well as gaseous and particulate forms are known to be emitted from point and area sources.
- Atmospheric chemistry data are incomplete. Some atmospheric reactions of mercury, such as the oxidation of elemental mercury to divalent mercury in cloud water droplets have been reported. Other chemical reactions in the atmosphere that may reduce divalent species to elemental mercury have not been reported.
- There is inadequate information on the atmospheric processes that affect wet and dry deposition of mercury. Atmospheric particulate forms and divalent species of mercury are thought to wet and dry deposit more rapidly than elemental mercury; however, the relative rates of deposition are uncertain. There is no validated air pollution model that estimates wet and dry deposition of vapor-phase compounds close to the emission source. In addition, there is uncertainty regarding the revolatilization of deposited mercury.

Analysis of the Local Atmospheric Fate of Mercury

An analysis of the local atmospheric fate of mercury (within 50 km) released from anthropogenic emission sources was undertaken using the ISC-3 model to estimate the impacts of mercury from selected, individual source types. The ISC-3 model was slightly modified to correspond more closely to the chemical properties of atmospheric mercury. This analysis addressed atmospheric mercury emissions from MWCs, coal- and oil-fired utility boilers, MWIs, and chlor-alkali plants. A model plant approach was utilized to develop facilities which represent actual sources from these four categories. The model plants were situated in hypothetical locations intended to simulate a site in either the western or eastern U.S.

The ISC-3 model was used in conjunction with the results from the RELMAP regional scale modeling in order to estimate the air concentrations and deposition rates for each hypothetical facility in each site. Once emitted from a source, mercury may be deposited to the ground via two main processes: wet and dry deposition. Wet deposition refers to the mass transfer of dissolved gaseous or suspended particulate mercury species from the atmosphere to the earth's surface by precipitation, while dry deposition refers to such mass transfer in the absence of precipitation.

The model parameters exerting the most influence on the deposition rates are these:

- total mercury emission rate (grams/second);
- assumptions regarding speciation of the total mercury;
- vapor/particle phase partition estimate;
- stack height for the plant; and
- exit gas velocity.

Combined Results of Local and Regional Scale Analyses

The results of the local scale ISC-3 modeling and the regional scale RELMAP modeling were combined to predict air concentrations and deposition rates for each hypothetical facility in each site. The predicted air concentrations are typically dominated by the regional values, even for the watersheds relatively close to the facility. In general, the predicted average air concentrations are quite low. The only source class for which significantly elevated air concentrations are predicted is the chlor-alkali facility. This is due to a very low stack height coupled with a high assumed mercury emission rate. The low stack height results in predicted plumes that are close to the receptors considered, and so there is less dispersion of the plume compared to the other facilities.

In contrast to the predicted air concentrations, the annual deposition rates are *cumulative*; they represent the sum of any deposition that occurs during the year, and hence are not affected by long periods of little deposition. Further, the ISC3 model predicts that significant deposition events occur infrequently, and it is these relatively rare events that are responsible for the majority of the annual deposition rate. The percentage of mercury deposited within 50 km depends on two main factors: facility characteristics that influence effective stack height (stack height plus plume rise) and the fraction of mercury emissions that is divalent mercury. In most cases, the effective stack height affects only the air concentrations, and hence dry deposition.

For any site with appreciable precipitation, wet deposition can dominate the total deposition for receptors close to the source. Single wet deposition events can deposit 300 times more Hg than a high dry deposition event. These events are even rarer than significant dry deposition events because not only must the wind direction be within a few degrees of the receptor's direction, but precipitation must be occurring as well. The predicted dry deposition rates depend ultimately on the predicted air concentrations. For this reason, dry deposition accounts for most of the total deposition for the facility with the highest predicted air concentrations, the chlor-alkali plant.

In general, 7-45% of the total mercury emitted is predicted to deposit within 50 km at the eastern site in flat terrain, while 2-38% is predicted to deposit at the western site. (The ranges represent values from the different sources considered.) This implies that at least 55% of the total mercury emissions is transported more than 50 km from any of the sources considered, and is consistent with the RELMAP results that predict that mercury may be transported across considerable distances.

The differences between the results for the eastern and western sites are due primarily to the differences in the frequency and intensity of precipitation. At the eastern site, precipitation occurs about 12 % of the year, with about 5% of this precipitation of moderate intensity (0.11 to 0.30 in/hr). By comparison, at the western site, precipitation occurs about 3% of the year, with about 2% of the precipitation of moderate intensity.

Assessment of Watershed Fate

The atmospheric mercury concentrations and deposition rates estimated using the RELMAP and ISC3 were then used as inputs in the watershed model, IEM-2M, to derive calculations of mercury in watershed soils and surface waters. The soil and water concentrations, in turn, drive calculations of concentrations in the associated biota and fish, which humans and other animals are assumed to consume.

IEM-2M is composed of two integrated modules that simulate mercury fate using mass balance equations describing watershed soils and a shallow lake. The mass balances are performed for each mercury component, with internal transformation rates linking Hg^0 , Hg^{2+} , and MeHg. Sources include wetfall and dryfall loadings of each component to watershed soils and to the water body. An additional source is diffusion of atmospheric Hg^0 vapor to watershed soils and the water body. Sinks include leaching of each component from watershed soils, burial of each component from lake sediments, volatilization of Hg^0 and MeHg from the soil and water column, and advection of each component out of the lake.

The nature of this methodology is basically steady with respect to time and homogeneous with respect to space. While it tracks the buildup of soil and water concentrations over the years given a steady depositional load and long-term average hydrological behavior, it does not respond to unsteady loading or meteorological events. There are, thus, limitations on the analysis and interpretations imposed by these simplifications. The model's calculations of average water body concentrations are less reliable for unsteady environments, such as streams, than for more steady environments, such as lakes.

The BAFs were used to estimate fish methylmercury concentrations based on measured concentrations of dissolved methylmercury in the water column. The distribution of the BAFs (Appendix D, Vol. III) was designed to estimate an average concentration of methylmercury in fish of a given trophic level from an average concentration of dissolved methylmercury in the epilimnion for a (single) randomly-selected lake in the continental U.S. The large amount of variability evidenced by the data and reflected in the output distributions arises from several sources, which were not quantified. Much of this variability depends on fish age, model uncertainty, and possibly the use of unrepresentative water column methylmercury measurements in the calculation of the BAFs.

Results of the Watershed Fate and Transport Analysis

For all facilities the contribution of the local source decreases as the distance from the facility increases. With the exception of the chlor-alkali plant, the facilities are generally predicted to contribute less than 50% to the total watershed soil concentration, with RELMAP (representing the regional anthropogenic sources) contributing up to 15% for the RELMAP 50th percentiles, and up to 60% for the RELMAP 90th percentiles.

The results for the MeHg water concentrations and trophic level 4 fish concentrations show a slightly higher contribution from the local sources. While the fractions are similar to those for watershed soil since the watershed serves as a mercury source for the waterbody, these values are slightly higher due to the direct deposition onto the waterbody.

The predicted fruit, leafy vegetable, and beef concentrations are generally dominated by the background values. For plants, this is because these products are assumed to take up most of the mercury from the air, and the local source usually does not impact the local air concentrations significantly. The exception is the chlor-alkali plant, for which the low stack results in higher mercury

air concentrations. The results for the beef concentrations are similar; however, there is a slightly higher contribution from the local source because the cattle are exposed through the ingestion of soil.

IEM-2M Model Sensitivity

For a specific atmospheric deposition rate, mercury concentrations in watersheds and water bodies can vary significantly. Several intrinsic and extrinsic watershed and water body characteristics influence the mercury concentrations in soil, water, and fish. These should cause significant variability in mercury concentrations between regions and among individual lakes within a region.

Mercury concentrations in watershed soils are strongly influenced by atmospheric loading and soil loss processes. The influence of plant canopy and roots in mediating both the loading to the soil and the loss from the soil is not well characterized at present, although published studies indicate its potential importance. Reduction of HgII in the upper soil layer appears to control the volatile loss of mercury, and variations in this reaction can cause significant variations in soil mercury levels. The factors controlling mercury reduction are not well characterized at present. Soil erosion from a watershed can vary more than 3 orders of magnitude depending on rainfall patterns, soil type, topography, and plant cover. High levels of soil erosion should significantly diminish soil mercury concentrations. Runoff and leaching are not expected to affect soil mercury concentrations significantly.

Total mercury concentrations in a water body are strongly influenced by atmospheric loading and, for drainage lakes, by watershed loading. Variations in watershed size and erosion rates can cause significant variability in lake mercury levels. Hydraulic residence time, the water body volume divided by total flow, affects the maximum possible level of total water column mercury for a given loading rate. Parameters controlling mercury loss through volatilization and net settling can also cause significant variations among lakes. Mercury loss through settling is affected by *in-situ* productivity, by the supply of solids from the watershed, and by the solids-water partition coefficient. DOC concentrations can significantly affect partitioning, and thus overall mercury levels. Mercury loss through volatilization is controlled by the reduction rate, which is a function of sunlight and water clarity. Reduction may also be controlled by pH, with lower pH values inhibiting this reduction, leading to higher total mercury levels.

Fish mercury levels are strongly influenced by the same factors that control total mercury levels. In addition, fish concentrations are sensitive to methylation and demethylation in the water column and sediments. A set of water body characteristics appear to affect these reactions, including DOC, sediment TOC, sunlight, and water clarity. Variations in these properties can cause significant variations in fish concentrations among lakes. Other factors not examined here, such as anoxia and sulfate concentrations, can stimulate methylation and lead to elevated fish concentrations. Fish mercury levels are sensitive to factors that promote methylmercury mobility from the sediments to the water column; these factors include sediment DOC and sediment-pore water partition coefficients.

Limitations of the Local Scale and Watershed Analyses

There are limitations associated with the fate and transport analyses. These have to do to a large degree with the current state-of-the-science concerning mercury fate and transport in the terrestrial and aquatic environments and variability between waterbodies. Some of the most important limitations are listed below.

- There is a lack of information characterizing the movement of mercury from watershed soils to water bodies and the rates at which mercury converts from one chemical species to another. There appears to be a great deal of variability in these factors among watersheds.

- There are not conclusive data on the amount of and rates of mercury methylation in different types of water bodies. In addition, there is a lack of data on the transfer of mercury between environmental compartments and biologic compartments; for example, the link between the amount of mercury in the water body and the levels in fish appears to vary from water body to water body.
- There is a lack of adequate mercury measurement data near mercury sources. Measurement data are needed to assess how well the modeled data predict actual mercury concentrations in different environmental media at a variety of geographic locations. Missing data include measured mercury deposition rates and measured concentrations in the atmosphere, soils, water bodies and biota.
- The IEM-2M has not been validated with site-specific data. The model was benchmarked against the independently-derived R-MCM, which itself has been calibrated to several Wisconsin lakes. When driven by the same atmospheric loading and solids concentrations, IEM-2M predictions of mercury concentrations compare well with those calculated by R-MCM for a set of Wisconsin lakes.

Conclusions Regarding Mercury Fate and Transport in the Environment

The uncertainty inherent in the modeled estimates arises from many individual assumptions present within the three models. Because of these uncertainties, U.S. EPA interpreted the model results qualitatively rather than quantitatively as follows.

The analysis of mercury fate and transport, in conjunction with available scientific knowledge, supports a plausible link between mercury emissions from anthropogenic combustion and industrial sources and mercury concentrations in air, soil, water and sediments. The critical variables contributing to this linkage are these:

- the species of mercury that are emitted from the sources, with Hg⁰ mostly contributing to concentrations in ambient air and Hg²⁺ mostly contributing to concentrations in soil, water and sediments;
- the overall amount of mercury emitted from a combustion source;
- the watershed soil loss rates, including reduction and erosion;
- the water body loss rates, including outflow, reduction, and settling; and
- the climate conditions.

In addition, the analysis of mercury fate and transport supports a plausible link between mercury emissions from anthropogenic combustion and industrial sources and methylmercury concentrations in freshwater fish. The critical variables contributing to this linkage are the following:

- the species of mercury that are emitted, with emitted divalent mercury mostly depositing into local watershed areas and, to a lesser extent the atmospheric conversion of elemental mercury to divalent species which are deposited over greater distances;
- the overall amount of mercury emitted from a source;
- the watershed soil loss rates, including reduction and erosion;

- the water body loss rates, including outflow, reduction, and settling;
- the extent of mercury methylation in the water body;
- the extent of food web bioaccumulation in the water body; and
- the climate conditions.

From the analysis of deposition and on a comparative basis, the deposition of Hg^{2+} close to an emission source is greater for receptors in elevated terrain (i.e., terrain above the elevation of the stack base) than from receptors located in flat terrain (i.e., terrain below the elevation of the stack base). The critical variables are parameters that influence the plume height, primarily the stack height and stack exit gas velocity.

On a national scale, an apportionment between sources of mercury and mercury in environmental media and biota cannot be described in quantitative terms with the current scientific understanding of the environmental fate and transport of this pollutant.

Assessment of Exposure

The exposure Volume consists of two parts; the first examines exposures predicted to result from the emitted mercury, and the second estimates exposures that result from seafood consumption. The first part of the exposure assessment draws upon the modeling analyses described above which assessed the long range transport of mercury from emission sources through the atmosphere, the transport of mercury from emission sources through the local atmosphere, and the aquatic and terrestrial fate and transport of mercury at hypothetical sites. The exposure assessment used the results of the atmospheric, terrestrial and aquatic models to estimate the resulting exposures to humans and animals that were assumed to inhabit the hypothetical sites explained above. In the second part of the exposure Volume, exposure to mercury from seafood was estimated using various dietary surveys and measurements of mercury concentrations in seafood.

The exposure assessment, which was based on environmental fate and exposure modeling, addressed atmospheric mercury emissions from the four sources described earlier; MWCs, MWIs, utility boilers and chlor-alkali plants. It did not address all anthropogenic emission sources. In addition, anthropogenic discharges of mercury to waterbodies were not addressed.

Human Exposure

The following human exposure routes were included: inhalation, consumption of water, consumption of fish, beef, beef liver, cow's milk, poultry, chicken eggs, pork, lamb, green plants (e.g., leafy vegetables, potatoes, fruits, grains and cereals) and ingestion of soil. Dermal exposures that resulted from contact with soil and water, as well as exposure through inhalation of resuspended dust particles and exposure through the consumption of human breast milk were not evaluated. The only exposure route considered for wildlife was the consumption of freshwater fish.

Consumption of fish is the dominant pathway of exposure to methylmercury for fish-consuming humans and wildlife. There is a great deal of variability among individuals in these populations with respect to food sources and fish consumption rates. As a result, there is a great deal of variability in exposure to methylmercury in these populations. The anthropogenic contribution to the total amount of methylmercury in fish is predicted to be, in part, the result of anthropogenic mercury releases from industrial and combustion sources increasing mercury body burdens in fish. Existing background

mercury concentrations are also predicted to contribute to methylmercury concentrations in fish. As a consequence of human and wildlife consumption of the affected fish, there is an incremental increase in exposure to methylmercury. Due to differences in fish consumption rates per body weight and differences in body weights among species, it is likely that piscivorous birds and mammals have much higher environmental exposures to methylmercury than humans through the consumption of contaminated fish. This is true even in the case of fish consumption by humans who consume above average amounts of fish. The critical variables contributing to these outcomes are these:

- the fish consumption rate;
- the body weight of the individual in relation to the fish consumption rate; and
- the rate of biomagnification between trophic levels within the aquatic food-chain.

A current assessment of U.S. general population methylmercury exposure through the consumption of fish is provided in Chapter 4 of Volume IV. This assessment was conducted to provide an estimate of mercury exposure through the consumption of fish to the general U.S. population. It is a national assessment rather than a site-specific assessment. This assessment utilizes data from the Continuing Surveys of Food Intake by Individuals (CSFII 89-91, CSFII 1994, CSFII 1995) and the third National Health and Nutrition Examination Survey (NHANES III) to estimate a range of fish consumption rates among U.S. fish and shellfish eaters. Per capita, per user (only individuals who reported fish consumption during the survey period) based on a single-day's intake, and month-long per user were considered. The month-long per user projections reflect the combined frequency distributions of NHANES III frequency of fish/shellfish consumption data and single day's data for per user consumption patterns. For each fish-eater, the number of fish meals, the quantities and species of fish consumed and the self-reported body weights were used to estimate mercury exposure on a body weight basis. The constitution of the survey population was weighted to reflect the actual U.S. population. Results of smaller surveys on "high-end" fish consumers are also included.

These estimates of fish consumption rates were combined with species-specific mean values for measured methylmercury concentrations. The marine fish methylmercury concentration data were obtained from the National Marine Fisheries Service Database. The freshwater fish methylmercury concentration data were obtained from Bahnick et al., (1994) and Lowe et al., (1985). Through the application of specific fish preparation factors (USDA, 1995), estimates of the range of methylmercury exposure from the consumption of fresh water fish were prepared for the fish-consuming segment of the U.S. population. Per body weight estimates of methylmercury exposure were determined by dividing the total daily methylmercury exposure from this pathway by the self-reported body weights of individuals in the USDA surveys and recorded body weights in the third NHANES data. The species of fish/shellfish consumed by children were identified from the 24-hour recalls on children in the USDA surveys and in the third NHANES.. The results of this analysis show that on a per kilogram body weight basis children have higher average exposure rates to methylmercury through the consumption of fish than adults. The higher exposures to children are considered biologically meaningful because month-long mercury exposures considerably in excess of the RfD are observed among some children. At the RfD or below, exposures are expected to be safe. The risk following exposures above the RfD is uncertain, but risk increases as exposures to methylmercury increase.

Wildlife Exposure

In terms of predicted methylmercury intake on a per body weight basis, the six wildlife species considered in this analysis can be ranked from high to low as follows:

- Kingfisher
- River Otter

- Loon, Mink, Osprey
- Bald Eagle

Methylmercury exposures for the most exposed wildlife species (the kingfisher) may be up to two orders of magnitude higher than human exposures from contaminated freshwater fish (on a kilogram fish consumed per body weight basis). This assumes that the fish within different trophic levels of a given lake are contaminated with the same concentrations of methylmercury.

Human Health Effects of Methylmercury

Data in both humans and experimental animals show that all three forms of mercury evaluated in this Report (elemental, inorganic and methylmercury) can produce adverse health effects at sufficiently high doses. Human exposure to elemental mercury occurs in some occupations, and exposure to inorganic mercury can arise from mercury amalgams used in dental restorative materials (U.S. PHS, Environmental Health Policy Committee, 1995). People, however, are primarily exposed to methylmercury in fish. The focus of this assessment, therefore, is on methylmercury, which can produce a variety of adverse effects, depending on the dose and time of exposure.

Individual risk assessors for specific organizations may apply risk assessment differently. Identification of subpopulations of concern is one of the decisions in the risk assessment process. Because methylmercury is a neurotoxin (particularly to the developing nervous system), the fetus and young child are of particular interest. More than one approach to selection of the population at risk of adverse effect is feasible. The RfD of 0.1 µg/kg bw/day was based on neurotoxic effects of methylmercury to the developing nervous system. Because nervous system development continues into postnatal life the young child may also be a subpopulation of interest. If children are judged to be a subpopulation of concern, specific age-groups within this subpopulation may be judged to be of greater interest; e.g., birth through 4 years of age. Alternatively other risk assessors may prefer to consider all children (e.g., birth through 14 years of age) as a group when evaluating risk to children.

Methylmercury-induced neurotoxicity is the effect of greatest concern when exposure occurs to the developing fetus. The RfD is a dose of methylmercury that is protective of the developing fetal nervous system. Post-natal brain development continues well into childhood. Methylmercury exposure adversely affects a number of cellular events in the developing brain both in utero and post-natally. The post-natal age when development of various regions of the brain is completed varies, but development of many functions continues through the first four to six years of life. The RfD of 0.1 µg/kg-bw/day (protective of fetal brain development) is anticipated to be protective of brain development in the young child.

Toxicokinetics of Mercury

The toxicokinetics (i.e., absorption, distribution, metabolism, and excretion) of mercury is highly dependent on the form of mercury to which a receptor has been exposed.

The absorption of elemental mercury vapor occurs rapidly through the lungs, but it is poorly absorbed from the gastrointestinal tract. Once absorbed, elemental mercury is readily distributed throughout the body; it crosses both placental and blood-brain barriers. The distribution of absorbed elemental mercury is limited primarily by the oxidation of elemental mercury to the mercuric ion as the mercuric ion has a limited ability to cross the placental and blood-brain barriers. Once elemental mercury crosses these barriers and is oxidized to the mercuric ion, return to the general circulation is impeded, and mercury can be retained in brain tissue. Elemental mercury is eliminated from the body via urine, feces, exhaled air, sweat, and saliva. The pattern of excretion changes depending upon the extent the elemental mercury has been oxidized to mercuric mercury.

Absorption of inorganic mercury through the gastrointestinal tract varies with the particular mercuric salt involved; absorption decreases with decreasing solubility. Estimates of the percentage of inorganic mercury that is absorbed vary; as much as 20% may be absorbed. Inorganic mercury has a reduced capacity for penetrating the blood-brain or placental barriers. There is some evidence indicating that mercuric mercury in the body following oral exposures can be reduced to elemental mercury and excreted via exhaled air. Because of the relatively poor absorption of orally administered inorganic mercury, the majority of the ingested dose in humans is excreted through the feces.

Methylmercury is rapidly and extensively absorbed through the gastrointestinal tract. Absorption information following inhalation exposures is limited. This form of mercury is distributed throughout the body and easily penetrates the blood-brain and placental barriers in humans and animals. Methylmercury in the body is considered to be relatively stable and is only slowly demethylated to form mercuric mercury in rats. It is hypothesized that methylmercury metabolism may be related to a latent or silent period observed in epidemiological studies observed as a delay in the onset of specific adverse effects. Methylmercury has a relatively long biological half-life in humans; estimates range from 44 to 80 days. Excretion occurs via the feces, breast milk, and urine.

The most common biological samples analyzed for mercury are blood, urine and scalp hair. The methods most frequently used to determine the mercury levels in these sample types include atomic absorption spectrometry, neutron activation analysis, X-ray fluorescence and gas chromatography.

Two major epidemics of methylmercury poisoning through fish consumption have occurred. The best known of these two epidemics occurred among people and wildlife living near Minamata City on the shores of Minamata Bay, Kyushu, Japan. The source of methylmercury was a chemical factory that used mercury as a catalyst. A series of chemical analyses identified methylmercury in the factory waste sludge, which was drained into Minamata Bay. Once present in Minamata Bay, the methylmercury accumulated in the tissue of shellfish and fish that were subsequently consumed by wildlife and humans. Fish was a routine part of the diet in these populations. An average fish consumption was reported to be in excess of 300 g/day (reviewed by Harada et al., 1995); this is a far greater level of fish consumption than is typical for the general U.S. population. For the general U.S. population, the average fish consumption level is between 8 and 10 g/day (based on month-long data in NHANES III).

The first poisoning case occurred in 1956 in a six year old girl who came to a hospital complaining of symptoms characteristic of nervous system damage. Symptoms of Minamata disease in children and adults included the following:

- Impairment of the peripheral vision;
- disturbances in sensations ("pins and needles" feelings, numbness) usually in the hands and feet and sometimes around the mouth;
- incoordination of movements as in writing;
- impairment of speech;
- impairment of hearing;
- impairment of walking; and
- mental disturbances.

It frequently took several years before people were aware that they were developing the signs and symptoms of methylmercury poisoning.

Over the next 20 years the number of people known to be affected with what became known as Minamata disease increased to thousands. In time the disease was recognized to result from methylmercury poisoning, and fish were subsequently identified as the source of methylmercury. As is often the situation with epidemics, the first cases noted were severe. Deaths occurred among both adults and children. It also was recognized that the nervous system damage could occur to the fetus if the mother ate fish contaminated with high concentrations of methylmercury during pregnancy. The nervous system damage of severe methylmercury poisoning among infants was very similar to congenital cerebral palsy. In the fishing villages of this region the occurrence of congenital cerebral palsy due to methylmercury was very high compared to the incidence for Japan in general. At the height of the epidemic, mercury concentrations in fish were between 10 and 30 ppm wet weight. After the source of mercury contamination was identified, efforts were made to reduce the release of mercury into the bay. After 1969, average mercury concentrations in fish had fallen below 0.5 ppm.

In 1965, an additional methylmercury poisoning outbreak occurred in the area of Niigata, Japan. As in Minamata, multiple chemical plant sources of the chemical were considered. Scientific detective work identified the source again to be a chemical factory releasing methylmercury into the Agano River. The signs and symptoms of disease in Niigata were those of methylmercury poisoning and strongly similar to the disease in Minamata.

The abnormalities (or pathology) in the human brain that result from methylmercury poisoning are well described. There is an extremely high level of scientific certainty that methylmercury causes these changes. Similar pathology has been identified in other countries where methylmercury poisonings have occurred. Methylmercury contamination of other food products (including grains and pork products) has resulted in severe methylmercury poisoning with pathological changes in the nervous system and clinical disease virtually identical to Minamata disease.

Methylmercury poisoning occurred in Iraq following consumption of seed grain that had been treated with a fungicide containing methylmercury. The first outbreak occurred prior to 1960 and resulted in severe human poisonings. The second outbreak of methylmercury poisoning from grain consumption occurred in the early 1970s. Imported mercury-treated seed grains arrived after the planting season and were subsequently used as grain to make into flour that was baked into bread. Unlike the long-term exposures in Japan, the epidemic of methylmercury poisoning in Iraq was short in duration, but the magnitude of the exposure was high. Because many of the people exposed to methylmercury in this way lived in small villages in very rural areas (and some were nomads), the number of people exposed to

these mercury-contaminated seed grains is not known. The number of people admitted to the hospital with symptoms of poisoning has been estimated to be approximately 6,500, with 459 fatalities reported.

As in the Japanese poisoning epidemics, the signs and symptoms of disease were predominantly those of the nervous system: difficulty with peripheral vision or blindness, sensory disturbances, incoordination, impairment of walking, slurred speech and in some cases, death. Children were affected, as well as adults. Of great concern was the observation that infants, born of mothers who had consumed the methylmercury-contaminated grain (particularly during the second trimester of pregnancy) could show nervous system damage even though the mother was only slightly affected herself.

Some Limitations of the Assessment

In both the Iraqi and Japanese epidemics, the levels of methylmercury consumed were much higher than the levels currently reported in the U.S. food supply. For example, in the Japanese epidemic, mercury concentrations in fish were between 10 and 30 ppm. Average concentrations in freshwater fish in the U.S. are roughly 0.3 ppm. The most frequently consumed marine species have mercury concentrations less than 0.2 ppm. While there are no data to indicate that methylmercury absorption is affected by food type, it must be noted that one of the severe poisoning episodes was through a means not expected to be prevalent in the U.S.; that is, the consumption of contaminated grain.

Health endpoints other than neurotoxicity were evaluated by U.S. EPA using established risk assessment Guidelines. Data for other endpoints than developmental neurotoxicity were limited. Methylmercury has been shown to cause tumors in mice at high doses that produce severe non-cancer toxicity. Low-dose exposures to methylmercury are not likely to cause cancer in humans. Data on effects related to mutation formation (changes in DNA) indicate that methylmercury could increase frequencies of mutation in human eggs and sperm. These data were not sufficient, however, to permit estimation of the amount of methylmercury that would cause a measurable mutagenic effect in a human population.

How Much Methylmercury is Harmful to Humans?

Information on the amount of methylmercury exposure producing particular combinations of signs and symptoms in people has been analyzed to yield what are called quantitative dose-response assessments. Both the Japanese and Iraqi epidemics are important to understanding how methylmercury from food produces neurological disease in humans. In the epidemics in Minamata and Niigata, the exposures were long-term, and the tissues of fish and shellfish were the sources of methylmercury exposure. This establishes with highest scientific confidence that methylmercury in fish can produce human disease. A limitation to these data is that many patients were severely affected. The extent of methylmercury poisoning was so severe that finding subtle indications of disease is difficult. Subtle indicators of poisoning are important for estimating the level of exposure that will not cause adverse effects. The U.S. EPA calculates one such estimate, called a reference dose or RfD (see the box "What Is a Reference Dose?").

U.S. EPA has on two occasions published RfDs for methylmercury which have represented the Agency consensus for that time. These are discussed at length in Volume IV, and the uncertainties and limitations are described in Volume VI. At the time of the generation of the Mercury Study Report to Congress, it became apparent that considerable new data on the health effect of methylmercury in humans were emerging. Among these are large studies of fish or fish and marine mammal consuming populations in the Seychelles and Faroes Islands. Smaller scale studies are in progress which describe effects in populations around the U.S. Great Lakes. In addition, there are new evaluations, including novel statistical approaches and application of physiologically-based pharmacokinetic models to

published work. The U.S. EPA has been advised by scientific reviewers to employ this RfD for this analysis. Because of various limitations and uncertainties in the Iraqi data set, the U.S. EPA and other federal agencies intend to participate in an interagency review of all the human data on methylmercury, including the more comprehensive studies from the Seychelles and Faroe Islands. The purpose of this review is to reduce the level of uncertainty attending current estimates of the level of exposure to mercury associated with subtle neurological endpoints. After this process, the U.S. EPA will re-assess its RfD for methylmercury to determine if change is warranted.

The current U.S. EPA RfD for methylmercury was based on data on neurologic changes in 81 Iraqi children who had been exposed *in utero*: their mothers had eaten methylmercury-contaminated bread during pregnancy. The data were collected by interviewing the mothers of the children and by clinical examination by pediatric neurologists approximately 30 months after the poisoning episode. The incidence of several endpoints (including late walking, late talking, seizures or delayed mental development, and scores on clinical tests of nervous system function) were mathematically modeled by U.S. EPA to determine a mercury level in hair (measured in all the mothers in the study) which was associated with no adverse effects. These effects were delays in motor and language development defined by the following:

- Inability to walk two steps without support by two years of age;
- inability to respond to simple verbal communication by age 2 years among children with good hearing;
- scores on physical examination by a neurologist that assessed cranial nerve signs, speech, involuntary movements, limb tone, strength, deep tendon reflexes, plantar responses, coordination, dexterity, primitive reflexes, sensation, posture, and ability to sit, stand, walk, and run; and
- assessment of mental development or the presence of seizures based on interviews with the child's mother.

In calculating the mercury level in hair which was associated with no adverse effects, the U.S. EPA chose a benchmark dose approach based on modeling of all effects in children. The benchmark dose is the intake of methylmercury associated with the lower bound (that is the lower limit) on a 95 percent confidence interval of a dose producing a 10 percent prevalence of adverse effects. The 95 percent confidence interval indicates there is a 5 percent likelihood that the effect reported was due to chance alone. The effects used as end-point of adverse neurological effects included delayed walking, delayed talking, and abnormal neurological scores (see p. 3-38 for details). This lower bound was 11 ppm hair concentration for methylmercury. A dose-conversion equation was used to estimate a daily intake of 1.1 μg methylmercury/kg body weight/day that when ingested by a 60 kg individual will maintain a blood concentration of approximately 44 $\mu\text{g}/\text{L}$ of blood or a hair concentration of 11 μg mercury/gram hair (11 ppm). Mothers with hair concentrations below that associated with the RfD (1 μg Hg/g hair) are unlikely to experience adverse effects.

Data on the behavior of mercury in the human body were used to estimate the amount of mercury ingested per day at this no adverse effect level. Due to variability in the way individuals process methylmercury in the body and the lack of data on observed adult male and female reproductive effects, an uncertainty factor of 10 was used to derive the RfD from the benchmark dose. The RfD for methylmercury was determined to be 1×10^{-4} mg/kg-day; that is a person could consume 0.1 μg methylmercury for every kg of his/her body weight every day for a lifetime without anticipation of risk of

What Is A Reference Dose?

A reference dose or RfD is defined in the following way by U.S. EPA: an estimate (with uncertainty spanning perhaps an order of magnitude) of a daily exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime. RfDs are reviewed by Agency scientists for accuracy, appropriate use of risk assessment methodology, appropriate use of data and other scientific issues. When consensus has been reached by the workgroup, information on the RfD is made available to the public through a U.S. EPA database; namely, the Integrated Risk Information System (IRIS).

The RfD is based on the best available data that indicate a "critical effect"; this is generally the first indicator or most subtle indicator of an adverse effect in the species under study. In calculating RfDs U.S. EPA generally uses a no observed adverse effect level (NOAEL). This is found from either inspection or modeling of dose-response data on the critical effect. It is a means of estimating the threshold for effect in the reported study. The NOAEL is most useful when it is from a study in which a determination of the lowest observed adverse effect level (LOAEL) can also be done. The LOAEL is the lowest tested dose at which the critical effect was seen in the species under study.

In calculating the RfD the U.S. EPA divides the NOAEL or LOAEL by a series of uncertainty and modifying factors in order to extrapolate to the general human population. The uncertainty factors (which may be as much as 10 each) are for the following areas: extrapolation of data to sensitive human subpopulations; extrapolation from animal data to conclusions for humans; lack of chronic data; lack of certain other critical data; and use of a LOAEL in the absence of a NOAEL.

The RfD is used for risk assessment judgments dealing with evaluations of general systemic toxicity. It is intended to account for sensitive (but not hypersensitive) members of the human population; the rationale is that if exposure to the RfD is likely to be without appreciable risk for sensitive members of the population, then it is without appreciable risk for all members of the population. The RfD is generally applicable to men and women and to adults, to children and to the aged, unless data support the calculation of separate RfDs for these groups.

The RfD is a quantitative estimate of levels expected to be without effect even if exposure persists over a lifetime. It is not intended to be compared with isolated or one time exposures. Exceedance of the RfD does not mean that risk will be present. Acceptability of uncertain risks is a risk management decision. Risk management decisions may consider the RfD but will take into account exposures, other risk factors and non-risk factors as well. At the RfD or below, exposures are expected to be safe. The risk following exposures above the RfD is uncertain, but risk increases with increasing exposures.

adverse effect. The RfD is a daily ingestion level anticipated to be without adverse effect to persons, including sensitive subpopulations, over a lifetime. The RfD may be considered the midpoint in an estimated range of about an order of magnitude. This range reflects variability and uncertainty in the estimate. At the RfD or below, exposures are expected to be safe. The risk following exposures above the RfD is uncertain but risk increases with increasing exposure.

The RfD is a risk assessment tool, not a risk management decision. Judgments as to a "safe" dose and exposure represent decisions that involve risk management components.

Limitations and Uncertainties in the Assessment

The range of uncertainty in the RfD and the factors contributing to this range were evaluated in qualitative and quantitative uncertainty analyses. The uncertainty analyses indicated that paresthesia (numbness or tingling) in the hands and feet, and occasionally around the mouth, in adults is not the most reliable endpoint for dose–response assessment because it is subject to the patient's recognition of the effect. Paresthesia in adults is no longer the basis for U.S. EPA's methylmercury RfD as it was in the mid-1980s. There are, however, uncertainties remaining on the current RfD based on developmental effects from methylmercury in children exposed *in utero*. There are difficulties with reliability in recording and classifying events like late walking in children, especially as the data were collected approximately 30 months after the child's birth. It should be noted, however, that the endpoints used represented substantial developmental delays; for example, a child's inability to walk two steps without support at two years of age, inability to talk based on use of two or three meaningful words by 24 months, or presence of generalized convulsive seizures. There is uncertainty in the physiologic factors which were used in estimating the ingested mercury dose. There is also a degree of uncertainty introduced by the size of the study population (81 mother-child pairs).

The RfD is supported by investigations of laboratory animals under controlled exposures to methylmercury. Data from experimental animals (including primates with long-term exposures to methylmercury) show methylmercury-induced nervous system damage, particularly on the visual system, although the animals appeared clinically normal. The endpoints described in the animal literature are important and these have been induced by dosing protocols that are relevant to human exposures. In experiments using nonhuman primates, sensory (visual, somatosensory, auditory), cognitive (learning under concurrent schedules, recognition of faces), social play, and schedule-controlled operant behavior are all identified as having been adversely affected by methylmercury. The sensory, cognitive, and motor deficits appear reliably over a consistent range of doses in nonhuman primates exposed to methylmercury during development. Subtle, but important deficits, appear in several functional domains. These are identifiable signs of methylmercury effects when appropriate testing conditions are applied.

The RfD is supported by additional studies in children exposed *in utero*. These include investigations among Cree Indians in Canada and New Zealanders consuming large amounts of fish. In these studies the hair concentration of mercury is used to monitor mercury exposure over time. Conclusions by the investigators in their official reports cite developmental delays among the children born of mothers whose hair mercury concentrations during pregnancy were 6 to 18 µg/g, consistent with the benchmark dose of 11 µg/g.

Currently a number of research studies are underway that further address the question of what exposures to methylmercury in fish are associated with neurological disease. These studies include more subjects than did the Iraqi study, are prospective in design, and utilize endpoints that are anticipated to be more sensitive than the clinical signs and symptoms of methylmercury poisoning observed in Iraq. These studies of developmental effects of mercury exposure secondary to fish and shellfish consumption, rather than poisoning, are conducted in the Seychelles Islands in the Indian Ocean (sponsored, in part by the Department of Health and Human Services), the Faroe Islands in the North Atlantic Ocean (sponsored, in part, by the United States Environmental Protection Agency and by the United States Department of Health and Human Services), and in the United States; this last study is sponsored by the Agency for Toxic Substances and Disease Registry (ATSDR).

Data from both the Seychelle Islands cohort and the Faroe Islands cohort have been published during 1996 and 1997. These data should be useful in decreasing the uncertainty surrounding both the benchmark dose and the RfD, however, statistical analyses for purposes of risk assessment have been

recommended by the U.S. EPA's Science Advisory Board. In addition to these two major prospective investigations, additional studies evaluating the effect of methylmercury exposures from fish and shellfish in human subjects from other geographic areas are anticipated to be published in the peer-reviewed literature within the period 1997/1998. The U.S. FDA has determined that revisions of its action level for mercury concentrations of fish in interstate commerce should wait until the new studies have reduced the level of uncertainty. The availability of results from the above studies will likewise enable U.S. EPA to re-examine and adjust its RfD as needed.

Levels of Methylmercury Exposure Addressed by the U.S. Food and Drug Administration, World Health Organization and State Recommendations

The U.S. EPA RfD is a daily intake level and is a risk assessment tool; the use of the RfD is not limited to fish. The discussion that follows covers risk assessment and risk management activities concerning fish. These consider fish consumption patterns and risk management policy factors.

There are numerous local and state warnings in the U.S. to limit intake of fish because of chemical contamination. Warnings are issued because of a number of contaminants. Methylmercury is most often included as one of the contaminants that form the basis for the warning. Often these warnings are issued based on local conditions.

Recommended limits on methylmercury exposure have been expressed in these units: $\mu\text{g}/\text{kg}$ body weight/day; concentrations of mercury in tissues such as blood, hair, feathers, liver, kidney, brain, etc.; grams of fish per day; number of fish meals per time interval (e.g., per week). Reference values for mercury concentrations (expressed as total mercury) in biological materials commonly used to indicate human exposures to mercury were published by the WHO/IPCS (1990). The mean concentration of mercury in whole blood is approximately $8 \mu\text{g}/\text{L}$, in hair about $2 \mu\text{g}/\text{g}$, and in urine approximately $4 \mu\text{g}/\text{L}$. Wide variation occurs about these values (WHO/IPCS, 1990).

A number of different estimates exist for hair mercury levels that are associated with low risks of neurological endpoints such as paresthesia. These estimates are sensitive to variables such as the half-life of mercury in the body (time to eliminate half the dose of mercury). Half-life is usually estimated as an average of 70 days, with extremes of about 35 to just over 200 days reported for different individuals. The half-life of mercury in pregnant women has not been directly measured. The half-life of mercury in women during lactation is shorter, possibly due to excretion of mercury into milk produced during lactation.

Cross-comparison of World Health Organization (WHO) recommendations regarding risk associated with hair mercury concentrations is facilitated by data reported by the WHO on mercury concentrations in 559 samples of human head hair from 32 locations in 13 countries. The WHO report found that mercury concentrations in hair increased with increasing frequency of fish consumption (see Table 3-4).

**Table 3-4
WHO Data on Mercury in Hair**

Fish Consumption Frequency	Average Mercury Concentration in Hair (μg mercury per g of hair)
No unusual mercury exposure	2
Less than one fish meal per month	1.4 (range 0.1 to 6.2)
Fish meals twice a month	1.9 (range 0.2 to 9.2)
One fish meal a week	2.5 (range 0.2 to 16.2)
One fish meal each day	11.6 (range 3.6 to 24.0)

The World Health Organization's International Programme for Chemical Safety (WHO/IPCS) provided estimates of risk of neurological effects from methylmercury exposures for adults and fetuses. Adult effects occur at higher exposures to methylmercury than do fetal effects. WHO/IPCS concluded that the general population of adults (males and non-pregnant females) does not face a significant health risk from methylmercury when hair mercury concentrations are under 50 μg mercury/gram hair. In recent evaluations of the Niigata epidemic of Minamata disease, study authors reported lower thresholds with mean values in the range of 25 to approximately 50 μg mercury/gram hair.

Clinical observations in Iraq suggest that women during pregnancy are more sensitive to the effects of methylmercury because of risk of neurological damage to the fetus. The WHO/IPCS (1990) analyzed the Iraqi data and identified a 30 percent risk to the infant of abnormal neurological signs when maternal hair mercury concentrations were over 70 $\mu\text{g/g}$. Using an additional statistical analysis, WHO/IPCS estimated a 5 percent risk of neurological disorder in the infant when the maternal hair concentration was 10 to 20 μg mercury/gram of hair. The recommendations of WHO/IPCS are based on clinically observable neurological changes as the indicator of effect. U. S. EPA's benchmark dose is associated with a hair mercury concentration of 11 $\mu\text{g/g}$ hair and clinically observable endpoints in the child following in utero methylmercury exposures to the mother. The RfD is one-tenth the benchmark dose because U.S. EPA applied an uncertainty factor of 10. The U.S. EPA RfD is within an order of magnitude of the dose described by WHO.

In addition to their recommendations on hair mercury concentrations WHO/IPCS recommended that as a preventive measure, in a subpopulation that consumes large amounts of fish (for example, one serving or 100 grams per day), hair levels for women of child-bearing age should be monitored for methylmercury.

The WHO/IPCS estimated (1990) that a daily methylmercury intake of 0.48 μg mercury/kg body weight will not cause any adverse effects to adults and that a methylmercury intake of 3 to 7 $\mu\text{g/kg}$ body weight/day would result in a <5 percent increase in the incidence of paresthesia in adults. Risk to this extent would be associated with hair mercury concentration of approximately 50 to 125 μg mercury per gram hair. By comparison, the U.S. EPA's reference dose, or the amount of methylmercury any person (including children and pregnant women) can ingest every day without harm is 0.1 $\mu\text{g/kg}$ body weight per day. This was based on a benchmark dose equal to 11 ppm ($\mu\text{g/g}$) hair. Children are expected to have a higher exposure to methylmercury (on a per kg body weight basis) than do adults.

In 1969, in response to the poisonings in Minamata Bay and Niigata, Japan, the U.S. FDA proposed an administrative guideline of 0.5 ppm for mercury in fish and shellfish moving in interstate

commerce. This limit was converted to an action level in 1974 (Federal Register 39, 42738, December 6, 1974) and increased to 1.0 ppm in 1979 (Federal Register 44, 3990, January 19, 1979) in recognition that exposure to mercury was less than originally considered. In 1984, the 1.0 ppm action level was converted from a mercury standard to one based on methylmercury (Federal Register 49, November 19, 1984).

FDA's action level is based on consideration of the tolerable daily intake (TDI) for methylmercury, as well as information on seafood consumption and associated exposure to methylmercury. The TDI is the amount of methylmercury that can be consumed daily over a long period of time with a reasonable certainty of no harm to adults. The neurological endpoint evaluated was paresthesia (see WHO description above for more information). U.S. FDA (and WHO) established a TDI based on a weekly tolerance of 0.3 mg of total mercury per person, of which no more than 0.2 mg should be present as methylmercury. These amounts are equivalent to 5 and 3.3 µg, respectively, per kilogram of body weight. Using the values for methylmercury, this tolerable level would correspond to approximately 230 µg/week for a 70 kg person or 33 µg/person/day. The TDI was calculated from data developed in part by Swedish studies of Japanese individuals poisoned in the episode of Niigata which resulted from the consumption of contaminated fish and shellfish and the consideration of other studies of fish-eating populations.

Based on observations from the poisoning event later in Iraq, U.S. FDA has acknowledged that the fetus may be more sensitive than adults to the effects of mercury (Federal Register 44: 3990, January 19, 1979; Cordle and Tollefson, 1984, U.S. FDA Consumer, September, 1994). In recognition of these concerns, U.S. FDA has provided advice to pregnant women and women of child-bearing age to limit their consumption of fish known to have high levels of mercury (U.S. FDA Consumer, 1994). U.S. FDA believes, however, that given existing patterns of fish consumption, few women (less than 1%) eating such high mercury fish will experience slight reductions in the margin of safety. However, due to the uncertainties associated with the Iraqi study, U.S. FDA has chosen not to use the Iraqi study as a basis for revising its action level. Instead, the U.S. FDA has chosen to wait for findings of prospective studies of fish-eating populations in the Seychelles Islands and in the Faroes Islands.

Characterization of Risk to Human Populations

The characterization of risk to U.S. human populations focuses on exposure to methylmercury. Although methylmercury is found in other media and biota, it accumulates to the highest concentrations in the muscle tissue of fish, particularly fish at the top of the aquatic food chain. As a result, fish ingestion is the dominant exposure pathway. The dominance of this pathway reflects both bioaccumulation of methylmercury in the fish and the efficiency with which methylmercury passes through intestinal walls. The critical elements in estimation of methylmercury exposure from fish are these: the species of fish consumed; the concentration of methylmercury in the fish; the quantity consumed and the frequency of consumption.

There are three ways to assess the risk to populations from methylmercury exposure. The first way used in this analysis was based on predicted increases in methylmercury concentrations in fish due to anthropogenic emissions coupled with predicted exposure to human (and wildlife) populations. This type of analysis has the advantage of predicting the direct impact of anthropogenic emissions on fish concentrations. The second way risk was assessed was by using dietary surveys to identify the amount and type of fish consumed by populations in the U.S. The advantage of this methodology is that a total exposure from fish can be evaluated, even though the contamination may have come from sources other than anthropogenic emissions. The third way to determine whether members of the population are at risk was to consider hair mercury levels as methylmercury exposures for the general populations are reflected by these levels. This type of assessment would be one appropriate measurement of actual mercury

exposure because biological samples are utilized. These three methodologies and conclusions regarding the risk characterization are presented below.

Modeled Anthropogenic Emissions and Predicted Fish Methylmercury Levels

The key issue addressed in the risk characterization was the extent to which anthropogenic mercury emissions from U.S. sources increase mercury concentrations in freshwater and marine fish such that subsequent consumption of these fish would result in increased risk to the consumer.

As described in previous sections, the U.S. EPA used models to evaluate exposures that result from atmospheric mercury emissions from U.S. sources. Exposure to mercury from fish consumption depends on both the mercury concentration in the fish and the amounts of fish consumed. The modeling analysis predicted that some of the mercury emitted from local emission sources deposits on local watersheds and water bodies where a fraction of it is methylated and incorporated into the aquatic food chain. Since mercury emissions are also transported across great distances, the deposition of mercury from distant sources as well as estimates of existing background concentrations were also considered to contribute to mercury around a single source. As noted in the discussion of the exposure analysis above, the U.S. EPA concludes that there is a plausible link between anthropogenic emissions and increases in methylmercury concentrations in freshwater fish.

Local water bodies in proximity (e.g., within 2.5 km) to industrial and combustion sources that emit substantial amounts of divalent mercury from low stacks or at a slow rate appear to be more highly impacted by atmospheric mercury releases. For water bodies located in remote areas, the predicted concentrations in fish are influenced by the overall proximity to anthropogenic sources, increased soot and ozone concentrations and elevated rainfall.

The highest levels of methylmercury in fish (e.g., greater than 1 ppm) predicted by the exposure model were in the trophic level 4 fish; that is, those predator species at the top of the food web. These high predictions generally result from using relatively conservative assumptions. By comparison, measured values in the U.S. range from less than 0.1 ppm to 8.42 ppm; typical values for trophic level 3 fish are about 0.08 ppm and for predatory fish in trophic level 4 about 0.3 ppm.

Given these potential methylmercury concentrations, the issue becomes the fish consumption rate of populations eating fish from these waters. Consumption of fish from these waters was assumed for three types of human populations: an adult with a high fish consumption rate ("high-end consumer"), a child of a high-end consumer and a recreational angler. The consumption and body weights used in the analysis are shown below in Table 3-5.

**Table 3-5
Body Weights and Fish Consumption Values Used in Exposure Modeling**

Subpopulation	Assumed Body Weight (kg)	Assumed Local Fish Consumption Rate (g/day)
Adult High-End Consumer	70	60
Child High-End Consumer	17	20
Adult Recreational Angler	70	30

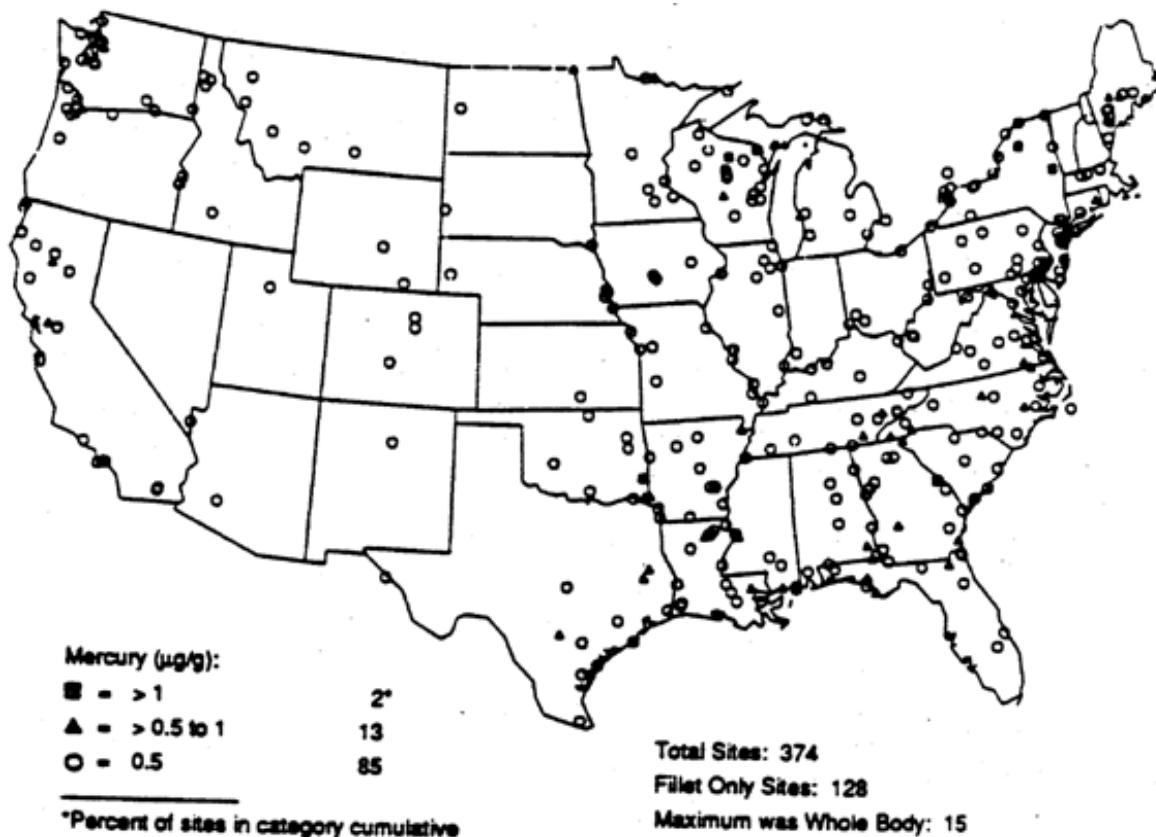
Results of the modeling analysis show that if humans consumed fish with mercury concentrations above 1 ppm at the above consumption rates, they would be ingesting mercury at levels approaching or exceeding the product of 10 times the U.S. EPA's RfD. Is it likely that the U.S. population would be consuming fish from inland waters with mercury levels this high? As noted above, the average concentration of mercury in freshwater fish in the U.S. is between 0.08 and 0.3 ppm depending on the size of the fish. For most consumers then, this scenario appears to be unlikely. However, it is known that there are locations in the U.S. where fish concentrations exceed do 1 ppm. The U.S. EPA has found mercury residues in fish at 92 percent of more than 370 surface water bodies tested in the U.S. Mercury levels above 1 ppm were found in at least one fish at 2 percent of the sites surveyed, and above 0.5 ppm in at least one fish at 15 percent of the sites. Figure 3-4 illustrates the geographic location of these sites.

The potential for a consumer to be at increased risk from fish consumption is modified by at least three important factors. First, many States have issued advisories regarding the consumption of certain species of fish from certain water bodies on account of mercury contamination. These advisories are meant to prevent the public from consuming fish with harmful levels of mercury in them. Thus, exposures to high concentrations are hopefully avoided. (It is known however, that not all anglers heed this advice.)

Second, most sport anglers fish from a variety of water bodies. Several studies indicate that many of these anglers may travel extended distances to fish; they may be traveling to places where fish have higher or lower mercury concentrations than those nearby. These individuals who consume fish from a variety of locations decrease their chance of exposure to methylmercury at toxicologically significant doses because the extent of mercury contamination can differ significantly between water bodies. Although some areas of the U.S. are known to have fish contaminated with levels above 1 ppm, the national average for freshwater fish is 0.3 ppm based on data from Bahnick et al., (1985).

Third, some members of the population, even though they consume large quantities of fish, are likely to obtain their fish from both local water bodies and from commercial sources. By eating a variety of fish in the diet, including fish obtained commercially, it is likely that fish with a range of mercury levels are being consumed. A consumer may be purchasing fish with lower mercury levels than those locally caught. Thus, overall exposure would be reduced. For example, the top ten seafood species all have methylmercury levels less than 0.2 ppm. These species are listed in Table 3-6. Note however, that there are some saltwater species, notably shark and swordfish, that do have elevated levels of mercury. These are not frequently consumed species, but their mercury levels are sufficiently high to have potential for increased risk if consumed regularly. Consequently, the FDA advises pregnant women, and women of childbearing age intending to become pregnant, to limit their consumption of shark and swordfish to no more than once a month.

Figure 3-4
Distribution of Mercury Concentrations in U.S. EPA-Sampled
Fish Tissue Throughout the U.S.



The FDA advises persons other than pregnant women and women of child-bearing age to limit their consumption of fish species with methylmercury levels around 1 ppm to about 7 ounces per week (about 1 serving). For fish with levels averaging 0.5 ppm, regular consumption should be limited to about 14 ounces per week (about two servings). Consumption advice is unnecessary for the top 10 seafood species listed in Table 3-6 as mercury levels are 0.2 ppm or less and few people eat more than the suggested weekly limit of fish (2.2 pounds) for this level of contamination. FDA made this latter statement for all segments of the population, including women who might become pregnant.

Human Exposure to Methylmercury Based on Dietary Surveys

Estimates of the number of individuals who exceed various recommendations on exposures to mercury are characterized by both uncertainty and variability. In its review of U.S. EPA's earlier draft of this Report, the Science Advisory Board noted that the high end of the distribution of methylmercury exposures is very uncertain with respect to exposures, total number of people (and percent of the population) who

Table 3-6
Mercury Concentrations in the Top Ten Types of Fish/Shellfish Consumed by U.S. Residents

Fish^a	Mercury Concentration (ppm, wet weight)^b	Comments
Tuna	0.206	The mercury content for tuna is the average of the mean concentrations measured in 3 types of tuna: albacore tuna (0.264 ppm), skipjack tuna (0.136 ppm) and yellowfin tuna (0.218 ppm). The U.S. FDA measured the methylmercury concentration in 220 samples of canned tuna in 1991; the average amount of methylmercury measured in these samples was 0.17 ppm and the measured range was <0.1 - 0.75 ppm (Yess, 1993).
Shrimp	0.047	The mercury content for shrimp is the average of the mean concentrations measured in seven types of shrimp: royal red shrimp (0.074 ppm), white shrimp (0.054 ppm), brown shrimp (0.048 ppm), ocean shrimp (0.053 ppm), pink shrimp (0.031 ppm), pink northern shrimp (0.024 ppm) and Alaska (sidestripe) shrimp (0.042 ppm).
Pollack	0.15	The Pesticide and Chemical Contaminant Data Base for U.S. FDA (1991/1992) reports the methylmercury concentration in pollack in commerce as 0.04 ppm.
Salmon	0.035	The mercury content for salmon is the average of the mean concentrations measured in five types of Salmon: pink (0.019 µg/g), chum (0.030 ppm), coho (0.038 ppm), sockeye (0.027 ppm), and chinook (0.063 ppm).
Cod	0.121	The mercury content for cod is the average of the mean concentrations in Atlantic Cod (0.114 ppm) and the Pacific Cod (0.127 ppm).
Catfish	0.088 0.16	The sources of mercury content in catfish are Bahnick et al., 1994 and Lowe et al., 1985. Both data sets were collected from U.S. freshwater sources. The Bahnick data (mean = 0.088) include channel, largemouth, rock, striped and white catfish, and the Lowe data (mean = 0.16) include channel and flathead catfish. It should be noted that neither survey included farm-raised catfish, which is the type of catfish predominantly consumed in the U.S. The mercury content of farm-raised catfish may be significantly different than freshwater sources. The Pesticide and Chemical Contaminant Data Base for U.S. FDA (1991/1992) reports the methylmercury concentration in farm-raised catfish as 0.02 ppm.
Clam	0.023	The mercury content for clam is the average of the mean concentrations measured in four types of clam: hard (or quahog) clam (0.034 ppm), Pacific littleneck clam (0 ppm), soft clam (0.027 ppm), and geoduck clam (0.032 ppm).

Table 3-6 (continued)
Mercury Concentrations in the Top Ten Types of Fish/Shellfish Consumed by U.S. Residents

Fish ^a	Mercury Concentration (ppm, wet weight) ^b	Comments
Flatfish (Flounder)	0.092	The mercury content for flounder is the average of the mean concentrations measured in nine types of flounder: Gulf (0.147 ppm), summer (0.127 ppm), southern (0.078 ppm), four-spot (0.090 ppm), windowpane (0.151 ppm), arrowtooth (0.020 ppm), witch (0.083 ppm), yellowtail (0.067 ppm), and winter (0.066 ppm).
Crab	0.117	The mercury content for crab is the average of the mean concentrations measured in five types of crab: blue crab (0.140 ppm), dungeness crab (0.183 ppm), king crab (0.070 ppm), tanner crab (<i>C. opilio</i>) (0.088 ppm), and tanner crab (<i>C. bairdi</i>) (0.102 ppm).
Scallop	0.042	The mercury content for scallop is the average of the mean concentrations measured in four types of scallop: sea (smooth) scallop (0.101 ppm), Atlantic Bay scallop (0.038 ppm), calico scallop (0.026 ppm), and pink scallop (0.004 ppm).

^a List of fish types from U.S. FDA (1995).

^b Mercury concentrations sources are described in the comments, refer to Volume III for complete citations.

may be experiencing exposures high enough to cause adverse health effects, and the actual subgroups who are highly exposed. Consequently, the total population at risk is not, and cannot be fully characterized at this time.

Because of the uncertainties intrinsic to describing fully the high-end of the distribution, where multiple estimates of the size of the highly exposed population are available, a range of values has been presented. Predicted high exposures to methylmercury are caused by one of two factors or their combination: 1) consumption of types of fish which exhibit elevated methylmercury concentrations in their tissues; and or 2) high consumption rates of methylmercury contaminated fish.

The discussion of the modeling analysis above focused on potential risk to human populations due to consumption of fish having relatively high concentrations of mercury. A limitation of the modeling analysis is that the size of the population potentially at increased risk cannot be estimated because hypothetical water body locations and exposure scenarios are employed. The analysis of mercury exposure using dietary surveys described below is aimed at identifying populations that eat much greater amounts of fish than the average consumer. Their potential for increased risk is not necessarily due to elevated concentrations in fish, it is more a function of the amount of fish consumed on a regular, usually daily, basis. The analysis of the at-risk population eating above average amounts of fish focuses on that part of the population which consumes on average 100 grams or more of fish or shellfish per day (approximately 3.5 ounces). The basis for this focus on persons eating 100 grams or more is a recommendation made by the World Health Organization's International Programme for Chemical Safety (WHO) that populations consuming large amounts of fish and shellfish require special consideration. The 100 gram per day recommendation by the WHO can be used as a screening analysis to identify populations potentially at increased risk; particularly risk among pregnant women. The

significance of the risk is, as mentioned above, is also a function of the methylmercury concentrations of the fish consumed. Figure 3-5 illustrates the distribution of fish consumption rates of various populations. As shown in Figure 3-5, the general U.S. population consumes, on average, far less fish than subsistence fishers and some Native American tribes which have been studied. Figure 3-5 also illustrates that some members of the U.S. population do consume fish in large amounts on a daily basis.

The second way dietary surveys were used in this analysis was to calculate methylmercury exposure over a month-long period. This can be achieved by combining the frequency distribution of month-long patterns of fish/shellfish consumption with dietary data indicating the species of fish consumed, average values for mercury concentrations in the species of fish consumed, the portion size consumed, and the individuals' body weights.

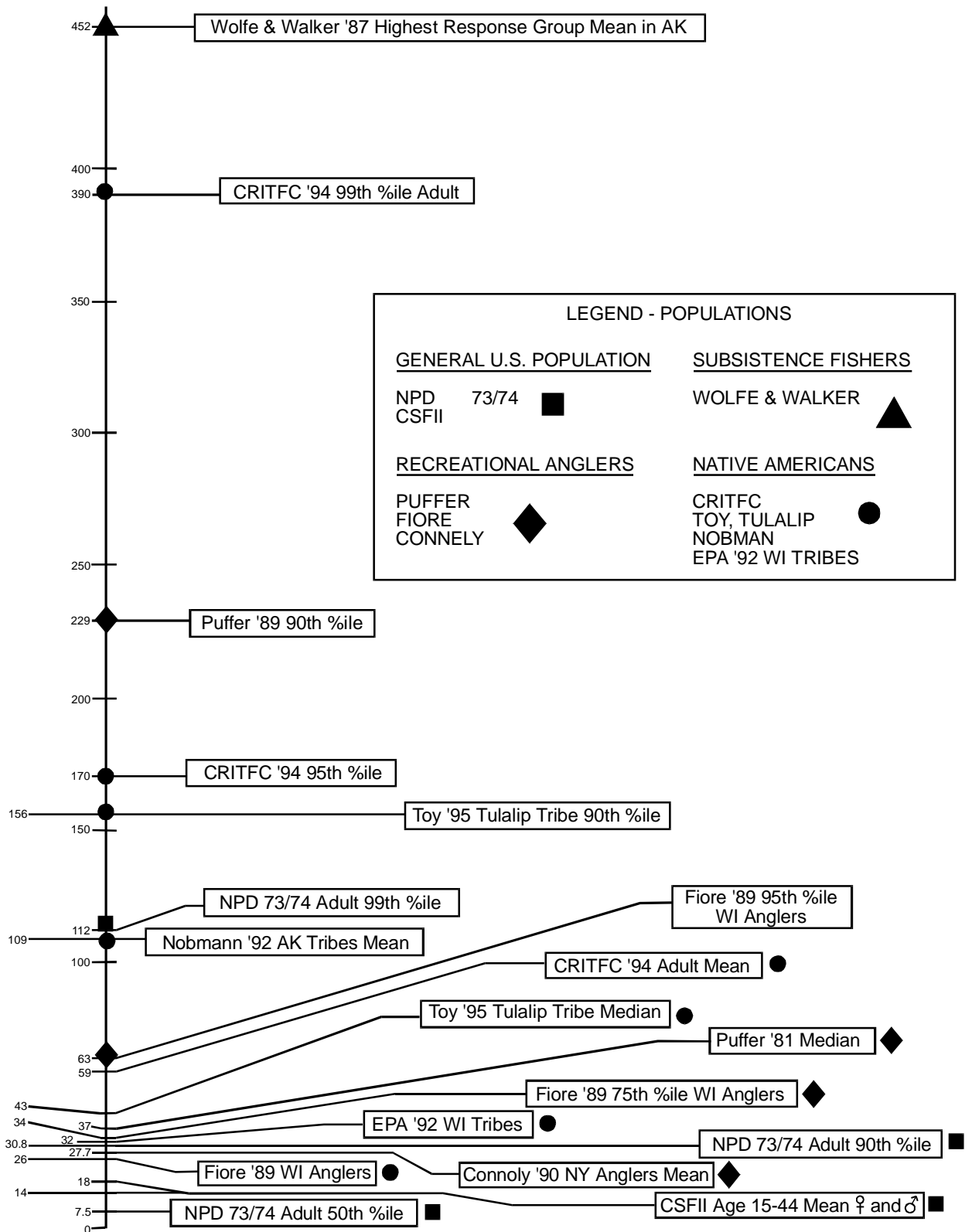
The U.S. EPA used two types of dietary surveys to identify these populations. Dietary surveys can be classified into longitudinal or cross-sectional surveys. Cross-sectional data are used to give a "snap shot" in time and are typically used to provide information on the distribution of intakes for groups within the population of interest. Cross-sectional data typically are for 24-hour or 3-day sampling periods and may rely on recall of foods consumed following questioning by a trained interviewer, or may rely on written records of foods consumed. The cross-sectional surveys used in this Report were the Continuing Surveys of Food Consumption by Individuals for the periods 1989 to 1991, 1994 and 1995 and the third National Health and Nutrition Examination Survey (NHANES III) conducted between 1988 and 1994.

General U.S. Population. NHANES III obtained data on the self-reported month-long frequency of consumption of fish and shellfish by respondents in this survey. Of the adults surveyed 86 percent reported they ate fish or shellfish at least once during the previous month. Major subgroups in the general population indicated they consumed fish and shellfish more frequently than did the overall population. Among persons who designated themselves as "White/NonHispanic," 1.9 percent consumed fish/shellfish 6.4 times or more a week. Within the subpopulation of persons who categorized themselves as "Black/NonHispanic," 3.3 percent consumed fish/shellfish 6 times or more per week. Among persons who categorized themselves as "Other" (typically individuals of Asian/Pacific Islander ethnicity, Native Americans, Alaskan Natives, and persons of Caribbean ethnicity), 8.9 percent indicated they consumed fish/shellfish 6 times or more a week.

Subpopulations of Concern. Three groups are potentially at increased risk from methylmercury: pregnant women, women of child-bearing age (i.e., between the ages of 15 and 44) and children ages 14 and younger. Pregnant women are of concern because of the adverse effects of methylmercury on the fetal nervous system. Women of child-bearing age rather than only pregnant women are of concern for two reasons. The first is that methylmercury persists in tissues. Measured half-lives for methylmercury in adults range from about 1 month to 9 months, although half-lives of just over 2 months are usually observed. Thus, dietary intakes just prior to pregnancy are of concern rather than only methylmercury intakes during pregnancy. The second reason is that women usually do not know they are pregnant until the pregnancy is past many of the critical stages of fetal development.

Children may be at a higher risk of methylmercury exposure than are adults because they appear to have higher exposures on a per kilogram body weight basis, and they may be inherently more sensitive than adults given the developmental state of the nervous system. In the methylmercury poisoning epidemics in Japan and Iraq, children were affected, as well as adults. These effects were not seen only in children exposed to methylmercury *in utero*, but included children exposed through ingesting methylmercury from food. Whether or not children differ from adults in sensitivity to methylmercury neurotoxicity is not known.

Figure 3 -5
Distribution of Fish Consumption Rates of Various Populations



Within the subpopulation of women of child-bearing age, it is useful to estimate the number of women whose dietary patterns include eating fish and shellfish in amount of 100 grams/day or more. When the distribution of fish and shellfish intakes among the overall population of women in the United States is considered, it appears that approximately 5% of women eat fish and shellfish in amount of 100 grams/day or more, *on any single day*. This type of data provide a distribution across the entire population of women in the United States. However, because mercury is a toxic element that accumulates in the body over time, it is relevant to know what percent of women consume fish at the 100 grams or more level consistently. Using the NHANES III dietary data (including the fish consumption frequency data), it was estimated that 3% of women consistently consume 100 grams of fish/shellfish per day or more.

An additional source of information on typically long-term or longitudinal estimates of intake of mercury from fish and shellfish is a longitudinal survey, the National Purchase Diary, Inc conducted in 1973. In this survey the 99th percentile of fish and shellfish consumption among adults was 112 grams per day.

Based on these three different approaches to estimating the amount of fish and shellfish consumed on a month-long basis by adult women of childbearing age, it is estimated that between 1 and 3 percent of women consistently consume 100 grams or more of fish and shellfish per day. Because occasional ingestion of greater quantities of fish and shellfish may result in very short-term higher exposures, the 5% of women who report consuming 100 grams of fish and shellfish per day (the per capita data) may also be considered for purpose of assessing risk of exposure to mercury from diet.

Census statistics (United States 1990) indicate that within the 48 contiguous states the estimated number of women of childbearing age (that is assumed to be 15 through 44 years) was approximately 58,222,000. It was estimated that in a given year 9.5 percent of women in this age group are pregnant. If the consumption of 100 grams of fish/shellfish per day or more is used as a screen for concern for mercury exposures, it is estimated that between approximately 52,000 and 166,000 pregnant women consume fish at these levels (based on 1% of the population from the NPD, Inc, survey and the 3% projection for month-long national estimates of consumption based on NHANES III (1988-1994)).

Estimates of Methylmercury Exposure Based on Dietary Surveys. Exposure to methylmercury from contaminated fish results in an incremental increase in mercury exposure. Methylmercury exposure rates on a per body weight basis among fish-consuming children are predicted to be higher than for fish-consuming adults. Data obtained in the NHANES III permits calculation of national estimates of month-long exposures to mercury from eating fish and shellfish. This can be achieved by combining the frequency distribution of month-long patterns of fish/shellfish consumption with dietary data indicating the species of fish consumed, mean values for mercury concentrations in the species of fish consumed, the portion size consumed, and the individuals' body weights.

As is the situation with adults, it is uncertain how often children consume fish and shellfish according to the patterns that are shown by the 24-hour recall data. In the NHANES III, there are no data for children which specify how frequently children eat fish/shellfish, although there are such data for adults. Consequently, a simplifying assumption was made that children of a particular ethnic/racial group ate fish as often as the adults of that particular ethnic/racial group. Unlike the frequency data, however, the smaller portion size of fish/shellfish and the different species of fish/shellfish selected by children were described with the 24-hour recall data specific for children in NHANES III. In the U.S. EPA's analysis only the data on frequency of fish consumption was extrapolated from adult data.

Using this approach, month-long estimates of mercury exposure were calculated. The results indicate that 7 percent of women ages 15 through 44 years of age exceed the RfD (0.1 µg/kg-bw/day). For 1 percent of women ages 15 through 44 years of age, the month-long estimate of mercury exposure

was 0.37 µg/kg-bw/day. Some children were also estimated to have exposures of 0.3 ug/kg-bw/day and higher. As noted above, exposures at or below the RfD are expected to be safe. The risk following exposures above the RfD are uncertain, but risk increases as exposure to methylmercury increases.

It is important to note that the above estimates of mercury exposure from fish and shellfish are based on average concentrations of mercury in fish and shellfish usually selected by people. The most commonly consumed fish/shellfish were tuna, shrimp, and Alaskan pollock. The typical mixture of fish and shellfish species consumed is associated with an average concentration of mercury in the range of 0.1 to 0.15 parts per million. This average mercury concentration for the mixture of fish and shellfish species usually eaten is a major factor in determining national estimates of mercury exposure. These concentrations are comparatively lower than found in a number of species of fish.

Consumption of fish with mercury levels higher than average may pose a significant source of methylmercury exposure to consumers of such fish. The magnitude of methylmercury exposure varies with local consumption rates and methylmercury concentrations in the fish. The average mercury concentrations measured throughout the inland waters of the U.S. provide an indication of the amounts of mercury in various freshwater fish. These are provided in Table 2-1.

The exposure analysis described above indicates that some of these methylmercury concentrations in freshwater fish species may be elevated as a result of mercury emissions from anthropogenic sources. As a result, exposures may be elevated as a result of mercury emissions from anthropogenic sources. Because people may select fish from limited geographic regions where fish mercury concentrations are lower or higher than those present in the general diet, they may experience quite different mercury exposures than does the general population as described by the national estimates from dietary surveys (e.g., national estimates of month-long consumption projected from NHANES III). Exposures may be elevated among some members of subpopulations of concern; these are evidenced by blood mercury measurements in excess of 10 micrograms per liter of whole blood that have been reported among multiple freshwater fish-consuming subpopulations.

Hair Mercury Measurements

Actual measurements of hair mercury levels would be an additional to assess mercury exposure and risk because mercury exposure is reflected by hair mercury levels. Because fish are the primary exposure pathway for methylmercury there is a broad-based scientific literature describing increases in hair mercury concentrations with increases in fish consumption. Maternal hair mercury concentrations predict mercury concentrations in fetal brain, fetal blood, umbilical cord blood and newborn hair.

The WHO has concluded that the general population of adults (males and non-pregnant females) does not face a significant health risk from methylmercury when hair mercury concentrations are under 50 µg mercury/gram hair. However, in recent evaluations of the Niigata epidemic of Minamata disease, study authors reported lower thresholds with mean values in the range of 25 to approximately 50 µg mercury/gram hair.

In addition, clinical observations in Iraq suggest that women during pregnancy are more sensitive to the effects of methylmercury with fetuses at particularly increased risk. The WHO analyzed the Iraqi data and identified a 30 percent risk to the infant of abnormal neurological signs when maternal hair mercury concentrations were over 70 µg/g. Using an additional statistical analysis, WHO estimated a 5 percent risk of neurological disorder in the infant when the maternal hair concentration was 10 to 20 µg mercury/gram of hair.

Although data on hair mercury concentrations from a sample representative of the United States population with adequate documentation of quality assurance/quality control do not exist, data from

individual studies conducted within the United States are available and are discussed in the Volume on exposure (Volume IV) and in the risk characterization Volume (Volume VII). These surveys were conducted in widely diverse geographic areas within the United States. The mean hair mercury concentrations identified for subjects in these studies are typically under 1 $\mu\text{g/g}$ or 1 ppm. There are a number of uncertainties surrounding this value which are discussed in the risk characterization Volume. The maximum values reported in these individual surveys conducted in widely diverse geographic areas of the United States range from 2.1 to 15.6 ppm.

Hair mercury concentrations of 1 ppm or less are associated with dietary intakes of mercury of an estimated 0.1 $\mu\text{g/kgbw/day}$ which is also the RfD. These mercury concentrations correspond to hair mercury concentrations associated with fish consumption at the level of less than one meal per month to one meal per week. Based on the higher hair mercury concentrations reported in additional studies of subpopulations expected to have higher than usual consumption of fish and shellfish, dietary intake of mercury considerably in excess of the RfD also occurs among some members of the United States population.

Until appropriate survey data for the general United States population exist, the overall distribution of hair mercury concentrations for the United States remains unclear. For adequate prediction of methylmercury exposure for the general United States population, the data should be obtained from subjects who are chosen based on a sampling strategy that can be extrapolated to the United States population, and must include appropriate quality assurance/quality control procedures.

Summary of the Risk Characterization

In summary, conclusions that can be drawn from the risk characterization are these.

- There is evidence from measurement data and modeling analyses that past and present emissions and releases of mercury from industrial sources can be plausibly linked with incremental increases in environmental mercury concentrations, including fish methylmercury levels in surface waters in the U.S.
- One U.S. EPA study found mercury levels above 1 ppm in at least one fish at 2 percent of the sites surveyed, and above 0.5 ppm in at least one fish at 15 percent of the sites. U.S. emissions contribute to local, regional and global atmospheric mercury and the resulting deposition to the oceans and land. These emissions ultimately contribute to total mercury loads in fish, since elemental mercury in the environment is neither created nor destroyed. Because mercury methylation and subsequent uptake in fish is complex and not well understood, it cannot be assumed that a change in total mercury emissions will be linearly related to any resulting change in methylmercury in fish, even taking into account the role of natural and old anthropogenic sources.
- The typical U.S. consumer eating fish from restaurants and grocery stores is not in danger of consuming harmful levels of mercury from fish and is not being advised to limit fish consumption. The levels of mercury found in the most frequently consumed commercial fish are generally low, especially compared to levels that might be expected in some non-commercial fish from fresh water bodies that have been affected by mercury emissions.
- While most U.S. consumers need not be concerned about their exposure to mercury, some exposures may be of concern. Those who regularly and frequently consume large amounts of fish -- either marine species that typically have much higher levels of

mercury than the rest of seafood or freshwater fish that have been affected by mercury pollution -- are the most highly exposed. Since the developing fetus may be more sensitive to the effects of mercury in fish, women of child-bearing age are as the population of greatest interest.

- In this Report, an analysis of dietary surveys led the U.S. EPA to conclude that between 1 and 3 percent of the women of child-bearing age (i.e., between the ages of 15 and 44) eat sufficient amounts of fish to be at risk from mercury exposure depending on the mercury concentrations in the fish. In addition, some Native Americans or subsistence fishers do consume fish in these large quantities for cultural or economic reasons. These consumers should be aware of the FDA and State fish advisories that suggest limiting the consumption of contaminated fish.

Limitations of the Risk Characterization

The primary purpose of the Mercury Study Report to Congress was to assess the impact of U.S. anthropogenic emissions on mercury exposure to humans and wildlife. The size of some populations of concern have been estimated; namely women of child-bearing age and children who eat fish. In the general population, people typically obtain their fish from many sources. The question on whether or not the impact of mercury from anthropogenic ambient emissions can be proportioned to the overall impact of methylmercury on wildlife is a much more difficult issue.

As with environmental monitoring data, information on body burden of mercury in populations of concern (blood and/or hair mercury concentrations) are not available for the general U.S. population. Data on higher-risk groups are currently too limited to discern a pattern more predictive of methylmercury exposure than information on quantities of fish consumed. The selenium content of certain foods has been suggestive as a basis for modifying estimates of the quantities of methylmercury that produce adverse effects, although there is no consistent evidence that selenium is protective against the neurotoxicity of methylmercury. Experimental investigations under controlled conditions indicated that feline species developed neurotoxicity from methylmercury as severely and as rapidly if the methylmercury was present naturally in fish or added as chemically pure methylmercury to the animals' diet. Currently, data on this mercury/selenium association form an inadequate basis to modify quantitative estimates of human response to a particular exposure to mercury.

Available data for human health risk assessment have limitations as described in the Report and in this summary. Studies of human fish-consuming populations in the Seychelles and Faroes Islands address some of these limitations. Additional studies on U.S. populations who consume fish from the Great Lakes are in progress, as well as, additional studies currently in review and expected to be published during the period 1997/1998. Public health agencies of the U.S. government and the U.S. EPA will evaluate these new data when they are available.

The benchmark dose methodology used in estimating the RfD required that data be clustered into dose groups. Most data on neurologically based developmental endpoints are continuous; that is, not assigned to dose groups. For example, scoring on scales of IQ involves points rather than a "yes/no" type of categorization. Measurements on the degree of constriction of the visual field involve a scaling rather than a "constricted/unconstricted" type of variable. Although arbitrary scales can be constructed, these groupings have generally not been done in current systems. Use of alternative dose groupings (as described in Volume IV) had no significant effect on calculated benchmark doses. An additional difficulty occurs in estimation of benchmark dose for multiple endpoints that have been measured. Further research on appropriate methods for mathematical modeling is needed. For some situations such information is known, but for methylmercury exposure and multiple endpoints assessing the same system

(i.e., developmentally sensitive neurological, neuromotor and neuropsychological effects) the time-course/dose-response of such changes have not been clearly established. Development of the mathematical models needs to be accompanied by understanding the physiological/pathological processes of methylmercury intoxication.

How Much Methylmercury Exposure is Harmful to Wildlife and What Are the Effects?

Massive poisonings of birds and wildlife from methylmercury-treated seed grains were identified during the decades preceding the 1970s. These findings resulted in substantial limitation on use of methylmercury-treated seed grains. However, methylmercury contamination of the aquatic foodchain from many sources continues to adversely affect wildlife and domestic mammals and wild birds. In Minamata, Japan from about 1950-1952 (prior to recognition of human poisonings) severe difficulties with flying and other grossly abnormal behavior was observed among birds. Signs of neurological disease including convulsions, fits, highly erratic movements (mad running, sudden jumping, bumping into objects) were observed among domestic animals, especially cats that consumed seafood.

Generally the place of wildlife in the aquatic foodchain of the ecosystem and their feeding habits determine the degree to which the species is exposed to methylmercury. Fish-eating (piscivorous) animals and those which prey on other fish-eaters accumulate more mercury than if they consumed food from terrestrial food chains. In a study of fur-bearing animals in Wisconsin, the species with the highest tissue levels of mercury were otter and mink, which are top mammalian predators in the aquatic food chain. Top avian predators of aquatic food chains include raptors such as the osprey and bald eagle. Smaller birds feeding at lower levels in the aquatic food chains also may be exposed to substantial amounts of mercury because of their high food consumption rate (consumption/day/gram of body weight) relative to larger birds.

Laboratory studies under controlled conditions can be used to assess the effects of methylmercury from fish on mink, otter and several avian species. Effects can occur at a dose of 0.25 µg/g bw/day or 1.1 µg/g methylmercury in diet. Death may occur in species at 0.1-0.5 µg/g body weight/day or 1.0-5.0 µg/g in the diet. Smaller animals (for example, minks, monkeys) are generally more susceptible to mercury poisoning than are larger animals (for example, mule deer, harp seals). Smaller mammals eat more per unit body weight than larger mammals. Thus, smaller mammals may be exposed to larger amounts of methylmercury on a body weight basis.

Whole body residues of mercury in acutely poisoned birds usually exceed 20 µg/g fresh weight. Although sublethal effects include a number of different organ systems, reproductive effects are the primary concern. These occur at concentrations far lower than those that cause overt toxicity.

The broad ecosystem effects of mercury are not completely understood. No applicable studies of the effects of mercury on intact ecosystems were found. Consequently, characterization of risk for non-human species did not attempt to quantify effects of mercury on ecosystems, communities, or species diversity. The characterization focused on quantities of mercury that adversely affects the health of sensitive subpopulations of wildlife species and on the co-location of these populations with areas of elevated mercury exposure secondary to ambient, anthropogenic emissions of methylmercury. To this end wildlife criteria (WC) were calculated for four piscivorous birds and two mammals (see Table 3-8). The WC is a methylmercury level in water which is expected to be without harm for the species. The WC considers the bioaccumulation of methylmercury in the large and small fish eaten by the mammals or birds. A bioaccumulation factor (BAF) was used in the WC calculation; the BAF was based on data on methylmercury in fish and the water from which they were taken. A review of literature from the last several years suggests that there is now sufficient information available to estimate BAFs for

methylmercury. Previously, it was thought that much of the variation around BAFs estimated on a total mercury basis could be attributed to differences among water bodies in the proportion of total mercury existing as the methylated form. The goal of the present analysis was to calculate a WC for the bioaccumulating form of mercury, thereby yielding an estimate with the lowest possible variation around the mean. The effects data for mammals were from a short-term study of neurotoxicity in mink. The data for fish-eating birds were from a three-generation study in mallard ducks.

The evaluation of data and calculation of WC in this Report was done in accordance with the methods and assessments published in the Final Water Quality Guidance for the Great Lakes System: Final Rule. Availability of additional data led to differences in calculated values of the WC in this Report and those published in the final rule. Differences were the result of several factors. First, this Report uses more recent data to derive BAFs. The Supplementary Information Document to the final Water Quality Guidance for the Great Lakes System noted that a preliminary draft of the Mercury Report to Congress was available but was not used because it had not been completed at the time the final guidance was published. Second, the Guidance appropriately used some region-specific assumptions that were not used in this nationwide assessment (e.g., consumption of herring gulls by eagles). Third, different endpoints were used. In the Guidance, a risk-management decision was made to base the WC on endpoints that compare direct effects on growth, reproduction, or development. In this Report, more sensitive endpoints were considered with the goal of assessing a greater range of toxic effects. Finally, different uncertainty factors were employed in the two assessments. In general, uncertainty factors used in the GLWQI are more conservative than those used in this Report.

**Table 3-7
Wildlife Criteria for Methylmercury**

Organism	Wildlife Criterion (pg/L)
Mink	57
River otter	42
Kingfisher	33
Loon	82
Osprey	82
Bald eagle	100

Derivation of a WC to protect the Florida panther is complicated by the possibility that prey items (e.g., raccoon) accumulate mercury to an even greater extent than the fish represented by trophic level 4. Other prey (e.g., deer) probably contain relatively lower levels of mercury. Calculation of a WC protective of the panther, therefore, requires collection of additional information on the diet of this species and mercury residues in that diet. Existing data are insufficient to support such an analysis. A chronic NOAEL for domestic cats was reported to be 20 $\mu\text{g}/\text{kg}/\text{d}$. This is close to that of 5.5 $\mu\text{g}/\text{kg}/\text{d}$ estimated for mink (that is, the subchronic NOAEL of 55 $\mu\text{g}/\text{kg}/\text{d}$ divided by a UF_s of 10). Cats (and presumably larger felines) do not, therefore, appear to be uniquely sensitive or insensitive to the toxic effects of mercury.

Methylmercury (as described in Volumes V and VI of this Report) has deleterious effects on the chordate nervous system. The human health endpoint of concern is developmental neurotoxicity. The

health endpoints of concern for the avian wildlife species are reproductive and behavioral deficits and for the mammalian quadrupeds are neurological effects. Assuming that the effects are of similar concern for the well-being of individuals within a species, the NOAELs, LOAELs and the human and wildlife WCs for these health endpoints can then be compared across species.

The human benchmark dose of 11 ppm mercury in hair was considered operationally equivalent to a NOAEL in the derivation of the methylmercury RfD. A LOAEL of 52.5 ppm mercury in hair was estimated for purposes of this risk characterization from inspection of data in Table 5-4 of Volume VI. The NOAEL of 11 ppm mercury in hair and the LOAEL of 52.5 ppm mercury in hair correspond to ingestion levels of 1 $\mu\text{g}/\text{kg}\text{-day}$ and 5.3 $\mu\text{g}/\text{kg}\text{-day}$, respectively; these dose conversions were made by applying the methods for converting hair mercury concentrations to ingestion levels used in the derivation of the RfD in Volume IV of this Report.

The avian RfD was based on the data from a series of studies by Heinz and collaborators on mallard ducks. A NOAEL could not be identified. The estimated LOAEL, based on reproductive and behavioral effects, was 78 $\mu\text{g}/\text{kg bw}/\text{day}$. The mammalian RfD was based on the data from a series of studies by Wobeser and collaborators done on ranch mink. A NOAEL of 55 $\mu\text{g}/\text{kg bw}/\text{day}$ was estimated from these studies. The estimated LOAEL, based on damage to the nervous system and liver, was 180 $\mu\text{g}/\text{kg bw}/\text{day}$.

Based on the data developed for the health assessment, the human LOAEL and RfD are orders of magnitude lower than the corresponding LOAELs and RfD of the other animals. There is a great deal of uncertainty in this comparison. It must be noted that the effects in humans are based on the RfD definition of a critical effect; that is the most sensitive reported adverse effect or indicator of adverse effect. The effects reported for mammals (i.e., neurologic damage in the mink) and birds (i.e., reproductive effects in mallards) would be considered frank effects in the human RfD methodology. The observations in laboratory animals indicate that it would be reasonable to expect more subtle and less damaging effects of methylmercury to occur at lower doses than the wildlife LOAEL and NOAEL.

The information assessed in this Report suggests that ecosystems most at risk from airborne releases of mercury exhibit one or more of the following characteristics:

- They are located in areas where atmospheric deposition of mercury is high;
- they include surface waters already impacted by acid deposition;
- they possess characteristics other than low pH that result in high levels of bioaccumulation; and/or
- they include sensitive species.

The adverse effects of methylmercury on wildlife have been described and quantified. For wildlife the importance of site-specific effects of mercury exposure are anticipated to be greater than for humans in the general population because wildlife obtain their fish from a much more limited geographic area than do people.

Limitations of the Wildlife Assessment

There is uncertainty and variability associated with each WC. These include lack of long-term studies for mammals, lack of a no adverse effects level for birds, and extrapolation from one species to another. It is not known if the species selected for WC development are the most sensitive or appropriate

species; also, it is not known if protecting individual animals or species will guarantee protection of their ecosystem from harmful effects of mercury. There are uncertainties and expected variability in the BAF; it was the subject of a quantitative uncertainty analysis.

For wildlife risk assessment, as for humans, mercury toxicity among wildlife involves neurological effects. Available toxicology data from laboratory-based studies of wildlife exposed to methylmercury have measured only gross clinical signs and symptoms of disease and death or pathological changes accompanying these clinically evident changes. Physiologically based evaluation of wildlife has not been done. The importance of more subtle endpoints of neurological function is anticipated to be relevant to such practical questions as the ability of visual hunters such as the loon to find food.

The risk assessment for wildlife made the assumption that the primary source of mercury exposure to the selected species was contaminated fish. Since mercury bioaccumulation is largely through aquatic ecosystems, it is reasonable to focus attention on wildlife species whose feeding habits are tied to these systems. Existing data permit a general treatment of mercury exposure and effects on such populations. For some species, such as the kingfisher and river otter, it can be reasonably assumed that fish always comprise a high percentage of the diet. For others, such as the eagle and mink, considerable variations in diet are likely to exist. Still others, such as the Florida panther, consume prey (such as the raccoon) which consume variable amounts of aquatic biota, but which in South Florida are closely linked to the aquatic food chain. A more accurate characterization of the risk posed by mercury to a specific group of animals occupying a given location will depend on the collection of necessary supporting information: food habits, migratory behavior, breeding biology, and mercury residues in preferred prey items.

To improve the characterization of risk, research needs highlighted in the preceding sections should be addressed. Additional work to decrease uncertainty should be directed toward the exposure assessment. Validated local and regional atmospheric fate and transport models are needed. This should utilize long-term national monitoring networks. Data to improve understanding of movement of mercury through environmental media are also needed. The bioaccumulation factors are major sources of uncertainty. This uncertainty will be decreased by improved data to use in the parameters of the bioaccumulation factor equations and by increased understanding of mercury biogeochemistry in water bodies.

4. MANAGEMENT ALTERNATIVES

Possible Control Strategies

Effective control of mercury emissions may require a mix of strategies. The four major types of control techniques reviewed include:

- Pollution prevention measures, including product substitution, process modification and materials separation;
- Coal cleaning;
- Alternative approaches; and
- Flue gas treatment technologies.

Table 4-1 summarizes mercury control techniques for selected source categories. Pollution prevention may be suitable for those processes or industries where a mercury substitute is demonstrated and available (e.g., mercury cell chlor-alkali plants). Another pollution prevention measure is material separation, which may be an appropriate approach for processes where mercury-containing products are disposed of by incineration, or where mercury can be reduced in the fuel prior to the fuel being combusted (e.g., medical waste incineration). Conventional regulatory strategies may be applicable when mercury is emitted to the environment as a result of trace contamination in fossil fuel or other essential feedstock in an industrial process (e.g., cement manufacturing). Other non-traditional approaches such as emissions trading or other market-based approaches may also prove feasible for mercury control. In addition, emissions control is only one possible means for reducing human exposure. For example, the issuance of fish advisories (or increased public education about advisories already in place) is an alternative that would need to be explored when selecting among strategies for reducing risks to human health (though not to ecosystems).

Cost-effective opportunities to deal with mercury during the product life-cycle, rather than just at the point of disposal, need to be pursued. A balanced strategy which integrates end-of-pipe control technologies with material substitution and separation, design-for-environment, and fundamental process change approaches is needed. In addition, international efforts to reduce mercury emissions as well as greenhouse gases will play an important role in reducing inputs to the global reservoir of mercury.

As noted above, because of the current, limited scientific understanding of the environmental fate and transport of this pollutant, it is not possible to quantify the contribution of U.S. anthropogenic emissions relative to other sources of mercury, including natural sources and re-emissions from the global pool, on methylmercury levels in fish consumed by the U.S. population. Mercury methylation and subsequent uptake in fish is complex and not well understood. 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 analyses of control technologies and costs presented in this Report are not intended to replace a thorough regulatory analysis, as would be performed for a rulemaking. The information presented is

**Table 4-1
Summary of Mercury Control Techniques for Selected Source Types**

Mercury Control Technique	Applicable Source Type	Estimated Mercury Removal Efficiency	Cross-Media Impacts?^a	Other Pollutants Controlled	Comments
Product substitution (e.g., batteries, fluorescent lights)	MWCs, MWIs	Variable, depending on the extent of substitution	Yes	Could include other components of mercury-containing batteries, fluorescent lights and other products	<ul style="list-style-type: none"> Product substitution has reduced the use of mercury in household batteries Use of mercury-containing fluorescent lights has increased because of their energy efficiency, but lower mercury content is being achieved The impact of product substitution to other areas depends on specific circumstances, including technical and economic feasibility
Process modification	Mercury cell chlor-alkali plants	100%	Yes	None directly	<ul style="list-style-type: none"> In 1994, about one-half of the chlor-alkali plants used mercury-free processes Because the membrane cell process has lower electricity demands than the mercury cell process, plant conversion results in an energy savings Additional savings presumably also result by avoiding costs of recycling or disposing of mercuric wastes
Materials separation	MWCs and MWIs	Variable, depending on the extent of separation	Yes	Could include other components of mercury-containing wastes burned in MWCs or MWIs	<ul style="list-style-type: none"> Separation of low-volume materials containing high mercury concentrations (e.g., batteries, fluorescent lights, thermostats and other electrical items) can reduce mercury input to a combustor without removing energy content of the waste stream Household battery separation has been implemented by several communities; program efficiency ranges from 3 to 25 percent Material separation programs at hospitals have been successful

**Table 4-1 (continued)
Summary of Mercury Control Techniques for Selected Source Types**

Mercury Control Technique	Applicable Source Type	Estimated Mercury Removal Efficiency	Cross-Media Impacts? ^a	Other Pollutants Controlled	Comments
Carbon filter beds	MWCs, utility boilers, industrial boilers	99%	Yes	Residual organic compounds, other heavy metals, SO ₂ , acid gases	<ul style="list-style-type: none"> Currently applied to five full-scale power plants in Germany, and planned to be installed on five hazardous waste incinerators in Europe Technically feasible to other sources, such as MWIs or smelters, but has not been applied Potential negative effects associated with the disposal of spent carbon and the potential for fires in the bed
Wet scrubbing	MWCs, MWIs, boilers	Can be >90% for water-soluble species; limited for elemental mercury	Yes	Acid gases, metals, particulate matter, dioxins, furans	<ul style="list-style-type: none"> Have not been applied to MWCs in the U.S., although they have been used at MWCs in Europe and MWIs in the U.S. 25 percent of coal-fired boilers currently have wet scrubbers Requires treatment of wastewater prior to disposal May form more toxic, lesser-chlorinated dioxin and furan congeners
Depleted brine scrubbing	Chlor-alkali plants	98%	Yes	None	<ul style="list-style-type: none"> Very little information is available on this technique
Treated activated carbon adsorption	Chlor-alkali plants	90%	Yes	Residual organic compounds, other heavy metals, SO ₂ , acid gases	<ul style="list-style-type: none"> Very little information is available on this technique In 1984, carbon bed systems were in use at 8 of the 20 chlor-alkali plants in operation in the U.S. at that time

**Table 4-1 (continued)
Summary of Mercury Control Techniques for Selected Source Types**

Mercury Control Technique	Applicable Source Type	Estimated Mercury Removal Efficiency	Cross-Media Impacts:^a	Other Pollutants Controlled	Comments
Selenium filters	Primary copper smelters, primary lead smelters, and (more limited) MWCs, crematories, power plants	90%	Yes	Particulate matter, acid gases	<ul style="list-style-type: none"> Factors that influence performance include inlet mercury concentrations, flue gas temperature and flue gas dust content Four known applications at smelters as well as a MWC and a crematory in Sweden; known installation at a German power plant; potentially applicable to MWIs Spent filter containing selenium and mercury must be landfilled after use More information needed on the possibility of selenium being emitted from the filter itself
Activated carbon injection	MWCs, MWIs, utility boilers	50-90+%	Yes	Chlorinated dioxins and furans, potentially other semi-volatile organics	<ul style="list-style-type: none"> Activated carbon injection efficiencies reported for utility boilers are based on pilot-scale data and as such have a high degree of uncertainty Factors that influence performance include flue gas temperature, amount of activated carbon injected, type of particulate matter collector, concentration and species of mercury in flue gas and type of carbon used Addition of carbon could have significant impact on amount of particulate matter requiring disposal from utility boilers, but not from MWCs or MWIs

^a For the purpose of this table, cross-media impacts refer to the potential to transfer and release mercury to media other than air, such as soil, ground water, and surface water. For example, carbon filter beds and wet scrubbers remove mercury from air emissions but result in the generation and disposal of mercury-containing solid and liquid wastes, respectively. In the case of product substitution, cross-media impacts refer to the potential to decrease airborne emissions of mercury at one site but increase such emissions elsewhere.

intended to present the range of available options and provide a relative sense of the extent of mercury reductions achievable and the general magnitude of the cost of such reductions.

Pollution Prevention Measures

One possible means of achieving reductions in mercury emissions is through the use of pollution prevention or source reduction. Such approaches to achieving reductions involve changes in processes or inputs to reduce or eliminate emissions of mercury from a particular product or process. They could include, for example, the replacement of mercury with an appropriate substitute or the use of low-mercury constituents.

In considering opportunities for pollution prevention or source reduction it is important to consider both the potential reductions achievable and the costs of these options. Any consideration of the potential reductions, should examine whether (and the extent to which) emission reductions from the particular sources in question will yield reductions in risk to public health and the environment. It is also essential to understand the costs associated with implementing a pollution prevention measure, including any changes in the quality of the end product.

Removing mercury-containing products such as batteries, fluorescent lights and thermostats from the waste stream can reduce the mercury input to waste combustors without lowering the energy content of the waste stream. The mercury removal efficiency would vary, however, depending on the extent of the separation. Many materials in wastes contain mercury. Materials that comprise a large portion of the waste stream, such as paper, plastic, dirt and grit and yard waste, contain very low concentrations of mercury. Therefore, obtaining appreciable mercury reduction from separation of these types of materials would require separating a large fraction of the total waste stream. Separating these materials would counter the intended purpose of the combustion process, which is to disinfect and reduce the volume of waste materials.

Other materials contain higher concentrations of mercury, but make up only a very small portion (less than 1 percent) of the total waste stream. These materials include mercuric oxide batteries, fluorescent lights, thermostats and other electrical items. Separation of such materials can reduce mercury input to a combustor without removing any of the energy content of the waste stream. To evaluate a materials separation program, the feasibility and costs of separating a particular material should be compared with the mercury emission reduction achieved. Furthermore, the current and future mercury reduction achieved by separating a certain material should be considered since the mercury content of some items such as household batteries has already declined considerably.

Coal Cleaning

Coal cleaning is another option for removing mercury from the fuel prior to combustion. In some states, certain kinds of coal are commonly cleaned to increase its quality and heating value. Approximately 77 percent of the eastern and midwestern bituminous coal shipments are cleaned in order to meet customer specifications for heating value, ash content and sulfur content. Any reduction in mercury content achieved by coal cleaning results in a direct decrease in mercury emissions from the boiler. The mercury removed by cleaning processes is transferred to coal-cleaning wastes, which are commonly in the form of slurries. No data are available to assess the emissions of mercury from coal-cleaning slurries.

Volume II of this Report (An Inventory of Anthropogenic Mercury Emissions in the United States) presents available data on the mercury concentrations in raw coal, cleaned coal and the percent reduction achieved by cleaning. These data, which cover a number of different coal seams in four states (Illinois, Pennsylvania, Kentucky and Alabama), indicate that mercury reductions range from 0 to 64

percent, with an overall average reduction of 21 percent. This variation may be explained by several factors, including different cleaning techniques, different mercury concentrations in the raw coal and different mercury analytical techniques. It is expected that significantly higher mercury reductions can be achieved with the application of emerging coal preparation processes. For example, in one bench-scale study, five types of raw coal were washed by conventional cleaning methods followed by column froth floatation or selective agglomeration. Conventional cleaning and column froth floatation reduced mercury concentrations from the raw coals by 40 to greater than 57 percent, with an average of 55 percent. Conventional cleaning and selective agglomeration reduced mercury concentrations from the raw coals by greater than 63 percent to 82 percent, with an average of 68 percent. In a second bench-scale study in which three types of coals were cleaned with a heavy-media-cyclone (a conventional cleaning method) followed by a water-only-cyclone and a column froth floatation system, mercury concentrations in the raw coal were reduced by as much as 63 to 65 percent. Bench-scale testing is also being carried out by DOE to investigate the use of naturally occurring microbes to reduce mercury (and other trace elements) from coal.

Alternative Approaches

There are a variety of flexible approaches for reducing the emissions of hazardous air pollutants. These include incentive- or market-based systems, “co-control,” and energy conservation and renewable energy initiatives.

Incentive-based systems are tools that provide industry with more flexibility than traditional regulatory programs. In such a system, the regulatory agency generally sets a ceiling on allowable emissions (a cap) for each source along with clear and certain penalties for missing the target, but regulated entities have complete choice in how these targets will be met. The cost to industry is determined by the market and by the innovation used in meeting the cap. Emissions cap programs allow for increased incentives because sources that reduce emissions below their cap can sell the surplus reduction to sources that cannot achieve their cap. Trading is promising where sources have different compliance costs, or where local environmental impacts are minimal. Sources that reduce emissions before they are required to do so can “bank” the excess reductions and save them for later. Examples of existing market-based programs include the SO₂ allowance trading and NO_x averaging programs implemented under Title IV of the CAA Amendments to reduce acid deposition; the Regional Clean Air Incentives Market Program and Rules developed in California to reduce emissions of NO_x, SO_x, and reactive organic compounds; and U.S. EPA’s Lead Trading Program designed to reduce the emissions of lead from gasoline in the mid-1980’s.

Incentive-based systems to reduce mercury emissions, either through regulation or voluntary means, may be attractive to utilities and other facilities for several reasons: to reduce mercury emissions at a lower per unit cost, to insure against future regulation, to reduce the compliance costs of regulation, to bank credits toward future regulatory requirements, to build experience with technology and to demonstrate environmental leadership. Also, incentive-based programs could provide financing for the control of mercury among different industries (and potentially other countries) and may be a viable option for utilities and other sources where cost-effective technologies have yet to be identified.

Co-control refers to the control of mercury by control devices or other management measures that were designed or prescribed to limit the emissions of pollutants other than mercury. Co-control can also be achieved through the implementation of the National Ambient Air Quality Standards (NAAQS) for ozone and particulate matter (PM). In support of the revised ozone and PM NAAQS, the U.S. EPA conducted numerous detailed analyses to predict what control approaches industry might use to achieve the new standards. Fuel switching, in which one fuel is switched to another (e.g., high-sulfur coal to low-sulfur coal, or coal to natural gas) to achieve emissions reductions, is also an alternative to direct control.

U.S. EPA estimates that implementation of the New Fine Particle Standard for ambient air quality through a regional control strategy that significantly reduces SO_x below the CAA's Title IV requirements can indirectly lower forecasted mercury emissions in 2010 by about 11 tons from electric power generation by units burning fossil fuels. This reduction occurs from both the additions of flue gas desulfurization units (scrubbers) at coal-fired boilers to lower SO_x emissions and through greater reliance by the power industry on producing electricity from natural gas as another way to reduce SO_x. In the Regulatory Impact Analysis for the new NAAQS, U.S. EPA estimated that in 2010 a regional SO_x reduction strategy for the electric power industry to lower fine particle formation will lead to the installation of scrubbers on additional 60 GWs of coal-fired capacity (increasing forecasted scrubber capacity under Title IV by about two-thirds). U.S. EPA assumes that scrubbers remove close to 30 percent of the mercury contained in coal flue gas. U.S. EPA also estimated that electricity produced from natural gas would increase by 16 percent above baseline levels. Natural gas combustion produces negligible levels of mercury emissions.

Title IV of the CAA also encourages energy conservation measures and use of renewable energy as a long-term strategy for reducing air pollution and other adverse effects of energy production and use. Renewable energy is defined as energy that is derived from biomass, solar, geothermal or wind.

Flue Gas Treatment Technologies

Most metals have sufficiently low vapor pressures at typical air pollution control device operating temperatures that condensation onto particulate matter is possible. Mercury, on the other hand, has a high vapor pressure at typical control device operating temperatures, and collection by particulate matter control devices is highly variable.

In Volume VIII of this Report (An Evaluation of Mercury Control Technologies and Costs), add-on controls to reduce mercury emissions are described in detail including information on commercial status, performance, applicability to the specified mercury emission sources, and secondary impacts and benefits. The controls described are:

- Carbon filter beds;
- Wet scrubbing for waste combustors and utility boilers;
- Depleted brine scrubbing;
- Treated activated carbon adsorption;
- Selenium filters; and
- Activated carbon injection.

The most important conclusions from the assessment of flue gas treatment technologies include:

- Factors that enhance mercury control are low temperature in the control device system (less than 150 °Celsius [°C] [300 to 400 °Fahrenheit (°F)]), the presence of an effective mercury sorbent and a method to collect the sorbent. In general, high levels of carbon in the fly ash enhance mercury sorption onto particulate matter which is subsequently removed by the particulate matter control device. Additionally, the presence of hydrogen chloride (HCl) in the flue gas stream can result in the formation of mercuric chloride (HgCl₂), which is readily adsorbed onto carbon-containing particulate matter, or can be efficiently scrubbed by a wet FGD system. Conversely, sulfur dioxide (SO₂) in flue gas can act as a reducing agent to convert oxidized mercury to elemental mercury, which is more difficult to collect.
- Conversion of mercury cell chlor-alkali plants to a mercury-free process is technically feasible and has been previously demonstrated.

- Control technologies designed for control of pollutants other than mercury (e.g., acid gases and particulate matter) vary in their mercury-removal capability, but in general achieve reductions no greater than 50 percent (except for high removal efficiencies for HgCl_2 by wet scrubbers) .
- Selenium filters are a demonstrated technology in Sweden for control of mercury emissions from lead smelters. Carbon filter beds have been used successfully in Germany for mercury control on utility boilers and MWC's. These technologies have not been demonstrated in the U.S. for any of these source types.
- Injection of activated carbon into the flue gas of MWC's and MWI's can achieve mercury reductions of at least 85 percent. The addition of activated carbon to the flue gas of these source types would not have a significant impact on the amount of particulate matter requiring disposal.
- No full-scale demonstrations of activated carbon injection for utility boilers have been conducted in the U.S. Based on limited pilot-scale testing, activated carbon injection provides variable control of mercury for utility boilers (e.g., the same technology might capture 20 percent of the mercury at one plant and 80 percent at another). The most important factors affecting mercury control on utility boilers include the flue gas volume, flue gas temperature and chloride content, the mercury concentration and chemical form of mercury being emitted.
- The chemical species of mercury emitted from utility boilers vary significantly from one plant to another. Removal effectiveness depends on the species of mercury present. To date, no single control technology has been identified that removes all forms of mercury.
- The addition of activated carbon to utility flue gas for mercury control would significantly increase the amount of particulate matter requiring disposal.

Cost of Controls

The overall approach for assessing the cost of "end-of-pipe" flue gas treatment technologies was to select a subset of source categories on the basis of either their source category emissions in the aggregate or their potential to be significant point sources of emissions. Consideration was also given to whether a particular source category was a feasible candidate for application of a control technology-based standard under section 112 of the CAA. The cost analyses cover four source categories: municipal waste combustors (MWC), medical waste incinerators (MWI), chlor-alkali plants, and utility boilers.

In addition to determining the cost effectiveness of applying mercury control technology, a financial analysis was performed to evaluate the affordability of mercury control (in terms of potential price increases or impacts on financial impact) for the selected source categories.

**Table 4-2
Potential Mercury Emission Reductions and Costs for Selected Source Categories**

<u>Mercury Source Category</u>	<u>Number of Facilities</u>	<u>% of U.S. Mercury Emission Inventory</u>	<u>Mercury Control Techniques</u>	<u>Potential National Reductions^a</u>	<u>Potential National Annual Costs^b</u>	<u>Cost-Effectiveness (\$/lb of mercury removed)^c</u>
Municipal waste combustors	129	18.7	<u>Material separation</u> <u>Product substitution</u> <u>Activated carbon injection</u> <u>Carbon filter beds</u> <u>Polishing wet scrubber</u>	26 tons (90% reduction) By 2000, based on final emissions guidelines	\$11.4-47 million	\$211-870
Medical waste incinerators	~2,400	10.1	<u>Material separation</u> <u>Wet scrubber or dry scrubber with carbon</u> <u>Activated carbon injection</u>	15 tons (95% reduction) By 2000, based on final emissions guidelines	\$60-120 million ^d	\$2,000-\$4,000 ^d
Coal-fired utility boilers	426 (1,043 boilers)	32.6	<u>Fuel switching</u> <u>Advanced coal cleaning</u> <u>Activated carbon injection</u> <u>Carbon filter beds</u>	37 tons (90% reductions) ^e	\$5 billion	\$67,700-70,000
Chlor-alkali plants using the mercury cell process	14	4.5	<u>Co-control: ozone and PM NAAQS</u>	11 tons ^f	No incremental mercury control costs	No incremental mercury control costs
			<u>Process modification</u> <u>Depleted brine scrubbing</u> <u>Treated activated carbon adsorption</u>	7.1 tons (100% reduction)	\$65 million	\$4,590
Total	~3,600	65.9			~\$5.2 billion	

NOTE: The underlined mercury control techniques are the techniques on which potential national reductions and potential national annual costs are based.

^a Estimated reductions assuming every facility could achieve the reduction listed.

^b Potential national costs are estimates only and assume all facilities would incur the same costs as the model plants used in the analysis.

^c Where cost-effectiveness values are presented as a range, the values reflect the range across facilities of different sizes.

^d Cost of control should not be attributed to mercury control alone. Wet scrubbers efficiently remove nine other pollutants from the MWI flue gas as required by the emission guidelines for MWIs.

^e The potential national reductions reflects sufficient amounts of activated carbon to control mercury emissions from coal-fired utility boilers by 90 percent. Activated carbon injection has not been demonstrated for a full-scale utility boiler application. Control costs are upper bound based on high temperature activated carbon injection. The 37 tons reduction is 90 percent of 41 tons, accounting for the 11 ton reduction from the ozone and PM NAAQS.

^f Assumes some fuel switching and additional installation of wet scrubbers which are assumed to remove 30 percent.

Table 4-2 presents the four source categories for which a control technology and cost analysis was performed. The selection of a particular type of control for the cost analysis should not be construed to mean that the U.S. EPA has selected, or has preference for, this technology for a given source category. The table presents the number of facilities in each category and the percent contribution of each to the national inventory. Potential national mercury reductions, potential national control costs and cost-effectiveness estimates are also presented. These estimates are based on the assumption that all plants within a source category will achieve the same reductions and incur the same costs as the model plants used in the analysis. Because this assumption would not be applicable in all circumstances, the estimates of potential reductions and costs should be used only for relative comparisons among the source categories to give an initial indication as to where mercury controls could provide the most emission reduction for the least cost.

The cost of mercury control incurred by any specific facility may be underestimated by the cost analysis presented in this Report because of variability inherent in the assumptions that were made in the analyses. These assumptions include the efficiency of the various control techniques for reducing mercury, the amount of mercury in the flue gas stream and other site-specific factors such as down-time and labor costs. In addition, costs for monitoring and recordkeeping were not included in the cost analyses. These requirements would be specific to a regulatory action. On the other hand, the costs represent retrofit application of controls. Installation of controls at new facilities can be significantly less expensive than retrofitting an existing facility.

The estimates of cost for mercury reductions also do not illustrate two important considerations. One is that, as presented, all of the cost of control could mistakenly be attributed to mercury removal. As described previously in this Report, many of these controls achieve reductions of other pollutants as well (e.g., acid gases, dioxin, other metals). In some cases (e.g., the emission guidelines for MWI), the choice of control technology or control strategy is aimed at reducing pollutants other than mercury. In these cases, there is a co-control benefit of mercury reduction. The benefits of reducing other pollutants should be considered when interpreting the mercury control costs. Second, the technologies available for mercury control represent relatively new applications of these technologies. Thus, in the future, it is likely that as new or emerging technologies develop, the cost-effectiveness of control will improve. Air pollution control and prevention techniques are continuously under development and improvement. There is a fairly rapid pace of innovation in the air pollution control sector. The demand for cleaner products and cleaner processes that lower overall costs, combined with the necessity for improved air and water quality, create strong incentives for technological innovation and a growing market for such innovations. As the demand for more innovative, cost-effective and cost-saving technologies increase, new technologies will move from the research and development or pilot program phase to commercial availability.

While existing technology will play a key role in reducing mercury from some sources, emerging technology may be more appropriate for others. Innovations in environmental policies may also play a key role in developing a national management strategy for mercury. These innovations could include multi-media approaches, greater emphasis on pollution prevention, regional control strategies and optimization of co-control opportunities.

Ongoing U.S. EPA Activities to Reduce Mercury in the Environment

Mercury is a priority pollutant across numerous U.S. EPA programs including air, water, hazardous waste and pollution prevention. There are numerous activities currently underway to reduce mercury emissions and releases to the environment. A number of these activities are described below which reflect the broad scope U.S. EPA's approach to the mercury issue.

Clean Air Act Initiatives

The U.S. EPA already has efforts underway to reduce mercury emissions from industrial sources. Specific actions being taken under the Clean Air Act include the following:

- The U.S. EPA has promulgated final emission limits for municipal waste combustors and medical waste incinerators under the authority of section 129 of the CAA. Emission standards have also been proposed for hazardous waste incinerators.
- The U.S. EPA is evaluating the impacts of mercury reductions for the following source categories: commercial/ industrial boilers, chlor-alkali plants using the mercury cell process and portland cement kilns.
- The U.S. EPA plans to evaluate whether secondary mercury production should be added to the source category list under section 112(c) of the CAA and subsequently evaluated for regulation under the authority of section 112(c)(6).
- Numerous CAA requirements involve utilities either directly or indirectly. Section 112(n)(1)(B) which required this Mercury Study Report to Congress specified utility boilers for analysis as did section 112(n)(1)(A) which is referred to as the Utility Air Toxics Report to Congress (Utility Study). The Utility Study is charged with evaluating the hazards to public health reasonably anticipated to occur as a result of emissions by electric utility steam generating units of pollutants listed under Section 112(b), including mercury, and to evaluate the impact of other provisions of the CAA on these emissions. The other provisions of the CAA would include the Acid Rain program as well as provisions pertaining to National Ambient Air Quality Standards. The Utility Study is also required to offer a regulatory recommendation with respect to regulation of utility boilers under section 112 of the CAA.
- The “Great Waters” program (section 112(m)) is an ongoing study with biennial reports to Congress required. The program must identify and assess the extent of atmospheric deposition of hazardous air pollutants (including mercury) to the Great Lakes and other specified waters, the environmental and public health attributable to atmospheric deposition and the contributing sources. Two reports have been submitted to Congress which address these issues.

Mercury Task Force

To address cross-media issues, additional pollution prevention options and regulatory authorities, the U.S. EPA has established a Mercury Task Force to consider strategies for coordinating various programs for use, management and disposal of mercury. The Task Force has recommended to the Department of Defense that the Defense Logistics Agency suspend sales of mercury from federal stockpiles through the fiscal year 1996 sales cycle, and are in the process of developing an environmental impact assessment.

The Mercury Task Force continues to consider several approaches for reducing mercury releases and environmental and human health risks associated with mercury exposure. A wide range of options, within a multi-media framework, advocating common-sense pollution prevention programs are being considered. Some areas which the Task Force will explore include evaluation and information transfer of ongoing prevention and control efforts at local, national and international levels; consideration of pollution prevention ideas including product substitution and innovation; recycling and disposal options;

research and science needs; and coordination within U.S. EPA for consistent mercury regulatory programs, as well as coordination with Federal agencies managing mercury.

The findings of the Mercury Study Report to Congress will be considered by the Mercury Task Force as it develops a U.S. EPA mercury strategy.

Virtual Elimination Project

U.S. EPA and Environment Canada are actively developing strategies to achieve the Great Lakes Water Quality Agreement's (GLWQA's) goal that persistent toxic substances should be "virtually eliminated" from the Great Lakes. Because toxic substances enter the Great Lakes from ongoing economic activities, as well as from sites contaminated by past activities, eliminating toxic substances from the Great Lakes requires a three-pronged approach that:

- Reduces the use of toxic substances at the source, through pollution prevention efforts;
- Reduces toxic substance discharges, emissions and other ongoing releases through treatment or other management techniques; and
- Cleans up sites of past contamination, such as contaminated sediments or areas of concern, through remediation efforts.

A central theme underlying the virtual elimination project is that opportunities may exist to alter the decisionmaking environment in which individuals and firms choose to use and release toxic chemicals in their ongoing activities. The project focuses on government actions -- or "signals" -- such as regulatory or voluntary programs that influence the economic and legal costs and benefits associated with using a particular chemical. These signals, which translate into costs for an affected entity, can motivate individuals and firms to choose pollution prevention based on their own economic interests.

Other Pollution Prevention Programs

U.S. EPA is working with state and local governments to develop a national network of prevention programs that will assist regulators at all levels of government in promoting pollution prevention. To that end, U.S. EPA is providing funding support, technical assistance, information dissemination and forming federal/state/local government partnerships to focus efforts on pollution prevention as the national goal for environmental management.

State and federal partnerships have already led to actions that will reduce mercury loadings to the environment. For example, the National Wildlife Federation, funded in part by U.S. EPA, has recently released a report detailing how hospitals in Detroit, Michigan; Grand Rapids, Michigan; Boston, Massachusetts; and Duluth, Minnesota have successfully reduced mercury releases by applying pollution prevention principles. This report contains practical and cost effective suggestions for improving the environmental performance of hospitals and to help them meet increasingly stringent limits in regulatory permits. Industry groups have also made advances in pollution prevention (see text box below on Chrysler Corporation).

U.S. EPA is working continually to incorporate pollution prevention into the mainstream of its work. Over the last six years, the agency has undertaken a concerted effort to find the best ways to incorporate prevention into regulations and permitting. For example, in 1992, U.S. EPA began an effort to evaluate pollution prevention options for a number of new regulations under development. This effort, called the Source Reduction Review Project, required U.S. EPA's media offices to identify multi-media approaches to addressing air, water and solid waste regulations. Another program, U.S. EPA's Common

Sense Initiative, created a pollution prevention framework for environmental protection on an industry-by-industry basis by focusing on opportunities to change complicated or inconsistent environmental requirements into comprehensive strategies. The goal of each of these programs is a cleaner environment at less costs to taxpayers.

In addition, and on a broader scale, U.S. EPA is currently developing a long-term plan to mitigate the risks associated with mercury and other chemicals of concern under its Persistent, Bioaccumulative and Toxic (PBT) Chemicals strategy by using pollution prevention principles. Through its current efforts on the PBT strategy, U.S. EPA will focus these activities more intensively on the key persistent, bioaccumulative pollutants, especially mercury. U.S. EPA expects that through partnership with states and local organizations, and in collaboration with industry, there will be more opportunities to use pollution prevention as a means to mitigate the potential risk to human health and the environment associated with exposure to mercury.

Life Cycle Cost Management in the Auto Industry

Industry remains at the center of pollution prevention activities. Studies have shown that the economic benefits can be compelling arguments in favor of pollution prevention, but only when managers are able to see the cost savings that pollution prevention would bring. Environmental accounting is the key factor in demonstrating to businesses the value of prevention. The following serves as a specific example of pollution prevention in practice to reduce mercury loadings.

The Chrysler Corporation is now removing or replacing all mercury switches that have been traditionally used in its underhood convenience light applications. Chrysler has done so as a result of the application of life cycle cost management methodologies that are advocated by the U.S. EPA Pollution Prevention Division's Environmental Accounting Project. This Project is a cooperative effort with business, academia and others to promote sound management accounting and capital budgeting practices which better address environmental costs. The project encourages and motivates business to understand the full spectrum of environmental costs and integrate these costs into decision making. Chrysler is partnering with the Project to share its environmental accounting experience and case studies with the 800-plus members of the Project-facilitated Environmental Accounting Network.

By applying the principles of environmental accounting, the Chrysler Corporation determined that it could cost-effectively replace the mercury switches with a rolling ball switch or remove the switches altogether. For the first group of cars on which Chrysler tested the feasibility of substitution and removal, it determined that it could avoid \$40,000 in costs. Most of those costs were associated with the documentation of the removal of mercury switches from the vehicle before disposal, and with the potential liability for any mercury that enters the environment following vehicle disposal. After conducting their own total cost analyses, other auto manufacturers are now following suit and are actively removing mercury switches from their own automobiles.

International Activities

On an international level, mercury is being addressed as part of the Great Lakes Binational Toxics Strategy and the North American Regional Action Plan, among other efforts. These two initiatives are summarized below.

Binational Strategy

The Great Lakes Binational Toxics Strategy, which was signed between Canada and the United States on April 7, 1997 (U.S. EPA and Environment Canada, 1997), was developed to help achieve the objectives of the 1987 GLWQA. Although both Canada and the United States have domestic virtual elimination strategies as described above, a coordinated strategy is necessary for the greatest reduction in toxic substances throughout the Great Lakes Basin.

The Binational Strategy provides the framework to achieve quantifiable goals in a specified time frame (1997 to 2006) for targeted persistent toxic substances, especially those which bioaccumulate. The Strategy recommends that goals be accomplished through a four-step process:

- Gather information on generation, uses, and sources of the pollutant within and outside the Great Lakes Basin;
- Analyze current regulatory and non-regulatory programs and initiatives that manage or control the pollutants and identify the gaps in these regulations that offer opportunities for reductions;
- Develop cost-effective options and provide recommendations for increasing the pace and level of reductions; and
- Recommend and implement actions to achieve goals.

Mercury and mercury compounds are considered immediate priorities and are targeted for reduction and eventual virtual elimination through pollution prevention and other incentive-based actions.

Both the United States and Canada have set “challenge” goals to achieve reductions through implementation of voluntary efforts and regulatory actions. One of these challenges is the commitment of these countries to work together to assess atmospheric inputs of persistent toxic substances 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. Efforts will be made to work within the existing international framework to reduce releases of such pollutants from remaining long-range sources.

North American Regional Action Plan

The North American Regional Action Plan (NARAP) is one of a number of regional undertakings that stem from the North American agreement on Environmental Cooperation between the governments of Canada, the united Mexican states and the United States of America (Parties). The NARAP calls for the development of regional action plans for selected persistent and toxic substances as a first priority in the Parties’ common desire to address national and regional concerns associated with the sound management of chemicals.

The action plans are designed to reflect a long-term commitment to regional action. The sharing and transfer of information and best practices are seen as an important means of enhancing national capacity for the sound management of chemicals. Other important elements and outcomes of these cooperative initiatives include collaboration and cooperation in the measurement, monitoring, modeling, research and assessment of selected persistent and toxic substances in environmental media. Such cooperation will improve the quality, availability and relevance of the “environmental information” needed to make informed and responsible decisions throughout the implementation of the action plans.

Mercury is one of the targeted chemicals and has its own action plan designed to unite the Parties in their joint and differentiated efforts to reduce the exposure of North American ecosystems, fish and

wildlife, and especially humans, to mercury through the prevention and reduction of anthropogenic releases of mercury to the North American environment. The objectives of the action plan are to reduce mercury levels in and fluxes among environmental media in order to prevent or minimize exposure to ecosystems, fish and wildlife, and humans.

Implementation on the mercury action plan is predicated on the following objectives:

- Building on existing initiatives. Examples include the Great Lakes Binational Toxics Strategy, described above.
- Promoting North American regional and global activities. The mercury action plan will promote regional actions to reduce mercury emissions and serve as an example for initiatives under development throughout the region and globally.
- Best practices. The action plan will promote the sharing, transfer, and general adoption of policies, programs, technologies, and other measures that have proven to be cost-effective and environmentally appropriate.
- Challenging stakeholders to take cooperative action on mercury. The action plan promotes stakeholder partnerships in information and technology exchanges.
- Improving scientific understanding. The action plan will use government and private-sector partnerships to fund research and monitoring, and to advance the science and technology state-of-knowledge for mercury.
- Capacity building in Mexico. The Parties are committed to working cooperatively to build Mexico's capacity with respect to the prevention and reduction of anthropogenic releases of mercury and the sound management of mercury.
- Extended Americas. The Parties agree to actively promote cooperation with other countries to promote pertinent initiatives.

Specific actions outlined for mercury include a series of workgroups and workshops to assess the current knowledge on mercury issues and compile information into shared databases. An implementation committee will provide oversight of the action plan.

5. RESEARCH NEEDS

The following sections summarize the major research needs identified for each of the study areas addressed in this Report.

Anthropogenic Mercury Emissions in the United States

An effort has been made to characterize the uncertainties (at least qualitatively) in the emissions estimates for the various source categories described. There are inherent uncertainties in estimating emissions using emission factors. To reduce these uncertainties, a number of research needs remain, including the following.

1. Source test data are needed from a number of source categories that have been identified as having insufficient data to estimate emissions. Notable among these are mobile sources, landfills, agricultural burning, sludge application, coke ovens, petroleum refining, residential woodstoves, mercury compounds production and zinc mining. A number of manufacturing sources were also identified as having highly uncertain emissions estimates. Notable among this category are secondary mercury production, commercial and industrial boilers, electric lamp breakage, primary metal smelting operations and iron and steel manufacturing. The possibility of using emissions data from other countries could be further investigated.
2. Development and validation of a stack test protocol for speciated mercury emissions is needed.
3. More data are needed on the efficacy of coal cleaning and the potential for slurries from the cleaning process to be a mercury emission source.
4. More data are needed on the mercury content of various coals and petroleum and the trends in the mercury content of coal burned at utilities and petroleum refined in the U.S.
5. Additional research is needed to address the potential for methylmercury to be emitted (or formed) in the flue gas of combustion sources.
6. The importance (quantitatively) of re-emission of mercury from previously deposited anthropogenic emissions and mercury-bearing mining waste needs to be investigated. This would include both terrestrial and water environments. Measuring the flux of mercury from various environments would allow a determination to be made of the relative importance of re-emitted mercury to the overall emissions of current anthropogenic sources.
7. Determination of the mercury flux from natural sources would help determine the impact of U.S. anthropogenic sources on the global mercury cycle as well as the impact of all mercury emissions in the United States.
8. The use of more sophisticated fate and transport models for mercury will require more detailed emissions data, particularly more information on the chemical species of mercury being emitted (including whether these species are particle-bound) and the temporal variability of the emissions.

Mercury Fate and Transport Modeling

During the development of the mercury fate and transport assessment, many areas of uncertainty and significant data gaps were identified. Many of these have been identified in the document, and several are presented in the following list.

1. Improved analytical techniques for measuring speciated mercury air emissions are needed as well as total mercury emissions from point sources. Laboratory evidence suggests that divalent mercury gas emissions will wet and dry deposit much more readily than elemental mercury gas. Particle-bound mercury is also likely to deposit relatively quickly. Current stack sampling methods do not provide sound information about the fraction of mercury emissions that are in oxidized form. While filters are used to determine particulate mercury fractions, high temperature stack samples may not be indicative of the fraction of mercury that is bound to particles after dilution and cooling in the first few seconds after emission to the atmosphere. Methods for determination of the chemical and physical forms of mercury air emissions after dilution and cooling need to be developed and used to characterize significant point sources.
2. Evaluated local and regional atmospheric fate and transport models are needed. These models should treat all important chemical and physical transformations which take place in the atmosphere. The development of these models will require comprehensive field investigations to determine the important atmospheric transformation pathways (e.g., aqueous cloud chemistry, gas-phase chemistry, particle attachment, photolytic reduction) for various climatic regions.
3. The evaluation of these models will require long-term national (possibly international) monitoring networks to quantify the actual air concentrations and surface deposition rates for the various chemical and physical forms of mercury.
4. Better understanding of mercury transport from watershed to water body including the soil chemistry of mercury, the temporal aspects of the soil equilibrium and the impact of low levels of volatile mercury species in surface soils and water bodies on total mercury concentrations and equilibrium.
5. Better understanding of foliar uptake of mercury and plant/mercury chemistry. (The most important questions: do plants convert elemental or divalent mercury into forms of mercury that are more readily bioaccumulated? Do plants then emit these different forms to the air?) A better understanding of the condensation point for mercury is needed.
6. Better understanding of mercury movement from plant into soil (detritus). May need to refine the models used to account for movement of mercury in leaf litter to soil.
7. The impact of anthropogenic mercury on the "natural," existing mercury levels and species formed in soil, water, and sediments needs better understanding. How does the addition of anthropogenic mercury affect "natural" soil and water mercury cycles? Natural emission sources need to be studied better and their impacts better evaluated.
8. Improved understanding of mercury flux in water bodies and impact of plant and animal biomass are needed. Unlike many other pollutants, most of the methylmercury in a water body appears to be in the biological compartment. The sedimentation rate as well

as benthic sediment:water partition coefficient require field evaluation. Important to consider rivers and other larger water bodies in these flux analyses.

Exposure from Anthropogenic Mercury Emissions in the United States

1. To improve the quantitative exposure assessment modeling component of the risk assessment for mercury and mercury compounds, U.S. EPA would need more and better mercury emissions data and measured data near sources of concern, as well as a better quantitative understanding of mercury chemistry in the emission plume, the atmosphere, soils, water bodies, and biota.
2. To improve the exposure estimated based on surveys of fish consumption, more study is needed among potentially high-end fish consumers, which examines specific biomarkers indicating mercury exposure (e.g., blood mercury concentrations and hair mercury concentrations).
3. A pharmacokinetic-based understanding of mercury partitioning in children is needed. Additional studies of fish intake and methylmercury exposure among children are needed.

Health Effects of Mercury and Mercury Compounds

1. In addition to the ongoing studies identified in the health effects review, further research is necessary for refinement of the U.S. EPA's risk assessments for mercury and mercury compounds. In order to reduce uncertainties in the current estimates of the oral reference doses (RfDs) and inhalation reference concentrations (RfCs), longer-term studies with low-dose exposures are necessary. In particular, epidemiological studies should emphasize comprehensive exposure data with respect to both dose and duration of exposure. Some studies should be targeted to populations identified in this Report as likely to experience methylmercury exposure in fish (e.g., subsistence fishers).
2. The current RfD and RfC values have been determined for the most sensitive toxicity endpoint for each compound; that is, the neurological effects observed following exposure to elemental or methylmercury, and the renal autoimmune glomerulonephritis following exposure to inorganic mercury. For each of these compounds, experiments conducted at increasingly lower doses with more sensitive measures of effect will improve understanding of the respective dose-response relationships at lower exposure levels and the anticipated thresholds for the respective effects in humans. Similar information from developmental toxicity studies would allow determination of RfDs for developmental toxicity (RfD_{dt}) for elemental and inorganic mercury.
3. Research needs include studies which will delineate the most appropriate indicators of neurotoxic effects for exposed adults, children and individuals exposed to methylmercury *in utero*. Well conducted studies are also needed to clarify critical levels at which other toxic effects could occur in humans.
4. Well-conducted studies are also needed to clarify exposure levels at which toxic effects other than those defined as "critical" could occur in humans. For all three forms of mercury, data are inadequate, conflicting, or absent for the following: adverse reproductive effects (effects on function or outcome, including multigeneration

exposure); impairment of immune function; and genotoxic effects on human somatic or germinal cells (elemental and inorganic mercury).

5. Investigations that relate the toxic effects to biomonitoring data will be invaluable in quantifying the risks posed by these mercury compounds. In addition, work should focus on subpopulations that have elevated risk because they are exposed to higher levels of mercury at home or in the workplace, because they are also simultaneously exposed to other hazardous chemicals, or because they have an increased sensitivity to mercury toxicity.
6. There are data gaps in the carcinogenicity assessments for each of the mercury compounds. The U.S. EPA's weight-of-evidence classification of elemental mercury (Group D) is based on studies in workers who were also potentially exposed to other hazardous compounds including radioactive isotopes, asbestos, or arsenic. There were no appropriate animal studies available for this compound. Studies providing information on the mode of action of inorganic mercury and methylmercury in producing tumors will be of particular use in defining the nature of the dose response relationship.
7. The assessment of both noncarcinogenic effects and carcinogenic effects will be improved by an increased understanding of the toxicokinetics of these mercury compounds. In particular, quantitative studies that compare the three forms of mercury across species and/or across routes of exposure are vital for the extrapolation of animal data when assessing human risk. For elemental mercury there is a need for quantitative assessment of the relationship between inhaled concentration and delivery to the brain or fetus; in particular the rate of elemental to mercuric conversion mediated by catalase and the effect of blood flow. Such assessment is needed for evaluation of the impact of mercury exposure from dental amalgam.
8. Work has been done on development of physiologically-based pharmacokinetic models. While one of these has developed a fetal submodel, data on fetal pharmacokinetics are generally lacking. The toxicokinetics of mercury as a function of various developmental stages should be explored. Elemental mercury and methylmercury appear to have the same site of action in adults; research is, therefore, needed on the potential for neurotoxicity in newborns when the mother is exposed. This work should be accompanied by pharmacokinetic studies and model development.

Ecological Assessment for Anthropogenic Mercury Emissions in the United States

1. *Process-based Research.* Mechanistic information is needed to understand the variability that presently typifies the mercury literature. This research includes laboratory and field studies to identify the determinants of mercury accumulation in aquatic food chains and kinetic information that would allow researchers to describe the dynamics of these systems. Areas of uncertainty include: (1) translocation of mercury from watersheds to waterbodies; (2) factors that determine net rates of methylation and demethylation; (3) dietary absorption efficiency from natural food sources; (4) effect of dietary choice; and (5) bioavailability of methylmercury in the presence of dissolved organic material and other potential ligands. In time, it is anticipated that this information can be used to develop process-based models for mercury bioaccumulation in fish and other aquatic biota. Significant progress in this direction is represented by the Mercury Cycling Model (MCM) (Hudson et al., 1994) and by the ISC3M model described in Volume III of this Report and employed in the wildlife exposure characterization.

2. *Wildlife Toxicity Data.* There is a need to reduce the present reliance on a relatively few toxicity studies for WC development. Additional data are needed for wildlife that constitute the most exposed organisms in various parts of the country, and in particular there is need to evaluate whether dietary selenium and endogenous demethylating pathways confer protection to piscivorous birds and mammals. Toxicity studies should examine endpoints relevant to the mode of action of methylmercury, including assessments of both reproductive and behavioral effects. There is also a critical requirement for toxicity data (e.g., growth and fecundity) that can be related to effects on populations, including effects on organisms that comprise the lower trophic levels.
3. *Improved Analytical Methods.* Efforts to develop and standardize methods for analysis of total mercury and methylmercury in environmental samples should be continued. Such methods must recognize the importance of contamination, both during the collection of such samples and during their analysis. It is particularly important that mercury measurements, which at present tend to be operationally defined (e.g., "soluble" or "adsorbed to organic material"), be made in such a way that mercury residues in fish can be correlated with the bioavailable mercury pool. Whenever possible, water samples should be filtered to obtain a measure of dissolved mercury species. As validated methods become available, it is important to analyze for both total and methylmercury so that differences between aquatic systems can be definitively linked to differences in methylmercury levels. Analyzing the two mercury species together will contribute to an understanding of existing data, much of which is reported as total mercury.
4. *Complexity of Aquatic Food Webs.* Present efforts to develop WC values for mercury are based on linear, four-tiered food chain models. Research is needed to determine the appropriateness of this simple paradigm and to develop alternatives if field data suggest otherwise. Of particular interest is whether zooplankton and phytoplankton should be modeled as two different trophic levels. Current information for detritivores and benthic invertebrates is extremely limited, even though their importance in mobilizing hydrophobic organic contaminants has been demonstrated.
5. *Accumulation in Trophic Levels 1 and 2.* Ongoing efforts to understand mercury bioaccumulation in aquatic systems continue to be focused on trophic levels 3 and 4, despite the fact that uncertainties in PPFs are relatively small. Additional emphasis should be placed on research at the lower trophic levels. In particular, there is a need to understand the determinants of mercury accumulation in phytoplankton and zooplankton and how rapid changes in plankton biomass impact these values.
6. *Field Residue Data.* High-quality field data are needed to support process-based research efforts and to determine residue concentrations in the fish and other aquatic biota that wildlife eat. Whenever possible, it is desirable to collect residue data at all trophic levels and to analyze mercury levels in the abiotic compartments of a system (e.g., water and sediments). It is particularly important that such measurements be made in a broader array of aquatic ecosystem types (including both lakes and rivers) so that a better understanding of mercury cycling and accumulation can be obtained.

Residue data from wildlife are needed to identify populations that are potentially at risk. Feathers and fur hold considerable promise in this regard due to the potential for "non-invasive" determination of mercury residues. Laboratory research is required, however, to allow interpretation of these data. Factors such as age, sex, and time to last molt are likely to result in variability among individuals of a single population and need to be understood. Whenever possible, tissue samples should be analyzed for both total and

methylmercury, as well as selenium. This is especially true of the liver. More attention should be given to analysis of mercury levels in brain tissue, since this is the primary site of toxic action. Sampling efforts with wildlife should be accompanied by analyses of likely food items.

7. *Natural History Data.* The development of WC values requires knowledge of what wildlife eat. Fish sampling efforts are frequently focused on species that are relevant to human consumers but that may be of little significance to wildlife. There is an additional need to collect information for macroinvertebrates and amphibians. Seasonal and spatial effects on predation should be explored and methods developed to describe this information adequately. Additional life history data is needed to characterize fully the nature and extent of exposure to mercury. Complicating factors must be considered, including migratory behaviors and sex-specific differences in distribution and resource allocation. It is particularly important that information be collected to support the development of predictive population models for sensitive species. Such models must account for immigration and emigration, density dependent factors, and the observation that mercury often bioaccumulates as animals age resulting in variable residues in breeding animals from a single population.

Risk Characterization

1. A monitoring program is needed to assess either blood mercury or feather/hair mercury of piscivorous wildlife; particularly those in highly impacted areas. This program should include assessment of health endpoints including neurotoxicity and reproductive effects.
2. There is a need to collect additional monitoring data on hair or blood mercury and assess health endpoints among women of child-bearing age and children. This study should focus on high-end fish consumers and on consumption of fish from contaminated water bodies.
3. There is a need for improved data on effects that influence survival of the wildlife species as well as on individual members of the species.
4. There is a need for controlled studies on mercury effects in intact ecosystems.
5. Monitoring data sufficient to validate or improve the local impact exposure models are needed.

Mercury Control Technologies

1. Data from full-scale testing of activated carbon injection at a coal-fired utility boiler.
2. Additional data on the efficiency of various sorbents including fly ash-based sorbents, activated carbon, impregnated carbons and other types of sorbents, in reducing the different chemical species of mercury present in flue gas.
3. Information on the cost-effectiveness and commercialization costs of other technologies for mercury control that are currently in the research stage. These include impregnated activated carbon, sodium sulfide injection, activated carbon fluidized bed and other types of sorbents.

4. Additional data on the ability and cost of conventional or advanced coal cleaning techniques to remove mercury from raw coal. The potential for mercury emissions from coal-cleaning slurries needs to be characterized.
5. Additional data on the fundamental mechanisms responsible for conversion of mercury to other chemical species as a result of combustion of certain coals or post-combustion conditions.
6. Additional information on improving the capture of mercury in wet FGD systems.
7. Additional analyses are required on the feasibility, cost-effectiveness of other mercury emission prevention measures such as emissions trading, emissions averaging, energy conservation, renewable energy, and fuel switching.