

Addressing Atmospheric Mercury: Science and Policy

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and the

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Editor's Note

Mercury, in its many forms and its associated compounds, is typically denoted by several symbols and abbreviations. In this report, the lexicon used by individual presenters in describing the various forms of mercury is employed in the portion of the report pertaining to their work. The following summary is meant to provide assistance in clarifying this usage.

Elemental mercury most frequently appears as Hg^0 and occasionally as $\text{Hg}(0)$. This form has no ionic charge.

Divalent mercury appears in the document as: Hg^{+2} $\text{Hg}(\text{II})$, HgII and Hg^{2+} or mercury II. It is considered quite reactive.

Mercury in particulate form appears as $\text{Hg}(\text{p})$

Methylmercury is also abbreviated as MeHg and is the organo-metallic form responsible for the majority of fish consumption advisories.

TGM - Total Gaseous Mercury encompasses all forms of mercury found in the gaseous state, including all gaseous Hg^{+2} compounds and Hg^0 . Estimation of the quantities present will vary with prevailing sampling conditions and selected sampling methodology.

RGM - Reactive Gaseous Mercury - the portion of TGM considered to be reactive, including ionic mercury in the divalent form (Hg^{+2}), all Hg^{+2} compounds in the gas phase but not gaseous elemental mercury (Hg^0). Estimation of the quantities present will vary with prevailing sampling conditions and selected sampling methodology.

Disclaimer

Every attempt has been made to accurately attribute the expert opinions and comments in this report to those attending the workshop on which it is based. The views and opinions expressed herein are those of the workshop organizers and participants and not those of the Commission for Environmental Cooperation or the International Joint Commission.

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FOREWORD

Opening remarks by Ms. Mary Gusella, Chair of Canadian Section - International Joint Commission

Good Morning.

While I am sorry I cannot be with you in person today, let me welcome you and thank you for sharing your knowledge and expertise at this most important workshop. I would like to extend my personal thanks to the organizers and speakers. John McDonald and the Air Board did an admirable job of organizing this workshop not once, but twice. The collaboration with the CEC and Environment Canada is impressive and bodes well for the co-ordinated effort needed to make progress to reduce mercury emissions and their harmful effects.

I attended my first meeting of the International Air Quality Advisory Board this spring in Bouctouche, New Brunswick. At that time I was very impressed with both the work of the board and with the challenge of the mercury issue. While I admit to not understanding all of the scientific issues associated with mercury, I do believe that it is critically important for the North American community, and the Great Lakes community in particular, to keep up the momentum on this issue, not just with respect to air deposition but also all the other sources. We cannot ignore the serious, long term, inter-generational health impacts of mercury.

As we all know, the modern warnings about the effects of mercury on human health first arose at Minamata Japan in the mid-1950s. The most serious effects were on brain development and functioning in children. These effects included a high incidence of cerebral palsy, seizures and mental retardation, particularly in male children. Methylmercury has been shown to have an affinity for the brain and nervous system. Even low doses have been shown to affect learning and cognitive abilities, as well as muscle coordination, especially in young children. There is enormous interest in the global scientific community in these more subtle effects on the brain and in developing new standards for protection of human health.

At the public meeting at the IJC Biennial Forum in Montreal, we heard that cerebral palsy in males is elevated in some of the Great Lakes Areas of Concern. This seems to be occurring in places where mercury was used in large quantities in the past but we will need a lot more work to state anything definitively on this point. We currently have a project that is being proposed by the Great Lakes Science Advisory Board to assist the Parties to investigate this concern raised by the public. There is also a proposal to host a large two-day Conference on Mercury in the Great Lakes in association with the next Biennial Meeting in the year 2003.

Your work here over the next two days is significant, therefore, in helping us to understand mercury sources and pathways. This understanding of sources and pathways will in turn help to answer the question: How can policy be shaped to minimize human exposure? We need your expertise and insights on this complex issue. I look forward to hearing about the results of this workshop and determining how the IJC can continue to work with you on this important issue. Let me close by extending my best wishes for a very productive workshop.

EXECUTIVE SUMMARY

The International Joint Commission (IJC) and the Commission for Environmental Cooperation (CEC), while acknowledging their distinct mandates and history, have long recognized their common interest in research associated with toxic contaminants, particularly persistent toxic substances.

In the year 1985, the Water Quality Board of the IJC developed a list of 11 Critical Pollutants (all persistent toxic substances) and the Commission began an assessment of the sources, dispersion and effects of these substances in the Great Lakes ecosystem. In the year 1987, with the addition of Annex 15 to the Great Lakes Water Quality Agreement, the Commission delved further into the deposition of these substances from external sources into the basin via transport through the atmosphere.

Shortly after its establishment in the year 1993, the CEC initiated its Sound Management of Chemicals program, focused on many of the same persistent toxic substances designated by the Water Quality Board. In October 1998, reflecting their concern for one of the most broadly dispersed persistent toxic substances, mercury, the two Commissions jointly sponsored a workshop on the State of Scientific Knowledge Related to Mercury. This first workshop brought together scientists from the United States, Canada, and Mexico on a trilateral basis. The scientists explored information on sources, ambient air concentrations, and wet and dry deposition of the various forms of mercury in the context of impacts on human and ecosystem health of that pollutant. The workshop participants concluded that enhancements to source and ecosystemic measurements of mercury and improvements to available atmospheric models were needed. Effective communication between the scientific community and policy makers would also be required to achieve further reductions in anthropogenic emissions.

In pursuit of improved communication, a second joint workshop *Addressing Atmospheric Mercury: Science and Policy* was organized. With support from Environment Canada, prominent scientists and policy makers from the United States, Canada, and Mexico met in Research Triangle Park, North Carolina in December 13-14, 2001 to review the state of mercury science and related policy in the three countries. The 20 presentations and associated panel discussions contained a wealth of scientific detail, much of which is reflected in this document.

Recognizing that, in some forms, mercury is a global pollutant, estimates of worldwide emissions as well as those of the three countries were reviewed. Continental monitoring activities were also reviewed, including determination of wet deposition, concentrations in ambient air, and related dry deposition. The use of these data in global, continental, and regional atmospheric mercury transport models was examined as were measurements of mercury in fish and seafood. While acknowledging the presence of mercury in nature, the need for a rigorous characterization of such sources was emphasized. Participants recognized the lesser but significant global anthropogenic contribution to mercury loading in North America and re-emphasized the need for further routine and specialized monitoring studies of sources, ambient air concentrations, and related wet and dry deposition determinations. Participants also acknowledged the progress made in modeling the transport and deposition of this contaminant.

In assessing the outcome of the meeting, the International Air Quality Advisory Board of the IJC and the Mercury Task Force of the Sound Management of Chemicals, CEC, recommended:

- further reductions in anthropogenic mercury emissions;
- improvements to the quality, comparability and scope of mercury source and ambient measurements, including levels in selected biota;
- enhancement of available appropriate meteorological data;
- continuation of programs in Canada and the United States to measure mercury content in freshwater fish, enhance measurements among marine food species, and support Mexico in the initiation and maintenance of such programs;
- continued model development, with an accounting for global sources, to guide the evolution of control programs and determine their outcome; and
- continued investigation of other possible effects of mercury on human health.

Further enhancement of co-ordination among Canada, the United States and Mexico, with joint technical programs on all aspects of mercury research and policy development was advocated along with interaction with other international and intergovernmental organizations, including the ongoing UNEP global assessment of mercury.

1.0 INTRODUCTION AND BACKGROUND

1.1 The International Joint Commission and the Commission for Environmental Cooperation: Ongoing Interest in Mercury

The International Joint Commission (IJC), largely through its International Air Quality Advisory Board (IAQAB), and the Commission for Environmental Cooperation (CEC), have long recognized their mutual concern with the levels of mercury in the ecosystem. The IJC has studied the issue of mercury since the year 1978, when the renegotiation of the Great Lakes Water Quality Agreement (GLWQA) between the United States and Canada brought a focus on it as one of several persistent toxic substances. In the year 1985, in its report to the Commission, the Water Quality Board designated mercury as one of the Critical Eleven persistent toxic substances in the Great Lakes basin. This list has been subsequently adapted by the governments of the United States and Canada in their Binational Toxics Strategy (April 1997) as a descriptor of the Level 1 substances to be among the first addressed under this Strategy.

Furthermore, Annex 15, added to the Agreement in the year 1987, recognized the atmosphere as a significant pathway for persistent toxic substances including mercury, and outlined the research, surveillance, monitoring and control measures needed to further quantify and reduce such transport to the Great Lakes basin.

In the early 1990s, the IJC struck the Virtual Elimination Task Force to review progress toward the Agreement goal of virtual elimination of the input of persistent toxic substances. The Task Force report, *A Strategy for Virtual Elimination of Persistent Toxic Substances* (year 1993), used mercury as one of the illustrative contaminants in its considerations. The presence of natural sources of mercury was acknowledged, while stating that “much of the chemical released from the soil has been deposited as a result of previous anthropogenic activity.” The report reviewed several sources of mercury associated with human activity and made specific recommendations as to their further prevention, control and elimination. The ultimate goal was restated as the virtual elimination of anthropogenic inputs of mercury into the Great Lakes basin, an approach that was embraced by the governments of the United States and Canada in their Binational Toxics Strategy of April 1997.

Under IJC’s 1995–1997 Great Lakes priorities, the IAQAB attempted an assessment of government efforts under Annex 15 toward immediate and forecasted reductions of emissions of persistent toxic substances from identified major sources. In another activity, the IAQAB also reviewed emissions of persistent toxic substances, including mercury, from mu-

nicipal solid waste incinerators and drafted an incineration policy statement that was subsequently adopted by the IJC. Also the North American Commission for Environmental Cooperation was established in the year 1993 under the North American Agreement on Environmental Cooperation among the governments of Canada, the United Mexican States, and the United States, developed as one of several outcomes of the Free Trade negotiations.

Since the year 1995, in accordance with Council Resolution 95-05 and under the Sound Management of Chemicals (SMOC) program, the CEC has fostered development of North American Regional Action Plans (NARAPs) for selected toxic chemicals that persist and accumulate in the environment and are transported across national boundaries via air and water pathways and in traded products.

In the year 1997, under Phase I of the Mercury NARAP, the three countries individually and cooperatively advocated a reduction in the extent of exposure of North American ecosystems to this contaminant. The emphasis was placed on the protection of fish and wildlife, and particularly humans, through the targeting of specific sources of anthropogenic mercury pollution for further control.

Under the Mercury NARAP Phase II, goals include the adoption of “best practices” across North America to prevent and reduce mercury releases from human activities to achieve naturally occurring levels in the environment, development of stakeholder partnerships to formulate interim targets for prevention and reduction of releases and to improve the scientific understanding of mercury, and recognition of the need to prevent or minimize releases of mercury used in regional commerce. Building Mexico’s capacity to further determine and control mercury releases, and the eventual dissemination of the NARAP experience in further cooperative work with other countries in Latin America and the Caribbean were fundamental parts of the Plan. More detailed information about the CEC’s NARAP on mercury and other NARAPs can be found on the CEC website at <http://www.cec.org/>.

Key to achieving the goals of the CEC’s North American Regional Action Plan on Mercury is enhancement of the tri-lateral capacity to measure, track, and monitor mercury uses and emissions. Such data are necessary to assess the impacts of mercury and support the evolution of appropriate policy measures.

1.1.1 First Science Experts Workshop on Mercury, October 1998, Las Vegas, Nevada

Sharing similar concerns, the two Commissions jointly sponsored a three-day workshop on the State of Scientific Knowledge Related to Mercury at the National Exposure Research Laboratory of the U.S. Environmental Protection

Agency (USEPA), Las Vegas, Nevada, October 6-8, 1998. Its purpose was to discuss, among a trilateral group of scientists, the current state of knowledge pertaining to mercury and to consider how research and monitoring could be applied on a trinational basis (Canada, United States, and Mexico) within the framework of the CEC North American Regional Action Plan (NARAP) on Mercury. The measurement of progress toward achieving the NARAP and the development of control strategies for significant sources of anthropogenic mercury were also considered.

The workshop drew, in part, on two consultative meetings organized by the CEC, the first in Zacatecas, Mexico, a site contaminated with mercury from colonial Spanish mining for silver, and the second in Mexico City to assess the private, public, and institutional concerns regarding mercury. The benefits of a trinational assessment by scientists and policy makers were important not only to the CEC and IJC efforts to identify and mitigate any adverse impacts of this contaminant, particularly those associated with long-range atmospheric transport, but also for national and multilateral agencies concerned with mercury contamination.

The focus of the meeting was on releases of mercury into the environment due to human activities. The contribution of natural sources, to the extent that they could be rigorously defined, was to be assessed as part of a clarification of the mercury inventory; however, no discussions on management of naturally occurring sources of mercury were planned.

The workshop was to seek recommendations on:

- science-based approaches to the reduction and prevention of releases of mercury.
- developing a trinational monitoring, research and development program.
- identifying candidate control strategies for significant sources and source sectors.

More than 60 experts from North America and Europe considered the following topics:

- the current state of knowledge on the fate and transport of mercury in environmental media.
- health effects of mercury on humans and wildlife.
- speciation of atmospheric emissions from major source sectors.
- monitoring and control technology, status and needs.
- sources or source segments with greatest reduction potential.
- management of current and accumulating stocks of mercury.

Critical Issues Raised at the Workshop

1. Human Health Effects

- a. Most severe impacts are usually manifest in the fetus and very young children. There may be a significant latency period before impacts become apparent.
- b. In assessing the Seychelles and Faeroe Islands studies, the latter appears to have been more sensitive to the detection of impacts, with a greater focus on children.
- c. Concentrations of methylmercury in some northern Canadian aboriginals are already well into the range where sub-clinical symptoms can be anticipated.
- d. The United States National Academy of Science findings on health impacts of methylmercury have the potential to strongly influence regulatory direction.

2. Wildlife Effects

- a. Risk assessment methodologies differ significantly between Canada and the United States. Generally, Canada assesses concentrations in wildlife and consequent human consumption (where appropriate) while the United States assesses concentrations in water which could lead to negative impacts.
- b. Some eastern Canadian loons, a fish predator species and a significant indicator of mercury-related stress, appear to have body burdens in excess of the no-effect threshold level.
- c. Mink and otters in some segments of Canada appear to have high mercury levels which may be associated with a significant decline in populations greater than five years of age.

3. Mercury Emissions Characterizations

- a. Considerable uncertainty prevails regarding the accuracy and comprehensiveness of any atmospheric emissions inventory in North America.
- b. Annual mercury emissions on a grams-per-capita basis were estimated as:

i.	Canada	1.40
ii.	USA	0.88
iii.	Mexico	1.10

c. Year 1990 Emissions from Major Anthropogenic Sources (tonnes (~tons))

Source Category	Global	North America
Stationary Combustion	1475 (1626)	105 (116)
Base Metal Production	394 (434)	25 (28)
Iron and Steel Production	28 (31)	5 (6)
Cement Production	115 (127)	13 (14)
Waste Disposal	139 (153)	66 (73)
Chlor-alkali process	172 (190)	4 (5)
Total	2323 (2561)	218 (240)

- d. Most U.S. urban areas demonstrate relatively high emissions of Hg²⁺ and particulate-bound Hg species.
- e. Dry deposition of mercury from the atmosphere may be at least as important as wet deposition in lakes and lake basins.
- f. While there will likely be significant variation from lake to lake, 40 percent (40%) of the mercury deposited into the Great Lakes basin may be from external sources.
- g. Mercury emissions from landfill sites may contribute significantly to both aquatic and atmospheric inventories. Mexican inventories of mercury emissions may be affected by open burning of waste.

4. Control Technologies

- a. Non-technological methods need to be addressed, including

i.	Energy conservation (reduction in generation of electricity from coal-fired generating stations)
ii.	Source segregation
iii.	Product bans

- b. Carbon absorption appears to be a proven control technology for incinerators but remains under development for coal-fired power plants
- c. Mercury speciated inventories are necessary to provide guidance on appropriate control technologies.
- d. Mercury emissions from waste incineration are 85 percent Hg²⁺ and 15 percent Hg⁰. Eighty percent (80%) reductions have been demonstrated to be feasible for this sector.
- e. A significant portion of the mercury content in the feed to current municipal waste incinerators comes from yard wastes as a result of atmospheric deposition.
- f. Coal-fired utilities need to develop a multipollutant strategy to maximize the benefit of mercury reductions;

flue gas desulfurization with subsequent filtration shows promise as one such multipollutant effort.

5. Atmospheric Fate and Transport

- a. Mercury circulates globally via the atmosphere; the elemental form has a residence time aloft of about one year.
- b. An Arctic depletion anomaly needs to be studied further to determine possible impact on biota and human inhabitants.
- c. Emissions and re-emissions of both natural and anthropogenic sources need to be differentiated and quantified.
- d. In some locations, the air/water exchange of mercury may indicate a net sink for mercury deposited from the atmosphere.

1.1.2 Impact of the Science Experts Workshop on Mercury

As a result of the discussions and workshop consultations noted above, a considerable number of recommendations were made that ultimately contributed to the development of the CEC Phase II North American Regional Action Plan (NARAP) on Mercury signed by the Environment Ministers of Canada, the United States, and Mexico in June 2000. This NARAP, as well as NARAPs for chlordane, PCBs and DDT, is available for viewing at <http://www.cec.org>, under the title of *Pollutants and Health*. Subsequent to the Science Experts meeting, it was determined that Monitoring/Assessment and Research were such critical aspects of this and similar work on other contaminants that the CEC initiated a separate action plan, to be known as the NARAP on Monitoring and Assessment.

The Science Experts Workshop also advocated a continued dialogue among the relevant agencies and scientific communities of the three countries. Collaborative discussions and support for further mercury-based study were seen as beneficial not only to the three North American countries but also as an influence on the actions of other nations toward reductions in anthropogenic releases.

Since that event, the scientific research and policy considerations regarding mercury have grown substantially. The need for an event where policy makers and scientists could review the science pertaining to mercury and its implications for policy development was most apparent. As a result the CEC and the IJC, through the IAQAB and with the support of Environment Canada, agreed to jointly sponsor this workshop *Addressing Atmospheric Mercury: Science and Policy*, held December 13-14, 2001 in Research Triangle Park, North Carolina.

TABLE 1. Selected Physical/Chemical Properties of Various Species of Mercury (tabular data from Schroeder and Munthe, 1998)

Property	Hg ⁰	HgCl ₂	HgO	HgS	CH ₃ HgCl	(CH ₃) ₂ Hg
Melting Point (°C)	-39	277	decomp. @ 500	584 (sublim.)	167 (sublim.)	?
Boiling Point (°C)	357 @ 1 atm	303 @ 1 atm	—	—	—	96 @ 1 atm
Vapor Pressure (Pa)	0.180 † @ 20° C	8.99 x 10 ⁻³ ‡ @ 20° C	9.20 x 10 ⁻¹² @ 25° C	?	1.76 @ 25° C	8.30 x 10 ⁻³ @ 25° C
Water Solubility (g L ⁻¹)	49.4 x 10 ⁻⁶ @ 20° C	66 @ 20° C	5.3 x 10 ⁻² @ 25° C	~2 x 10 ⁻²⁴ @ 25° C	~5-6 @ 25° C	2.95 @ 25° C

† Implies a saturation air concentration of about 14 mg per cubic meter at 1 atmosphere

‡ Implies a saturation air concentration of about 1 mg per cubic meter at 1 atmosphere

This second event focused on the interaction of science and policy in addressing atmospheric mercury. The objectives of the workshop were to:

- Review current developments in source and ambient monitoring of mercury
- Examine the transport and fate of mercury in ecosystems
- Identify the policy implications arising from current scientific research

During the workshop, presentations from several researchers and modelers were followed by panel discussions considering the policy implications of their work. Invited scientists and policy makers from the United States, Canada, and Mexico, as well as European experts, presented current scientific findings and analysis and examined associated policy implications as they pertain to mercury.

The conclusions and recommendations from this latter workshop should prove instrumental in advancing the objectives of the NARAP under the CEC and the continuing effort of the IJC in advocating implementation of the virtual elimination goal under the Great Lakes Water Quality Agreement.

1.2 Mercury: Physical and Chemical Properties

Mercury is a toxic metal occurring naturally in the environment in several forms. The physical properties associated with elemental mercury and selected mercury compounds are given in **Table 1**.

The most important distinction regarding the presence of mercury in the environment is between the inorganic and organic forms.

1. Inorganic mercury:

Inorganic mercury comprises the metallic or elemental form of mercury (Hg(0)), the ionic form (Hg⁺), and related inorganic mercury compounds. Elemental mercury is insoluble, can exist in a rather inert gaseous

form at low concentrations and is widely distributed throughout the global atmosphere.

In the ionic form, mercury can exist in two oxidation states: Hg+1 (the mercurous ion), and Hg+2 (mercuric ion). Oxidized mercury (Hg(II)) is relatively soluble and has a tendency to associate with particles. When ionic mercury is combined with other elements, such as oxygen or chlorine, it forms other inorganic mercury compounds.

2. Organic mercury:

Methylmercury, dimethylmercury, phenylmercury, and thimerosal are organomercurial compounds resulting from a chemical bond between mercury and carbon. When such a bond is created, mercury is commonly referred to as "organic." Methylmercury (MeHg) is the most toxic and prevalent form of these compounds and is largely responsible for the fish advisories in place in thousands of the large and small waterbodies in the United States and Canada. Consumption of contaminated fish is the major route of human exposure to this toxic substance.

The specific state (solid, liquid, or gas) and form of mercury compounds present can vary depending on the biological, chemical and physical conditions in the environment (such as temperature, acidity, microbial activity).

1.3 Speciation

In his presentation, Russ Bullock of the USEPA presented several primary factors influencing the transport range of mercury emissions in the atmosphere. These include the chemical and physical forms of mercury emissions, emission plume characteristics (temperature, velocity, moisture content), subsequent chemical and physical reactions, including those taking place within cloud formations, and surface dry deposition characteristics. Because of the several forms mercury can take in the environment and the unique behavioral characteristics of each form, the identification of individual species is crucial.

TABLE 2. Typical ambient air concentrations of mercury species (Misra)

	Concentration Ng/m ³	Henry's Constant	Temporal Scale
Elementary Mercury: – Hg ⁰	1 – 3	0.3	Global Lifetime: Months to a year
Divalent Mercury: – HgCl ₂ – HgO	0 – 0.1 ?	4 x 10 ⁻⁵ 4 x 10 ⁻⁵	Local/Regional Lifetime: hours to a day
Particulate Mercury: – Hg(p)	0.02 – 0.1	–	Regional Lifetime: 1 – 3 days

Mercury is emitted from anthropogenic activities such as incineration, coal combustion, and metallurgical refining into the atmosphere in three principal forms: elemental vapor (Hg(0)), gaseous divalent mercury Hg(II) and particulate phase mercury (Hg(p)). The major chemical form of mercury emitted from anthropogenic sources is elemental in the vapor state at source. The remainder of atmospheric mercury is mostly associated or absorbed with particles, aerosols and Hg(II). During his presentation, Dr. Walcek reviewed the speciation ratio used in his model: Hg(0) represented 48 percent; Hg(II) - 35 percent and Hg(p) - 17 percent. These are only estimates, but they appear to be a reasonable first estimate of mercury speciation at source. The various forms of mercury and their typical atmospheric concentrations and lifetimes are given in **Table 2**.

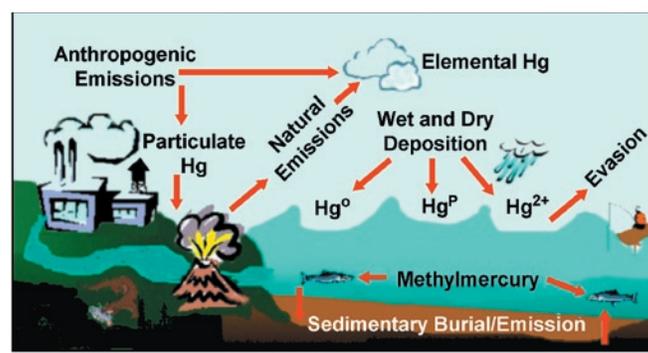
Given its relatively inert characteristics and low water solubility, elemental mercury vapor has a residence time of approximately one year in the atmosphere. As a result it can be transported for long distances before wet and dry deposition processes return it to land and water surfaces from where, on occasion, it can be volatilized and once again transported in the atmosphere. Dry deposition refers to the transfer of mercury in the form of gaseous or particulate species from the air to surfaces (water or land) outside of precipitation events. Wet deposition implies wet scavenging by precipitation events, in which cloud chemistry is a significant factor.

The gaseous divalent form of mercury (Hg(II)) is the result of oxidation of elemental mercury vapor. The most important gas phase oxidation pathways in the atmosphere are reactions with ozone and OH radicals (*Ref. 1*). Hg(II) is less volatile than Hg(0), and thus has a shorter residence time in the atmosphere and tends to condense onto atmospheric particulate matter or be deposited to marine or terrestrial surfaces on a local or regional basis. Different species of Hg(II) exist, among them reactive gaseous mercury (RGM), chemically reactive gaseous compounds of mercury that are quickly deposited to the surface by wet and dry processes. Particulate mercury (Hg(p)) is also subject to rapid wet and dry deposition and, along with Hg(II), accounts for most of the regional or local deposition.

Mercury transformation in emission plumes is still not well understood but several mechanisms play a significant role in the dispersion of the pollutant in the environment. Mercury speciation in plumes is subject to a wide range of factors. For

instance, in emissions from coal-fired utility boilers, oxidized mercury typically ranges from 30 to 70 percent of the total mercury in the flue gas but this concentration depends on the amount of mercury in coal and the manner of combustion of that coal (*Ref. 2*). Distance of transport and ultimate fate of mercury in ecosystems are closely correlated with mercury's speciation in emission plumes and the extent of subsequent transformation. Information about chemical speciation of mercury is critical for modelers attempting to simulate the transport and fate of mercury in the environment.

Once deposited in a water body, inorganic mercury must be converted to an organic form, principally methylmercury

FIGURE 1. Simplified Mercury Cycle

from EPS DOE

(MeHg) before it can be accumulated in fish and other biota. Consumption of contaminated fish is the main non-occupational route of mercury exposure for humans. Particulate-bound mercury can be transformed and mobilized by biotic and abiotic oxidation and reduction and can be converted to insoluble mercury compounds and precipitated. This transformation of mercuric mercury into metallic mercury in aqueous systems is enhanced by light and occurs under both aerobic and anaerobic conditions (*Ref. 3*). Inorganic mercury can also be methylated by microorganisms indigenous to soil and fresh water under both aerobic or/and anaerobic conditions. Transformation of methylmercury compounds back to volatile elemental mercury may also occur as a result of microbial demethylation. Anaerobic conditions, as may be found in sediments, favor the demethylation of methylmercury (*Ref. 4*).

Figure 1 gives some sense of the complexity of the biogeochemical cycle of mercury in the environment, highlighting the speciation phenomena in the atmosphere.

1.4 Concerns for Human Health

Dr. Marc Lucotte from the University du Québec à Montréal opened the workshop by highlighting the effects of mercury on human health as determined in his studies on fish consuming populations living adjacent to portions of the Amazon River in South America.

His studies revealed widespread contamination of the Tapajós River in northwest Brazil. High concentrations of mercury were found in river sediments. However, the contamination did not appear to be strongly associated with mercury used in gold mining but rather with that released from soils due to deforestation. Indeed, there was essentially no difference in mercury exposure levels between villagers living 100 kilometers (~ 62 miles) downstream from the gold-mining area and those residing 300 kilometers (~ 186 miles) away. He suggested that deposition of mercury has occurred over decades and centuries onto the forest canopy, but with the clear-cutting of the forests, this accumulated mercury has been leached out of the soil, into the water-courses and into the food chain.

Dr. Lucotte examined mercury concentrations in the hair of indigenous peoples whose diet was composed largely of fish. The data demonstrated that a high fish diet coincided with a high concentration of mercury in the hair of certain subjects. High concentrations of mercury in the hair were co-incident with a reduction of the subject's field of vision. This response occurred with concentrations as low as 10 ppm in the hair; a decreasing ability to discern colors was also observed. In addition, simple tests indicated that the dexterity of such subjects was also impaired and this impairment increased with increased levels of mercury in hair. Other studies have shown that similar low doses affect learning and cognitive abilities, as well as muscle coordination, particularly in young children.

Lucotte pointed out that it is important to understand and manage the linkages among the mercury levels in various fish in a given aquatic system and their implications for fish consumption habits. His study noted a significant difference in mercury levels in herbivorous and largely piscivorous fish with the latter having higher mercury content. By advising the local population in this region on appropriate fish consumption, a 30 percent decrease in hair mercury concentration was seen over a five year period.

2.0 SOURCES AND PATHWAYS OF MERCURY

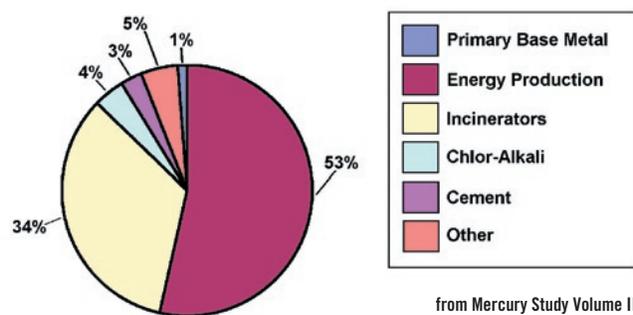
2.1 Sources

2.1.1 Overview

Mercury can be emitted into the atmosphere from natural or anthropogenic (associated with human activity) sources. Natural sources are mainly associated with volcanic emissions, volatilization from marine and aquatic environments, releases associated with wind-blown dust and the weathering of rock formations. Anthropogenic releases can be due to intentional or incidental human use and are associated with both point sources or diffuse areal releases.

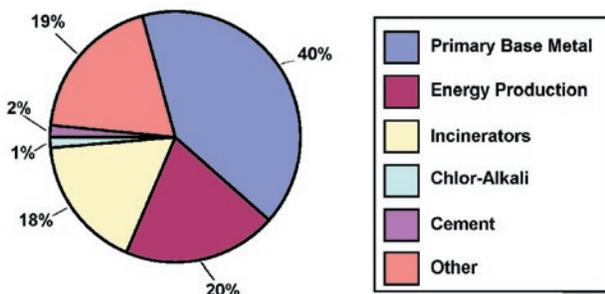
In North America combustion point sources are the largest source of mercury to the atmosphere, with stationary combustion of fuels being the main contributor. There are four significant categories of combustion sources: municipal and medical waste incineration, coal-fired electrical utilities, commercial and industrial boilers, and metallurgical processes. The coal-fired electric utilities are currently the principal contributor of atmospheric mercury emissions in the United States. In Canada emissions are dominated by primary metal production (Figures 2 and 3).

FIGURE 2. U.S. Mercury Emissions by Sector, 1994-1995



from Mercury Study Volume II
USEPA, December 1997

FIGURE 3. Canadian Mercury Emissions by Sector - 1995



from DOE - submission to UNEP
Hg Assessment - September 2001

Incidental use of mercury describes the release in the environment of mercury when other raw feed material is being processed and mercury does not play a substantive role in the process. Energy production (utilities) and manufacturing are considered activities associated with incidental releases of mercury. Intentional use of mercury in production processes or consumer products result in mercury emissions when such products or byproducts are ultimately managed as waste (Ref. 5). Thus, incinerators and waste disposal facilities are considered as contributing to intentional releases.

2.1.2 Estimated Global Emission of Mercury

During the workshop, Luke Trip, of the Environmental Protection Service of Environment Canada, presented a global overview of the emission sources of mercury, based on estimates prepared by J.M. Pacyna and E.G. Pacyna. These estimates were based on the collection of emission data at a country level; in cases where no official estimates were available, they were determined on the basis of emissions factors and statistical data on the production of industrial goods and/or the consumption of raw materials. National estimates were provided from 17 countries and checked for completeness and comparability. The authors noted that it was very difficult to entirely verify the data obtained.

During the year 1995 the total mercury released to the atmosphere from anthropogenic sources was estimated to be approximately 1,900 tonnes (~ 2,094 tons), compared to about 2,100 tonnes (~ 2,314 tons) during the year 1990. These data suggest that there has not been a significant change in total mercury released annually to the atmosphere over that five-year period. The authors acknowledged that the year 1995 data do not contain emissions of mercury from gold production processes because of the highly speculative nature of such releases. These can however be substantial and it is thought that as much as 325 tonnes (~ 358 tons) can be emitted from this process annually, half of it from Africa. While the total emissions did not significantly change, the authors nevertheless observed a change in the dominant sources and their geographic locations.

In the year 1995, stationary combustion of fuel represented 77 percent of total emissions of mercury, an increase of 17 percent since the year 1990 (Table 3), due to the increased combustion of coal to produce electricity and heat over the five year period. The use of mercury in battery production and chlor-alkali cell processes to produce chlorine gas has significantly decreased. No major changes in the emissions of mercury from pig iron and steel production have been observed between the years 1990 and 1995. With the exception of uses in gold refining, noted above, emissions of mercury resulting directly from its various applications or uses were believed to be insignificant on a global scale in the year 1995 although its presence in the waste stream resulted in significant releases.

TABLE 3. Global Emissions in tonnes (~tons) of Total Mercury from major Anthropogenic Sources in the year 1995 - (Pacyna & Pacyna)

Continent	Stationary combustion	Non-ferrous metal production	Pig Iron and steel production	Cement production	Waste disposal	Total	%
Europe	185.5 (204)	15.4 (17)	10.2 (11)	26.2 (29)	12.4 (14)	249.7 (275)	13.1
Africa	197 (217)	7.9 (9)	0.5 (0.6)	5.2 (6)		210.6 (232)	11
Asia	860.4 (948)	87.4 (96)	12.1 (13)	81.8 (90)	32.6 (36)	1074.3 (1184)	56.1
North America	104.8 (116)	25.1 (28)	4.6 (5)	12.9 (13)	66.1 (73)	213.5 (235)	11.2
South America	26.9 (30)	25.4 (28)	1.4 (2)	5.5 (6)		59.2 (65)	3.1
Australia & Oceania	99.9 (110)	4.4 (5)	0.3 (0.3)	0.8 (0.9)	0.1 (0.1)	105.5 (116)	5.5
TOTAL The year 1995	1474.5 (1625)	165.6 (183)	29.1 (32)	132.4 (146)	111.2 (123)	1912.8 (2108)	
TOTAL The year 1990 *1	1295.1 (1428)	394.4 (435)	28.4 (31)	114.5 (126)	139 (153)	2143.1 *2 (2362)	

*1. Estimates of maximum values, which are regarded as close to the best estimate value.

*2. The total emission estimates for the year 1990 include also 171.1 tonnes (189 tons) of mercury emission from chlor-alkali production and other less significant sources.

There have been two major changes with respect to the geographic distribution of major emission regions:

- A decrease in mercury emissions in Europe and North America. This can be explained by pollution prevention activities, further installation of emission and other control equipment and procedures, and a decrease of emissions from combustion sources, mostly in Europe. In Central Europe this decrease appears to be correlated to a decrease in industrial activities, but this statement needs to be verified.
- The Asian contribution to global emissions of total mercury has increased by over 25 percent, primarily due to the increase in combustion of coal in China. In fact, Asia now dominates the emissions from stationary combustion sources. **Figure 4** indicates that, along with South Africa, Asia contains areas with among the highest total mercury emissions (from all sources) in the world.

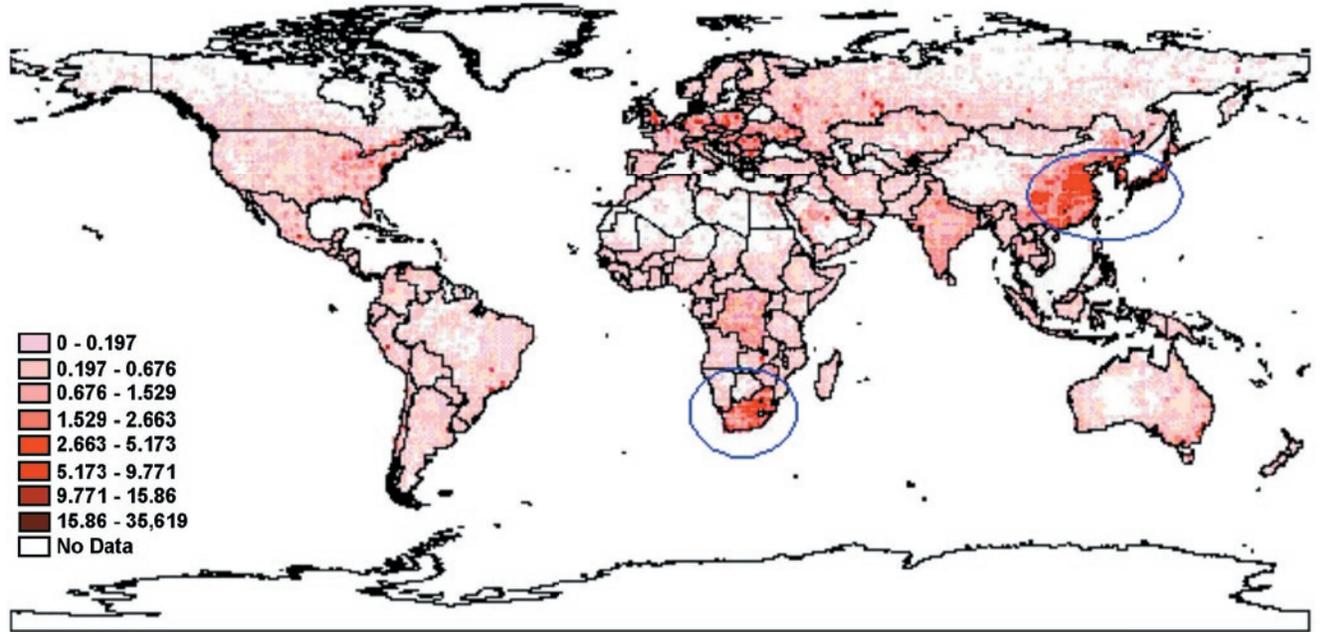
2.1.3 United States

Mercury is used in industrial processes because of its distinctive physio-chemical properties (i.e. conducts electricity, acts as a biocide, is useful in the measurement of temperature and pressure, and forms alloys with almost all other metals). Mercury is widely used in metallurgy, manufacturing and dentistry, with chlorine production as the major user of mercury in the United States. The annual U.S. demand for mercury has decreased from 554 tonnes (~ 611 tons) in the year 1991 to 436 tonnes (~ 481 tons) in the year 1995 (*Ref. 6*). The most significant changes in reported mercury consumption are the dramatic reduction in use in paints and batteries. In addition, since the 1980s and 1990s there have been significant reductions in the use of mercury in laboratories, wiring devices and switches, and measuring and control instruments.

Mercury mining facilities are no longer in operation in the United States; the last mine (in Nevada) was closed in the year 1990. Primary production mainly comes as a byproduct of gold mining. In the year 1995, the total U.S. commercially available supply of mercury was 911 tonnes (~ 1004 tons), with around 41 percent resulting from imports. Exports of mercury have decreased by more than 80 percent in the last several years, from 977 tonnes (~ 1077 tons) to 179 tonnes (~ 197 tons) due to the suspension of sale of mercury from U.S. federal stockpiles (*Ref. 7*). In the year 1994 the government stopped selling stockpiled mercury to U.S. and foreign companies because of concerns raised by EPA regarding accumulation of mercury in the atmosphere. The Water Quality Board of the International Joint Commission in its 1995-1997 Priorities and Progress Report shared the same concern, arguing that federal sale of mercury would increase the world supply, thereby lowering price and increasing use, and recommended that sales be halted. Stockpiled excess mercury and mercury waste remain an important issue in the United States. Currently 4,408 metric tons (~ 4,859 tons) of excess mercury are located in four Defense National Stock Pile centers across the United States (*Ref. 8*).

Anne Pope of the Emissions, Monitoring and Analysis Division of the USEPA reviewed the years 1996 and 1999 inventory of sources of mercury to the atmosphere in the United States. She emphasized that modelers using emission estimates in their models should first understand how the data were compiled. She then presented the methodology behind the emission estimates. The U.S. National Emission Inventory (NEI) is an annual national repository of emissions data for the 188 hazardous air pollutants (HAPs) and Criteria air pollutants identified in the U.S. Clean Air Act. It is assembled by the USEPA to support policy making and regulatory impact studies and will be available to the public on the EPA website in the summer of 2002, when it will replace the National Toxic Inventory (NTI).

FIGURE 4. Global Emissions of Mercury (Total Hg 1995 - All sources (tonnes))



(Pacyna NILU, Norway)

The current inventory includes both stationary sources and mobile sources. Stationary sources of HAPs include both major point and area designations. Major sources, as defined in the Clean Air Act, are facilities that have the potential to emit 10 tons per year of a single HAP or 25 tons of multiple HAPs. Area sources of HAPs are defined as those facilities that have the potential to emit less than 10 tons (nine tonnes) of a single HAP or 25 tons (22.5 tonnes) per year of a combination of HAPs. For HAPs, all major sources are inventoried as point sources; area sources are inventoried either as point or nonpoint sources depending on the availability of facility-specific information. Nonpoint and mobile source data are aggregated at the county level. Point sources or facility entries are allocated at the individual stack level (Ref. 9).

The NEI blends and merges different sources of data, beginning with available EPA data, then information from state and local agencies, which represent 90 percent of the data set. If necessary, Toxic Release Inventory (TRI) data are used but their use is minimal, representing only one percent of the data set. TRI is not a suitable input for models because it does not include all the sources (13,000 facilities are reported instead of the 19,000 reported through EPA and the States and local agencies). There are concerns about the quality of emission inventories for individual sources in the TRI databases as well, as it is largely self-reported information.

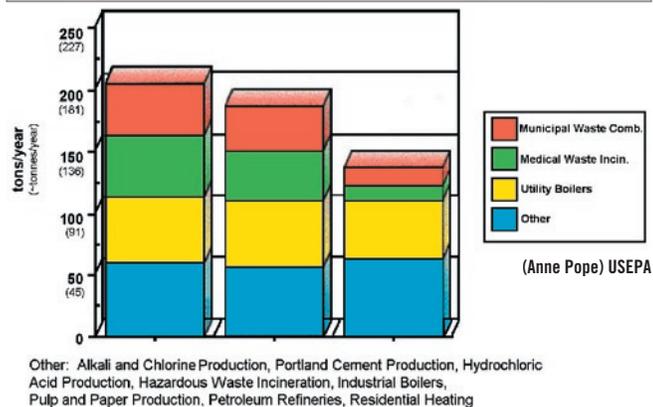
The final version of the year 1999 NEI currently under preparation is to contain estimates of process level emissions within a facility and the source-specific parameters necessary for modeling, such as precise location and detailed emission data. Location data and emission parameters are

crucial for an accurate inventory and this segment of the inventory still needs improvement. In the year 1999, 10 percent of data did not include accurate information. Location errors, including incorrect or missing latitude/longitude and incorrect or missing county Federal Information Processing Standards (FIPS) codes, require resolution for modeling. Obviously if the location of a facility is reported incorrectly or is missing, estimating release, transport, exposures and ultimately risks with any accuracy is very difficult (Ref. 10).

The data presented by Anne Pope show a decrease in emissions of mercury largely due to the introduction of controls on municipal waste combustion and medical waste incineration from the years 1990 to 1999. This can be attributed to more stringent emission standards and improved sorting and control at such facilities. However, mercury emission maps of the United States continue to show high levels in some individual counties due to current municipal waste combustion. This trend is especially apparent in Florida, the West Coast and the Great Lakes region. Furthermore, utility boilers do not follow a decreasing trend; there is only a four ton difference between the years 1990 and 1999 inventory for this category (Figure 5).

The issue of speciation of mercury in the inventory was also addressed. It was noted that very little testing data are available on emissions of elemental gaseous, gaseous divalent and particulate divalent species of mercury except for coal-fired electric utility boilers. For utility coal boilers, the type of coal influences speciation. While individual sources will vary, generally ionic mercury represents 40 percent of the emissions of utility boilers, particulate less than five per-

FIGURE 5. National Emissions Trends, Preliminary Data - 1990, 1995, 1999



cent and elemental 55 percent. Table 4 shows the emissions profiles of mercury releases, illustrating the different speciations possible, depending on the source.

Ms. Pope emphasized the need for improved measurements of both speciation and associated stack parameters. Modelers need to recognize and consider the uncertainty of inventories when using data in models. Furthermore, the diurnal and monthly variation of mercury emissions need more study. Also the inventories are only for anthropogenic sources and do not estimate emissions from natural sources, which could be comparable or greater. Other sources estimate that global natural emission rates are in the vicinity of 1,000 tonnes/year (~ 1,102 tons/year) (Ref. 11). Although these estimates are highly uncertain, it appears that natural emissions could account for about 50 percent of the total global emissions.

In a recent study (Ref. 12), the relative contribution of natural and anthropogenic sources to the deposition of atmospheric mercury over several centuries was examined. A mercury deposition record over the last 270 years was established from ice cores collected in Wyoming at the Upper Fremont Glacier, which allowed estimation of the impact of atmospheric releases of mercury from both natural and anthropogenic regional and global sources.

Preindustrial (before the year 1840) measurements of mercury were used to extrapolate a background value of three nanograms per litre (3 ng/L) throughout the ice-core record. Since that time, anthropogenic inputs were associated with 52 percent of the total deposition, while background concentrations contributed to 42 percent of the total mercury in the ice core, with volcanic events (including three major eruptions at Tambora, Krakatau, and Mount St Helens) contributing a further six percent of the total. These values are in agreement with the aforementioned estimate of global natural emissions accounting for about 50 percent of total emissions; however, the study notes that, in the past 100 year period, the estimated contribution to mercury deposition from anthropogenic sources was 70 percent.

2.1.4 Canada

In Canada, mercury is not commercially produced; the last Canadian mercury mine closed in the year 1975. Demand for mercury is largely met by imports from the United States and was estimated at 2.8 tonnes (~ 3.08 tons) in the year 1999 (Ref. 13). The major uses of mercury are in electrical apparatus, industrial applications, and control instruments. The use of mercury in the electrolytic preparation of chlorine is less widespread than in the United States. While there were 15 active chlor-alkali plants in Canada in the 1970s, only one now currently remains in operation. Consumption for applications such as gold recovery, industrial chemicals, and paints and pigments has been phased out.

Marc Deslauriers of the Pollution Data Branch of Environment Canada discussed the methodology behind the Canadian mercury emission inventory. Data estimates are assembled from a number of sources, including the National Pollutant Release Inventory (NPRI, 73 percent of data, based on mandatory reporting), the Criteria Air Contaminants emissions inventory (CAC, 16 percent of data), Accelerated Reduction/Elimination of Toxics Program (ARET, five percent (5%) of data, voluntary reporting), industry supplied estimates (five percent (5%)) and further consultation with the industry (one percent (1%)).

TABLE 4. Emission Profiles of Mercury Releases – tons per year 1999 (Anne Pope USEPA, 1999 U.S. NEI Draft)

	Ionic (tpy) Ionic (%)	Particulate (tpy) Particulate (%)	Elemental (tpy) Elemental (%)	TOTAL
Coal-Fired Electric Utility Boilers	20.4 40%	1.5 <5%	26.1 5%	48
Municipal Waste Combustors	8.8 58%	3.0 20%	3.3 22%	15.1
Medical Waste Incinerators	9.3 75%	2.5 20%	0.6 5%	12.4
Other	12.5 20%	8 13%	41 67%	61.5
Total	51 37%	15 11%	71 52%	137

The Canadian National Pollutant Release Inventory (NPRI), created in the year 1992, is a multimedia database reporting on releases to water, air, and land (solid waste) of over 260 pollutants. Significant changes were made to the NPRI reporting requirements for the year 2000. In previous years, a facility was exempt from reporting to the NPRI if its employees worked less than 20,000 hours during the reporting year (equivalent to 10 full-time employees) (Ref. 14). Also, very limited, if any, mercury data were being reported at the original 10-tonnes (~ 11-tons) and one percent (1%) concentration reporting threshold.

Environment Canada has removed the 20,000-hour employee threshold as well as the one percent concentration exemption for mercury. Manufacturers processing or otherwise releasing five kg (~ 11 lbs) of mercury annually are now subject to reporting, with an exemption for dentists. As a result the data set has undergone some changes:

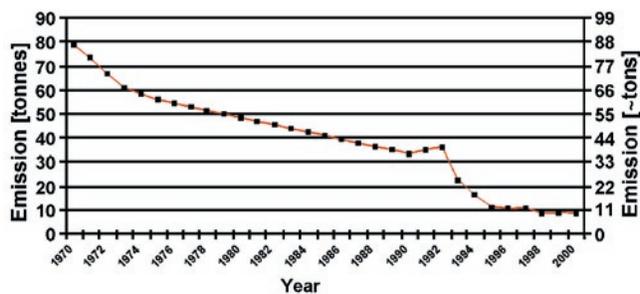
- 1000 percent increase in facilities reporting mercury releases (from 18 to 199)
- 350 percent increase in total releases reported (from 1.98 to 8.9 tonnes (~2.19 to 9.8 tons))
- 230 percent increase in atmospheric releases reported (from 1.76 to 5.82 tonnes (~ 1.94 to 6.42 tons))

Lower thresholds mean better data for specific facilities. In the new inventory, facilities must report emissions from their major stacks (stack height greater than 50 meters (~ 54.5 yds.)). The obligation to report improves the quality of stack parameters, which is especially critical for any modeling effort. With the new thresholds, NPRI now contains more information on air and land emission but water releases require further examination. NPRI year 2000 information accounts for 73 percent of the air releases, compared to 21 percent in the year 1995. The new inventory is the most comprehensive to date and is used by most modelers.

Deslauriers also presented trends regarding mercury emissions in Canada. Since the year 1970, emissions have been reduced by 90 percent (Figure 6). Major reductions have been achieved in:

- Base Metal Smelting (98 percent or 27 tonnes (~ 30 tons))
- Chlor-alkali Industry (99 percent or 23.9 tonnes (~ 26 tons) - largely through closure of facilities)

FIGURE 6. Trends in Mercury Emissions - Canada - 1970 to 2000



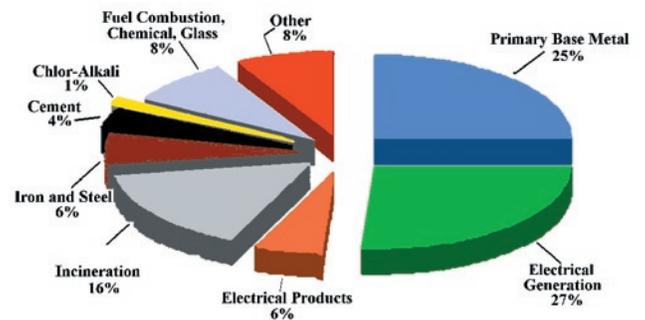
(Deslauriers) Environment Canada

- Removal from Agriculture/Household Fungicides (100 percent or 6.5 tonnes (~ 7 tons))
- Incineration Sectors (80 percent or three tonnes (~ 3.3 tons))

Technological changes in the base metal smelting sector are a significant factor in the reduction of emissions. Smaller reductions were also achieved in many applications such as pharmaceutical products and fluorescent lamp manufacture.

Because of the observed reduction in the base metal smelting sector, power generation now has more relative importance in the emission inventory, accounting for 27 percent of the annual estimations (Figure 7). When asked if the decrease in mercury emissions in Canada over the past years was similar to the trend in the United States, Deslauriers pointed out that emissions have decreased in Canada largely due to reductions from mining and smelting activities. In the United States, the decreases have been largely associated with further control of municipal and medical waste incinerators (90 percent reduction). The main distinction between the United States and Canada is that emissions from the coal-fired utility sector have been relatively stable in the United States.

FIGURE 7. Preliminary Mercury Emissions in Canada (2002)



(Deslauriers) Environment Canada

Deslauriers envisioned additional reductions through the Canada Wide Standard (CWS) and the Multi-Pollutant Emission Reduction Strategies (MERS) under development by the Canadian government. The CWS addresses two specific sources of mercury: base metal smelting and incineration. Despite significant reductions from the base metal sector, more can be done by employing the “best available technologies” at all plants emitting less than two grams (~ .07 ounces) of mercury per tonne (~ 1.1 tons) of product, which could reduce mercury emissions a further 800 kg/year (~ 0.9 tons/year) by the year 2008 (Ref. 15). Standards also address mercury from medical, hazardous, sludge and municipal waste incineration.

MERS is a national suite of sectorial emission reduction plans, to be built from jurisdictional plans on particulate matter, ozone, and national multi-pollutant analysis (*Ref. 16*). MERS affects several key sectors such as electric power generation, base metals, iron and steel, pulp and paper.

Relevant pending projects of the Pollution Data Branch of Environment Canada were also reviewed:

- publication of mercury trends in Canada from the years 1970 to 2000 in early 2002
- compilation of mercury emissions on an annual basis beginning in the year 2002
- provision of an enhanced data set for modelers in the year 2002
- a forecast for mercury emissions from mid-2002 up to the year 2015

Deslauriers also emphasized the need for research and data on speciation and the important effect the latter information has on modeling and policy development.

2.1.5 Mexico

In Mexico, the majority of mercury consumption, generally of secondary origin, is related to the manufacturing of chlorine, light bulbs, dental amalgams and instruments. Mercury consumption in Mexico in the year 1996 was estimated to be between 30 and 33 tons (~ 27 and 30 metric tonnes) (*Ref. 17*). It has been reported that mercury production has occurred in Mexico since the year 1891 and reached its peak in the mid 1940s. The declining price of mercury in the following years has depressed production. Between the years 1995 and 1999, no primary production was officially recorded for Mexico (*Ref. 18*).

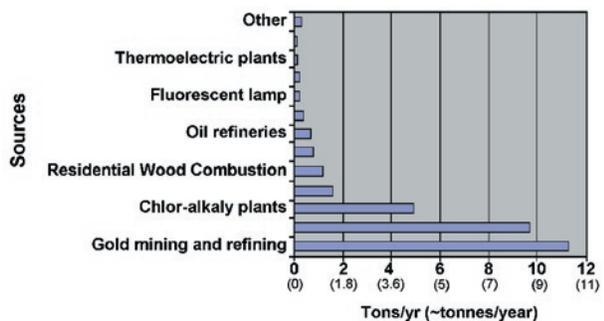
Gildardo Acosta Ruiz from Acosta y Asociados reviewed the methodology behind the assembly of emission inventories in Mexico and the current level of knowledge regarding the major sources of mercury. The previous inventories for mercury compiled in Mexico by the Electric Power Research Institute (EPRI) included the six northernmost Mexican states. In the absence of emissions estimation methodologies developed specifically for Mexico, EPRI relied on estimates based on emission factors developed by the USEPA and Parcom-Atmos of the Netherlands. The results do not take into account Mexico's technological context and the fact that the mercury content in extracted minerals has not yet been analyzed. In the year 2000, the Instituto Nacional de Ecología (INE) identified the major sources of mercury and developed an approach to estimate usage, disposal and emission. For INE, consumption and usage estimates are taken as a surrogate for emissions. Regarding consumption data, these were estimated from official statistics and data provided by the various industries (*Ref. 19*).

The estimates presented by Acosta Ruiz were developed using two approaches drawn from the EPRI, INE and Parcom-Atmos inventories. The first one was to include the activity level of electrical facilities, mercury use in the chlor-alkali industry and mercury process characteristics and behavior. In a second step, data were compared with U.S. information. It was observed that the largest emissions are from gold mining and refining which represent 36 percent of total estimated emissions in the year 1999. Processing of this ore requires two thermal operations: the smelting and the roasting to eliminate organic matter and mercury sulfide during the refining of gold. During these processes mercury is being released and there are no data on recovery of such mercury. Thus it is assumed that the entire release goes into the atmosphere. Using data from 60 mines producing more than 14 kg (~ 31 lbs) of gold per day and comparing them to data from Nevada, Acosta Ruiz was able to produce the estimates shown in **Figure 8**.

Mercury mining in Mexico ceased in the year 1995. Secondary production occurs through reprocessing of old mine tailings at former metal processing plants (*Ref. 20*). This accounts for 31 percent of the known emissions. Chlor-alkali plants, with nearly 16 percent of the total emissions, are also an important source of mercury (**Figure 8 and Table 5**). Mercury consumption data were obtained from these facilities. USEPA in its Mercury Report to Congress estimates emitted losses of 3.5 grams per ton of mercury used in this process as an emission factor whereas INE uses 40 g of mercury emitted per ton of mercury used. In his study, Acosta Ruiz used a factor in between those two. Other contributions, such as the flux from old mercury mining and amalgamation sites and the absence of recycling of thermometers and devices that contain mercury, were not addressed in this study. Furthermore the open burning of refuse at dumps and landfill sites in Mexico may release significant amounts of mercury but there are no data available on such releases.

There is very limited official information available on mercury emissions and mercury content in feedstock or waste streams. This is partly due to the embryonic nature of mercury regulation. Only two types of activity are regulated: ce-

FIGURE 8. Estimated Emissions of Mercury by Sector in Mexico (1999)



Adapted from Gildardo Acosta Ruiz

ment production and burning of waste fuels. Standards for incinerators, hazardous waste and pharmaceuticals are currently proposed and are not yet promulgated and enforced. This inventory is a first step toward a better data set.

TABLE 5. Estimated Mercury Emissions by Sector – Mexico Year 1999
(Adapted from Gildardo Acosta Ruiz)

Source of Emission	tonnes/yr (~ tons/yr)	percent
Gold mining and refining	10 (11.27)	36.03
Mercury mining/refining	9 (9.666)	30.9
Chlor-alkali plants	4 (4.902)	15.67
Copper smelters	1 (1.543)	4.93
Residential Wood Combustion	1 (1.158)	3.70
Carboelectric plants	0.7 (0.7855)	2.51
Oil refineries	0.6 (0.68)	2.17
Amalgams	0.3 (0.378)	1.21
Fluorescent lamps	0.21 (0.229)	0.73
Primary Lead and Zinc smelters	0.19 (0.208)	0.66
Thermoelectric plants	0.01 (0.1263)	0.40
Industrial commercial boilers	0.09 (0.0954)	0.3
Other	0.22 (0.2418)	0.77
Total mercury emissions estimated	28.38 (31.283)	100

2.2 Transport and Deposition

2.2.1 Transport of Mercury

2.2.1.1 Florida Everglades Study

The atmosphere is the dominant transport vector of mercury to most ecosystems, particularly those that are remote from mercury point sources. As mentioned earlier, mercury residence time in the atmosphere is related to its speciation. Long-range transport of mercury is often associated with elemental mercury, while transport on a regional and or local scale is mostly due to Hg(II) and Hg(p) or particulate mercury.

Before presenting his results from the aircraft measurement campaign in Coral Springs Florida, located north of Ft. Lauderdale, and mercury speciation experiments in Point Barrow Alaska, (this latter in cooperation with Dr. Steve Lindberg and others), Dr. Matt Landis of the National Exposure Research Laboratory (NERL) USEPA noted several needs in the study of mercury transport and deposition. Specifically, continued support for improved emissions inventories,

methodology to further determine ambient speciation of mercury, a further determination of defined impacts from specific source types, and linkage to aquatic modeling to allow for bioaccumulation estimates and potential exposure risk assessments were all seen as crucial.

The Florida study was an attempt to identify the sources responsible for the relatively high levels of mercury found in fish in the Everglades, on the southeastern coast of Florida. An aircraft containing sampling equipment able to measure various species of mercury, as well as particulates, NO_x, CO₂, and other parameters necessary to assist in the identification of the sources of prevailing ambient mercury concentrations was used in the study. Supportive measurements were also taken at ground based sites.

Specifically, the objectives of the aircraft sampling were to:

1. Obtain vertical atmospheric profiles (60 - 3500 meters (~ 65 - 3,800 yards)) of speciated ambient mercury off the South Florida coast;
2. identify any vertical mercury gradients that might indicate the presence of rapid mercury chemical reactions in air or in cloud water; and
3. investigate the role of long-range transport of RGM (Reactive Gaseous Mercury) in the marine troposphere.

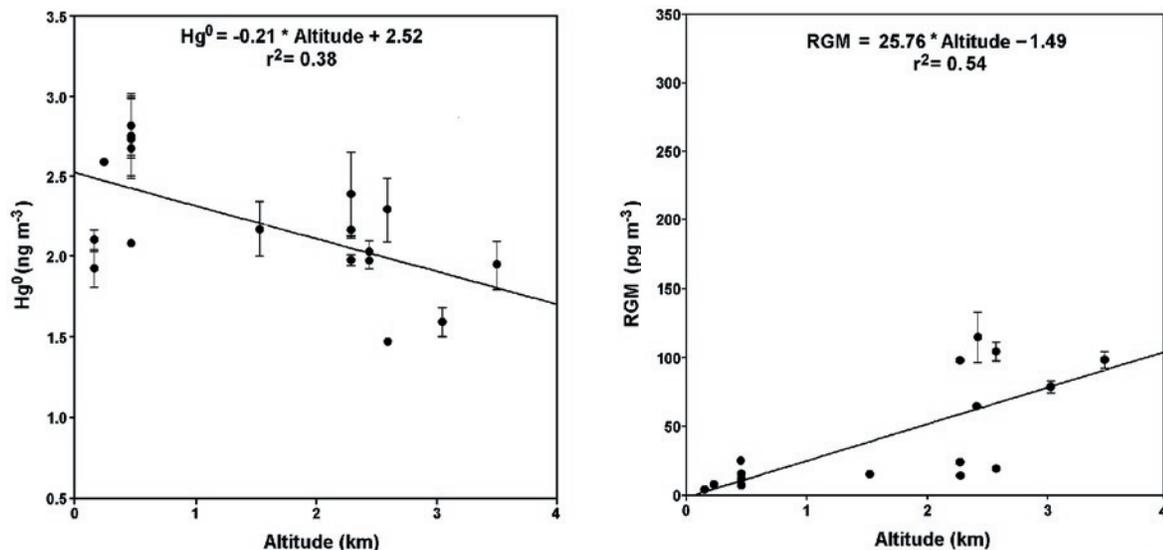
Aircraft samples taken in January 2000, with prevailing winds from west to east, showed that concentrations of elemental mercury were in the vicinity of 2.25 to 2.5 ng/m³ at ground level—above the global background—and decreased with increasing altitude, while reactive gaseous mercury (RGM) increased with altitude (**Figure 9**). In June 2000, with the prevailing wind now from east to west, similar trends occurred.

Thus far, aircraft studies have generated some preliminary conclusions.

1. No evidence supporting the hypothesis that the Atlantic Ocean as a source of RGM was found; however, further sampling above the ocean should allow for more definitive comment.
2. Elevated levels of RGM, accompanied by high sulfur oxide concentrations, were observed at the surface in Coral Springs, Florida, suggesting an association with anthropogenic sources, possibly in Fort Lauderdale and Miami.
3. Elevated concentrations of RGM observed in the marine free troposphere suggest an elemental mercury oxidation mechanism aloft—a reaction with relatively rapid kinetics.

The study of dry deposition of mercury in the form of RGM is important to the evaluation of regional and global mercury budgets. By attempting to determine the forms of mer-

FIGURE 9. Gaseous Hg and RGM aircraft data summary - January 2000 - Florida



(M. Landis) USEPA

cury present in the Florida skies that could be subsequently deposited on land and water, identification of possible mercury sources and associated transport mechanisms should be improved. The data from this study imply that regional transport of mercury, combined with oxidation mechanisms in the atmosphere, explain the high levels of RGM in the Everglades.

2.2.1.2 The Arctic Studies – Barrow Alaska

Mercury levels in Arctic wildlife are elevated above normal levels, notwithstanding some apparent reduction in global anthropogenic emissions over the last decade. Given there are few known mercury sources in the Arctic, an association with long-range transport must be considered (*Ref. 21*), including an exploration of possible mechanisms contributing to the accumulation of mercury from the global pool in the Arctic.

In the fall of 1998, Canadian researchers working at Alert Nunavut in the Arctic described what they referred to as Mercury Depletion Events (MDEs), during which concentrations of depositional elemental mercury dropped to very low levels, well below the global background concentrations. One hypothesis for this depletion was a transformation of the elemental mercury to reactive gaseous mercury, a much more soluble and thus bioavailable form of this contaminant. The presence of RGM could account for the relatively elevated levels of mercury seen in Arctic wildlife, a dietary staple of northern peoples.

Dr. Landis joined with Dr. Steve Lindberg of the Oak Ridge National Laboratory, Steve Brooks of NOAA and others to investigate the accumulation, speciation and cycling of

mercury in the Arctic environment through an intensive sampling program at Barrow Alaska.

This program confirmed the occurrence of MDEs at the Barrow location, 1600 km (~ 994 mi) south of Alert, suggesting that this phenomenon could be widespread in the Arctic, and perhaps in Antarctica. The production of RGM during mercury depletion events was also confirmed.

The conversion of elemental mercury into reactive gaseous mercury appears to be associated with the arrival of the Arctic dawn. Depletion events begin with polar sunrise and persist until snow melt, which in Barrow is early to mid June. During the events, there is evidence of a strong correlation between elemental mercury and ozone, suggesting a link with chemical reactions driving ozone depletion, as the Canadians earlier suggested. No correlation between these two substances is apparent in the months prior to polar sunrise. The reactions with ozone likely also involve sunlight and reactive bromine, as both gaseous and aerosol bromide show strong seasonal peaks up to 100 ng/m³ at Barrow, occurring midway between sunrise and snow melt. The bromide may originate in sea water, and the melting of the ice cap due to climate change may be further encouraging these interactions.

RGM concentrations at three Arctic locations can reach in excess of 300 pg/m³, with occasional levels up to 900 pg/m³, much in excess of measurements at rural sites in the eastern United States and comparable to levels of RGM normally seen near major mercury point sources.

The estimated resulting flux of mercury to the surface (principally by dry deposition) is in the vicinity of 40 μg/m² at peak times. By comparison, total (wet + dry) mercury depo-

sition in the northeastern United States has been estimated at 10 to 30 $\mu\text{g}/\text{m}^2/\text{year}$.

These presentations reinforce the fact that a sophisticated knowledge of the chemistry of mercury in various media is vital to an understanding of its presence and impact in those media, including biota. This chemistry is complex, with many drivers and possible factors for consideration, including the significant transformation of a relatively unreactive and persistent form of mercury (elemental) to a highly reactive, transitory and available form (reactive gaseous mercury).

2.2.2 Putting Deposition in Context with Other Sources and Pathways

Once deposited into bodies of water, inorganic mercury must be converted to toxic methylmercury by methylating bacteria before accumulation in fish and other biota can occur. Methylmercury is soluble, mobile, and is rapidly accumulated by aquatic organisms. Biomagnification associated with methylmercury in the foodchain results in significant mercury concentrations in fish, the main route for human exposure to this toxic compound. According to the Great Lakes Water Quality Board, this contaminant is subject to environmental cycling of mercury previously introduced into the environment (*Ref. 22*). Volatilization of mercury from land and water surfaces into the atmosphere can result in subsequent transport and deposition followed by revolatilization. Mercury contaminated sediments may resuspend mercury compounds in the water, allowing for bioaccumulation in the food web. In Maryland it was estimated that less than 20 percent of the mercury deposited in wet and dry deposition would be exported in streams and rivers (*Ref. 23*). Rather, there has been a buildup of mercury in soils and sediments.

During the workshop Greg Mierle, from the Dorset Environmental Science Centre of the Ontario Ministry of the Environment, presented similar ideas. He argued that, despite the significant direct input of mercury to lake surfaces from the atmosphere, methylmercury in fish is derived either directly or indirectly from mercury in watersheds and that watersheds contain large pools of mercury accumulated over very long periods of time. Mierle pointed out that a study he conducted eleven years ago showed that, in an average lake, precipitation directly to the lake surface accounts for about half the total load of mercury deposition.

Several studies have attempted to explain where the mercury in streams originated. One interesting clue about the factors controlling mercury release from watersheds was the relationship between dissolved organic matter and mercury in stream water. A quantitative measure of colour was used as a surrogate for the presence of organic matter in waterbodies, and the trends in colour and mercury concentration in

a typical stream very closely track each other over the two year monitoring period. It could then be concluded that bioavailability of mercury in the water column is influenced by dissolved organic carbon concentration. In the study, the colour in stream water was due to humic substances, and humic substances are associated with wetlands. This association suggested that the release of mercury could be associated with wetlands. John Rudd from the Experimental Lakes area in northwestern Ontario established in the early 1990s that wetlands are major sources of methylmercury which corroborates Mierle's results. At the end of the study it was concluded that:

1. Wetlands are sources of methylmercury

- Most methylmercury in lakes comes from either wetlands or in-lake production.
- Current precipitation (wet deposition) is a minor source of this particular species (except for extreme scenarios of methylmercury in precipitation).
- No assumptions are made about source of in-lake mercury.

2. Watersheds as sources of methylmercury

- Total mercury inputs appear dependent on wetland area.
- The in-lake production of methylmercury can be allocated based on loading of total mercury.
- Watersheds, directly or indirectly, dominate methylmercury inputs to lakes.

Mierle noted that if these models and observations show that mercury found in fish is dominated, in most cases, by mercury exported from watersheds, one question remains: where does the mercury in watersheds come from?

In general, mercury in soil is stable for long periods of time, usually remaining on the surface of the sediment or soil, rather than moving through the soil to groundwater. Indeed, once incorporated into soils, mercury is tightly bound to organic matter and is not easily released. Thus, freshwater and marine sediments are important repositories for mercury. This strong adsorption of mercury to particulate matter also suggests that the transport of mercury-contaminated particles carried in surface runoff is an important mechanism for moving mercury from soil to water (*Ref. 24*).

Mierle presented a study done in Québec showing high concentrations of mercury at the surface of soils. This is consistent with the idea that watersheds store mercury deposited from the atmosphere. Studies related to mercury deposition patterns in soil in Québec and Ontario raised a number of uncertainties about mercury pools:

- Are the pools of mercury in soil active?
- Does the age of organic matter indicate the age of associated mercury?

- How do soil disturbances (logging, fires, flooding) affect the pools?
- How will climate change affect mercury pools in soil?

Mierle pointed out that the interaction between mercury in soils and incoming deposition was not well known. Climate change, with the increasing average ambient temperatures and dryness, might accelerate decomposition and promote mobilization of mercury in soils. Mierle concluded that the outcome of emission reductions on mercury in fish is quite uncertain. There are large pools of mercury in watersheds, and they may modulate the response to any changes in deposition. Furthermore the existing pools may be susceptible to mobilization and ultimately further contamination of fish. There is a present need for models which elucidate the interactions of mercury in the watershed.

In his report, Robert Mason from the University of Maryland Center for Environmental Science expressed similar concerns and conclusions advanced by Mierle. “While current and pending legislation should lead to a decrease

in anthropogenic input of mercury to the atmosphere, there will be a long legacy of mercury in watersheds where it is typically strongly retained. The mercury in soils will slowly be released to watersheds even after curtailment of anthropogenic sources, and could exacerbate the mercury problem for decades to come.” (*Ref. 25*) It will thus be more prudent to take into account existing pools of mercury in the development of programs and policies concerning mercury. When asked if he meant that efforts towards investing further in the abatement of mercury emissions were not appropriate, Mierle responded that investing in emission reduction should not cease. However, this investment should be balanced with more investments in research. Indeed, in the opinion of the presenter, more needs to be known about how mercury behaves before committing to further funding of control programs, to ensure that large investments for reduction of emissions yield the desired results. To some extent, the METAALICUS research project currently underway in the Experimental Lakes Area of northwest Ontario will respond to many of these research needs.

3.0 MONITORING

3.1 United States

3.1.1 Monitoring Measurements of Mercury Speciation in Coal and Waste Incinerator Flue Gas

During the workshop, Eric Prestbo presented data from his study of mercury species and their interaction in combustion source plumes from a coal-fired power plant and a municipal waste combustion unit using a stack linked mercury dilution sampling technique to simulate the plumes from these sources. The goal of his work was to examine the physical and chemical transformations of mercury in the plumes of the two types of combustion sources to allow better determination of the evolution of individual mercury species and volumes of mercury contained in such combustion source plumes.

For the workshop presentation, recent work at a coal-fired facility was reviewed. A Static Plume Dilution Chamber (SPDC) was used; this technology allows simulation of the conditions typically encountered by a plume discharged from a combustion source, and the quantification of different species of mercury which exist and interact in the plumes of the monitored sources (*Ref. 26*) (particulate Hg, gas phase Hg(0), Hg(II), and total Hg; dry deposited Hg; and dissolved and particulate phase Hg in simulated rainwater (SRW)). The following conclusions arose from this work:

- The gaseous Hg(II) input to the SPDC is rapidly dry deposited to the chamber surface in the absence of simulated rain.
- In the absence of simulated rain, on average only 4.0 percent \pm 2.7 percent of the total injected gaseous Hg(II) was detected in the SPDC.
- The amount of elemental mercury (Hg(0)) increases in the plume, suggesting rapid (less than 5 minutes) conversion of Hg(II) to Hg(0).
- The gaseous Hg(II) input to the SPDC is efficiently scavenged by rain water.
- In the presence of simulated rain, on average only 0.67 percent \pm 0.74 percent of the injected gaseous Hg(II) is observed in the SPDC.
- The wall-wash or rain-out Hg(II) is found overwhelmingly in the dissolved phase.

Other SPDC observations included a greater conversion of Hg(II) to Hg(0) during SPDC daytime simulations. The conversion of Hg(II) to Hg(0) was the largest (by a factor of 6) for tests in which 200 ppb of ozone were added to the SPDC. Also there was significantly more particulate mercury observed in the air and water fraction under these conditions. There was

no evidence of significant amounts of gaseous Hg(II) adsorbing to the particulate phase: this contrasts with past Tennessee Valley Authority (TVA) plume studies.

The following comments on coal-fired utility plume mercury chemistry were put forward.

- Conversion of Hg(II) to Hg(0) was significant, which is consistent with the reactivity of the flue gas matrix and observations of HgCl₂ to Hg(0) behavior in the laboratory.
- This Hg(II) to Hg(0) conversion has been observed in three different power plants, two different SPDC devices and at various dilution ratios.
- This conversion is too rapid to be observed by the SPDC.
- Initial results of the ground-based SEARCH program to measure downwind plume chemistry supports SPDC observation of Hg(II) to Hg(0) conversion.

It was concluded that mercury emissions follow a dual pathway, one which contributes to the global burden of elemental mercury and the second which results in the local wet and dry deposition of more reactive forms of this contaminant.

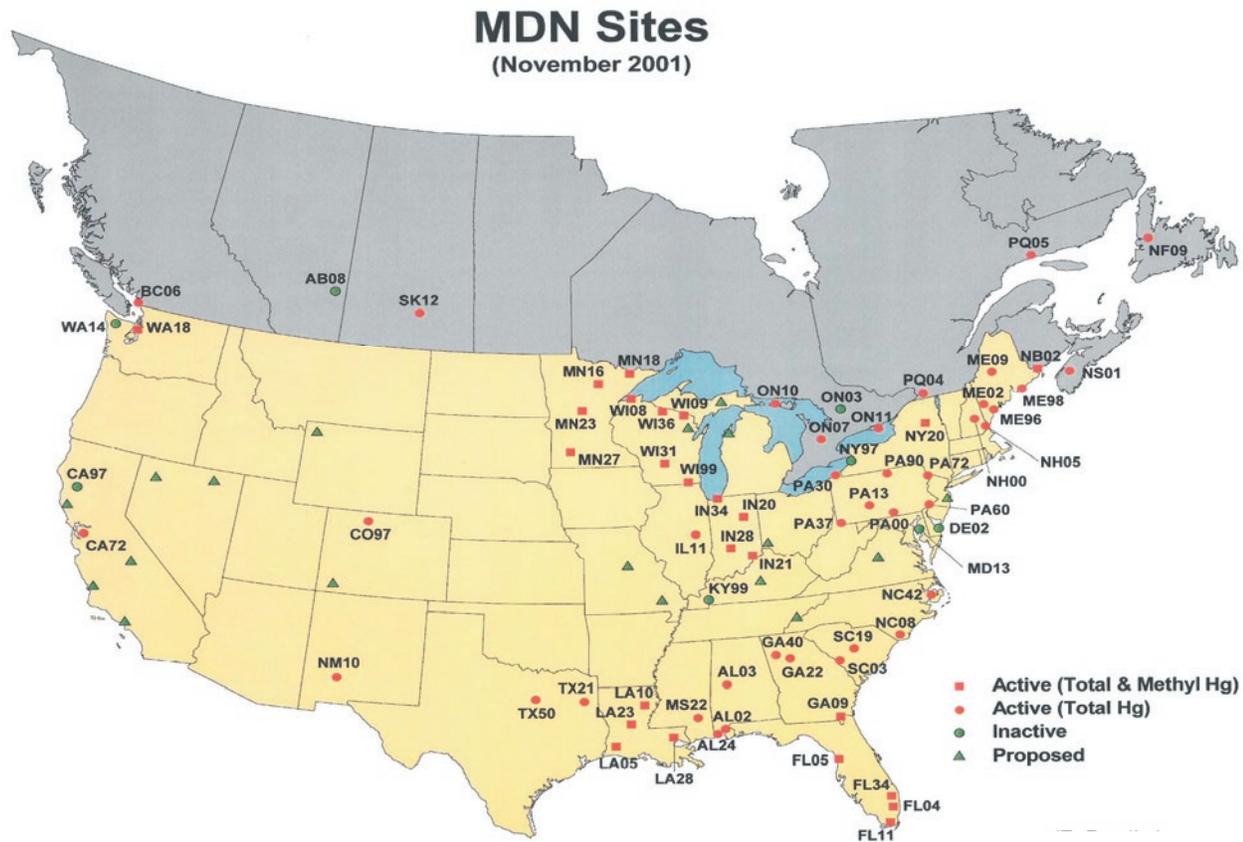
3.1.2 Monitoring Networks

In the United States and Canada, the majority of locations measuring mercury wet deposition rates are part of the Mercury Deposition Network (MDN). In the United States, the MDN is one of three networks within the National Atmospheric Deposition Program (NADP). The NADP, initiated in the late 1970s, is a cooperative effort among many different groups, including the State Agricultural Experiment Stations, U.S. Geological Survey, U.S. Department of Agriculture, and numerous other governmental and private entities.

The purpose of NADP is to collect data on the chemistry of precipitation for monitoring of geographical and temporal long-term trends nationwide. The objective of the MDN is to develop a national database of weekly concentrations of total mercury and methylmercury in precipitation and the seasonal and annual flux of total mercury in wet deposition. The data would be used to characterize the extent of the mercury problem, describe the regional patterns of mercury deposition, and assess deposition changes over time. Over 50 sites were in operation during the year 2000 (*Ref. 27*) (**Figure 10**).

The network uses standardized methods for collection and analyses. Weekly precipitation samples are collected in modified Aerochem Metrics model 301 sampling units. The “wet-side” sampling glassware is removed from the collector every Tuesday and mailed to the Hg Analytical Laboratory (HAL) at Frontier Geosciences in Seattle, Washington for

FIGURE 10. Mercury Deposition Network (MDN) site map - November 2001



analysis by cold vapor atomic fluorescence. The MDN provides data for total mercury, but also includes methylmercury if desired by a site sponsor. Data are available online at: <http://nadp.sws.uiuc.edu/mdn/>, for the transition network (year 1995) and for the years 1996 through 2001. Network operation is expected to continue to the year 2005 and perhaps beyond.

Eric Prestbo from Frontier GeoSciences gave an overview of the characteristic elements of NADP and MDN. Both share the following characteristics:

- Regional/National/International in Scope
- Regionally Representative, Mostly in Rural Settings
- Uniform Sampling and Analysis Procedures
- A Single Central Laboratory for Analysis
- Rigorous Field and Laboratory Quality Assurance Program
- Rapid and Open Data Dissemination (Web)
<http://nadp.sws.uiuc.edu/mdn/>
- Independent Site Audit Program

3.1.3 Measuring Near-Source Wet and Dry Deposition

Eric Prestbo also presented research on mercury monitoring in ambient air near anthropogenic sources. The purpose of his observations of elevated near-source wet and dry deposition is to provide field data in support of the static plume dilution chamber study previously reviewed, particularly much needed mercury deposition data near known mercury emission sources. The USEPA 1997 Mercury Report to Congress established a plausible link between anthropogenic sources and mercury deposition to aquatic ecosystems but there is a lack of adequate mercury measurement data near the sources considered in that report.

The lack of such measured data precludes a comparison between concentrations estimated from modeling results and those measured adjacent to sources. Dr. Prestbo indicated that there was a need for future near-source mercury wet and dry deposition studies with high resolution sampling and meteorological support data. Such studies should include air measurements of mercury speciation (RGM, Hg(0), Hg(p)) on an event basis, with a temporal resolution brief enough to capture before- and after-storm mercury concentrations. These data should then be used to improve plume model chemistry and verify the outputs of such models.

3.1.4 Mercury Dry Deposition

Dr. Jerry Keeler from the University of Michigan presented his research (undertaken in collaboration with Matt Landis and Steve Lindberg) on mercury dry deposition in the Florida Everglades. The Florida Everglades Dry Deposition Study (FEDDS) was conceived as an intensive effort to improve understanding of the processes associated with mercury dry deposition, provide information to enhance the representation and parameterization of dry deposition in atmospheric models, and make deposition estimates which could serve as validation checks on the models applied (*Ref. 28*).

Despite the fact that the measurements were made 25 miles (~ 40 km) from the coast and approximately 10 miles (~ 16 km) away from any significant anthropogenic sources of mercury, high levels of RGM were observed along with the diurnal cycling behavior observed in other studies. It was also noted that, in the presence of dew, RGM disappears.

Southeasterly winds from the Miami urban area, an area containing significant mercury emission sources (waste incinerators), were present during much of the sampling, allowing a correlation between high levels of RGM and these sources of mercury. The project also measured particulate phase mercury to water surfaces by a technique developed with USEPA in the mid 1990s to measure deposition on the water surface of the Everglades. The water surface technique enabled the modeling of dry deposition onto the water interface and a better understanding of dry deposition in general.

3.1.5 Monitoring Rural, Urban, and Mobile Sources

The behavior of RGM was further analyzed by Dr. Keeler in samplings near Ann Arbor, Michigan, where large sources of mercury are more than 15 miles (~ 24 km) away in Jackson, Michigan, (also incineration facilities). Again the data showed a diurnal profile and, on occasion, fairly high values were observed. During precipitation events, RGM was depleted. The question arose as to whether precipitation was removing some of the RGM or whether there was a frontal passage with an air-mass shift. Levels of RGM appeared to be correlated with levels of ozone concentration. This suggested a relationship between the photochemistry of ozone and the formation of RGM which was counter to those developed by Steve Lindberg; in Keeler's study RGM levels increased in the presence of elevated ozone, whereas in the Arctic RGM increased when ozone was depleted. This suggests that RGM behavior in the Arctic, coastal regions and rural areas could be distinct from that observed in urban environments. Studies in any of these areas should reflect these distinctions. A correlation between RGM and elemental mercury was also observed; when RGM levels

rose, elemental mercury decreased in an inverse relationship. This occurred particularly on days when elevated ozone concentrations were present.

Samples were also taken in Detroit Michigan. In urban areas, there appears to be, on average, two or three times more elemental mercury present as well as some RGM. It was observed that RGM and elemental mercury concentration levels did not appear to be correlated in this environment.

Keeler also presented his ongoing research on mercury emissions from mobile sources. One potential source of emissions of reactive gaseous mercury (RGM) not previously examined is emissions from mobile sources - gasoline or diesel powered vehicles. The objectives of this research are to develop a mobile source signature for automobiles and heavy diesel trucks and to estimate the magnitude of mercury emission rates from the two classes of vehicles (*Ref. 29*). The study was conducted at the Interstate 95 Fort McHenry tunnel complex near Baltimore, Maryland. With respect to mercury levels due to motor vehicles, the tunnel studies in the vicinity of Baltimore are not yet conclusive.

3.1.6 Future Monitoring Needs

In presenting his research projects, Dr Keeler emphasized the importance of environmental monitoring and highlighted the fact that there was no adequate national effort to properly assess the trends in ambient mercury or the dry deposition of mercury. This lack of ambient gaseous and dry deposition data is coupled with insufficient measurements of mercury speciation in the atmosphere. Even though several states use the MDN wet deposition network to monitor weekly mercury levels, daily and hourly deposition data are needed if methylation rates and their linkage to fish are to be better understood. As he testified to the U.S. House of Representatives in May 2001: "(...) spatial coverage at present is not adequate to assess emissions trends or to assess the efficacy of reduction programs. The highest deposition (wet and dry) is found in and downwind of (...) large urban areas where there are at present few monitoring sites. [Support is needed] to improve the spatial coverage (...)." (*Ref. 30*) During the workshop Dr. Keeler stressed the importance of monitoring in different environments such as the Arctic, rural and urban areas and the marine environment. One study in a given environment cannot be taken as a surrogate for studies in other distinct locations because of the significant variation in ambient data secured at different locales.

Similarly, Eric Prestbo highlighted the need for data resolved both spatially and temporally in order to calibrate regional and continental models, and monitoring of mercury in marine environments. Since no data are available on oceanographic deposition of mercury, this should be explored along with some evaluation or estimation of mercury transport from Asia. Furthermore he emphasized the need to support

technology transfer and new monitoring site development in Mexico, while addressing the conversion of divalent mercury to elemental mercury in plume chemistry (particularly in plumes from coal-fired utilities). The lack of adequate ambient mercury data near anthropogenic sources impairs the understanding of plume chemistry. As well, the rationale behind the accumulation of particulate mercury in the tropopause needs to be explored.

Both speakers emphasized that, given that a clear understanding of the limitations and boundaries of models is considered necessary, good monitoring increases confidence in the delineation of source-receptor relationships via application of various models.

3.2 Canada

3.2.1 Monitoring Networks

In Canada, monitoring of mercury deposition is done by the Air Quality Processes Research Division (ARQP) of Environment Canada. Atmospheric mercury has been measured continuously at the Alert site in Nunavut since the year 1995 as part of the Northern Contaminants Program (NCP), and it is currently being measured as a vapor, on particles and concentrations in snowfall (Ref. 31). These measurements at Alert are also part of the Canadian Atmospheric Mercury Network (CAMNet), which also includes operation of the Canadian segment of the Mercury Deposition Network. CAMNet was initiated in the year 1996 to provide a better understanding of mercury trends and processes in the environment. Pierrette Blanchard from ARQP presented the main objectives of CAMNet as follows:

- To improve the current understanding of the atmospheric transport, transformation and removal processes for mercury and its ecologically significant compounds released into the atmosphere.
- To examine spatial and temporal variability in mercury in air and precipitation on a regional/national basis.
- To examine source-receptor/transboundary transport of mercury.
- To investigate atmospheric mercury chemistry.
- To support ecosystem and human health research.

Currently, there are 13 sites across Canada (Figure 11) measuring total gaseous mercury (TGM) and, as part of the MDN operation, mercury in precipitation.

CAMNet initially focused on the measurement of total gaseous mercury (TGM) at selected sites across Canada. Efforts were initiated to establish standardized operating

FIGURE 11. Canadian Atmospheric Mercury Network (CAMNet) and Mercury Deposition Network (MDN) in Canada



procedures and audit and data management/quality control protocols to ensure that TGM data gathered at various sites were comparable and of high scientific quality. The CAMNet mandate has recently been expanded to share knowledge and coordinate measurements of mercury, including methylmercury, in precipitation (largely as part of MDN). New initiatives include measurement of reactive gaseous mercury. In the future, sampling particulate mercury in the atmosphere (Ref. 32) may also be included.

3.2.2 Monitoring Atmospheric Mercury in Atlantic Canada

Dr. Blanchard also reviewed the monitoring activities on the Canadian east coast. The Atlantic Provinces lie downwind of North American anthropogenic emission source regions and are receptors of some forms of these mercury emissions (Ref. 33). In the year 1995 measurement of total gaseous mercury (TGM) with a continuous gas analyzer began in Kejimikujik National Park, Nova Scotia and at St. Andrews, New Brunswick. Total mercury in precipitation for a six-month period in 1996 in Kejimikujik averaged 11.4 ng/L compared to 9.1 ng/L in St. Andrews. Mercury deposition via wet precipitation was estimated to be 8.4 $\mu\text{g}/\text{m}^2/\text{yr}$ in the St. Andrews area and 10.5 $\mu\text{g}/\text{m}^2/\text{yr}$ in Kejimikujik (Ref. 34).

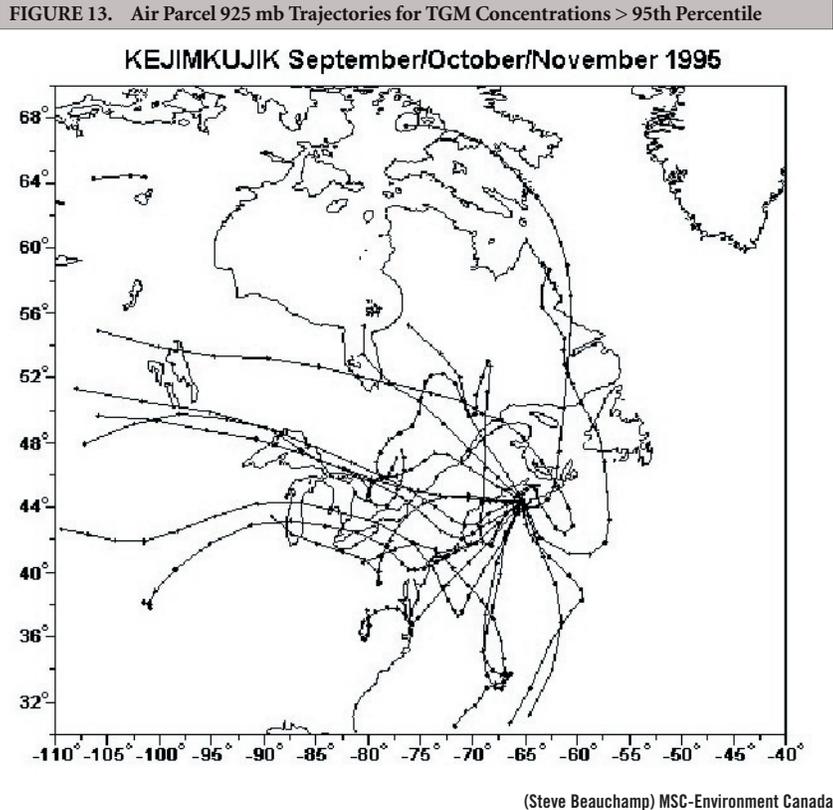
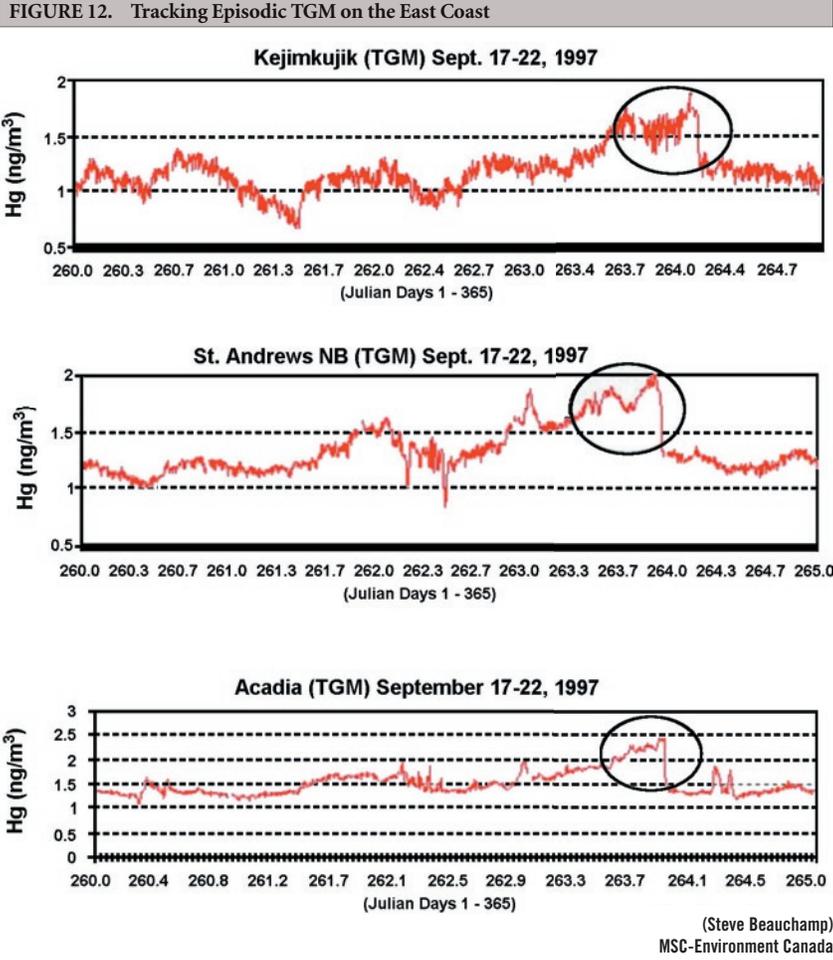
Data presented during the workshop suggest a spatial and temporal variability in total gaseous mercury linked to anthropogenic pollution by other contaminants (e.g. elevated ozone and particulate concentrations). From September 17 to September 22, 1997 total gaseous mercury increased over a two day period in a west to southwest flow but dropped back to “normal” levels following the passage of a cold front. Higher mercury concentrations and wind speeds together increased horizontal mercury flux by two- or threefold leading to the peaks observed in Figure 12.

Furthermore, a study of air parcel trajectory climatology during periods of elevated total gaseous mercury concentrations indicated that such higher concentrations may reflect the influence of a combination of local and long-range sources (Figure 13).

Also, seasonal patterns were observed for total mercury between the years 1996 and 1998, with higher concentrations occurring during the summer months (Figure 14).

3.2.3 Atmospheric Mercury in Ontario

To further the understanding of the temporal and spatial distributions of gas phase mercury in Ontario, two TGM monitors were installed, one at the Integrated Atmospheric Deposition Network master station at Point Petre (near Hamilton) and the other at a satellite site at Egbert (near Toronto). Comparisons between TGM and ground level ozone measurements for Egbert emphasize the non-linearity of gas phase oxidation of mercury by ozone. This lack of correlation with ozone was observed at high levels (above 20 ppb) of the latter contaminant and Blanchard suggested that it may be due to secondary aerosol formation. Indeed, below five ppb, a negative correlation was observed in plumes from Toronto. A greater linearity was observed with PM_{2.5} and SO₂. This would likely be due to the commonality of sources of these pollutants and mercury. A particular episode of larger TGM concentrations at Point Petre can be traced to the more populated and industrialized areas southwest of the site, demonstrating the impact of anthropogenic activities on the background ambient air concentrations (Ref. 35). A Tekran instrument was placed on a buoy in Lake Ontario, between the Hamilton and Toronto sites. When high mercury concentrations are observed at the buoy, Point Petre also experienced elevated levels shortly thereafter. In Egbert, a peak in concentration was still present, but was less significant because of its northern location (Figure 15).



3.2.4 Monitoring in Québec

Dr. Blanchard also presented data from monitoring efforts in the province of Québec. With an area of over 1,600,000 km² (~ 617,761 mi²), anthropogenic emissions in Québec in the year 1995 were estimated as 1.6 tonnes (~ 1.76 tons) per year; however, the estimated total wet deposition was 4.2 tonnes (~ 4.63 tons) per year as estimated by Dr. L. Poissant. Québec could thus be considered a net receptor of mercury via deposition.

Concentrations in precipitation have been estimated as approximately 5 ng/L. Mercury concentrations in precipitation have been monitored at two sites along the St. Lawrence River on a weekly schedule since April 1998. The first site, St. Anicet, is located near the Québec-Ontario border and the second site is located at Mingan, a more inland location on Anticosti Island. Both sites cover the entrance and the exit of the main St. Lawrence river winds corridor (up-downwind). The volume-weighted averages of total mercury concentrations between May 1998 and December 2000 were 8.75 ng/L and 4.92 ng/L at St. Anicet and Mingan respectively. Temporal trends have also been observed; similar to Nova Scotia, a seasonal pattern was present, with higher concentrations of mercury during the spring and summer months (Figure 16).

3.2.5 Other Canadian Atmospheric Mercury Network (CAMNet) Activities

Dr. Blanchard also pointed out that CAMNet was involved in regional mercury modeling and ecosystem modeling studies. The aim of those studies is to understand how mercury is exchanged between the different compartments of the environment. The study of the biogeochemical cycling of mercury should allow a better understanding of the mercury linkages with human health. Research on contaminated sites is also underway, with studies of mine tailings, sewage treatment, mercury from forest fires and landfills; source studies mainly focus on plume characterization. Dr. Blanchard also presented the different national and international initiatives in which CAMNet is involved such as the protocol to the Convention on Long-Range Transport of Air Pollution on Heavy Metals (United Nations – Economic Commission for Europe) or Canadian Environmental Protection Act (CEPA).

FIGURE 14. Total Mercury deposition at Kejimikujik National Park 1996-1998

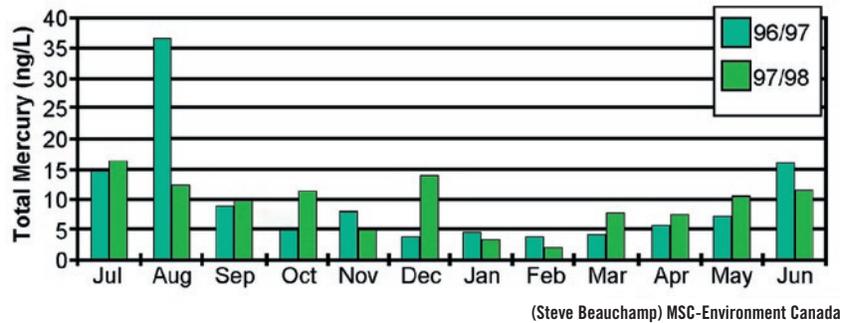


FIGURE 15. Total Gaseous Mercury (TGM) concentrations at Egbert, Point Petre and Lake Ontario (Buoy)

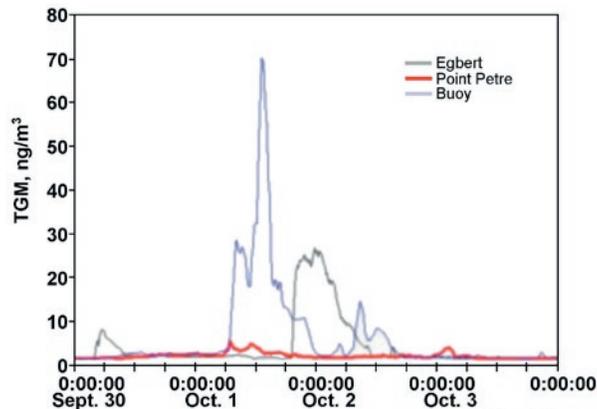
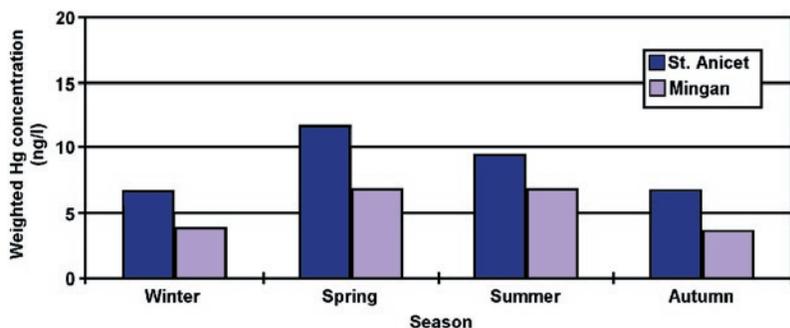


FIGURE 16. Seasonal Hg Concentration at St. Anicet and Mingan



The presentation was concluded by a summary of CAMNet characteristics:

- Systematic measurements of mercury in air and precipitation are being made across Canada.
- Spatial (E-W, N-S), temporal (high-resolution), Audit Protocols, QA/QC and Data Management Protocols are included.
- Spatial and temporal variations are being observed and interpreted in terms of meteorology and other factors.
- Support is provided to atmospheric and ecosystem model development and evaluation.

- Linkages are made to ecosystem and human health issues.
- CAMNet fits within the CEC NARAP Environmental Monitoring and Assessment objectives concerning mercury.

3.3 Mexico

3.3.1 Monitoring Needs

There is no current atmospheric deposition or ambient atmospheric concentration monitoring for mercury in Mexico. Effor 1991 indicate that the Cuatitlán River in the State of Mexico and the Grand Canal in the Federal District (Mexico City) exceeded the limits of 0.001 mg/L (Mexican ecological water quality criteria) and 0.2 mg/L (Mexican technical ecological standard on hazardous waste) with measurements in the vicinity of 0.3 and 0.2 mg/L, respectively. Both rivers receive discharges from a nearby metropolitan zone. Between the years 1994 and 1998, the CNA did not report measurement of any excessive concentrations of mercury in any river. This lack of consistent and continual mercury monitoring is caused by the absence of mercury regulations in the country. The National Institute of Ecology (Instituto Nacional de Ecología-INE) has undertaken the development of mercury measurement capacity in anticipation of further monitoring.

3.3.2 Current Monitoring Capacity and Needs in Mexico

Pablo Maiz Larralde from Gamatek, S. A de C.v laboratories in Mexico presented some key issues for mercury monitoring in Mexico. He noted that mercury source testing began in the 1990s for incineration facilities (medical waste and industrial hazardous waste) and cement kilns burning hazardous waste. Other sources of mercury such as combustion facilities, mercury production, miscellaneous stationary sources and fugitive and area sources are not regulated and monitored. The testing requirements for cement kilns and incineration facilities have grown slowly. According to Larralde this is due to the lack of:

- regulatory emission standards for stationary sources;
- capable and reliable testing firms in Mexico;
- an approach to environmental responsibility by the emitting facilities.

The regulatory mercury scenario for these two stationary emission sources is presently based on permits. These permits include several operating conditions and emission standards. However, they do not establish the source testing reference methods to be used in most of the cases, nor the testing protocol to be followed; thus, results obtained between facilities cannot be compared. A new standard for incineration facilities (which does not include industrial combustors, crematories and combustion units that burn

alternative fuels) is under development. This standard will also establish a reference method for the measurement of mercury, but it will not include a compliance testing protocol. The reference method adopted in Mexico currently is equivalent to USEPA Reference Method 29, but has significant deviations from the sampling quality assurance/quality control (QA/QC) requirements. The deviations consist mainly in calibration requirements, and the use of appropriate acceptance/rejection criteria for several method performance indicators.

The cause of these deviations is partially due to a lack of reliable basic source test methods for parameters such as:

- gas velocity
- molecular weight (global gas composition)
- water vapor content
- isokinetic testing

Because of the QA/QC procedures that are partially or totally omitted by Mexican laboratories when using Method 29, a strengthening of basic source test methods is needed in Mexico. Larralde identified these methods as presented in **Table 6**.

Larralde also noted that mercury concentrations in ambient air and/or fugitive or area source emissions have not been monitored on a regular basis in Mexico as laboratories have not implemented sampling and analytical methods in support of such measurements.

TABLE 6. Basic Ancillary Source Test Methods when Using USEPA Reference Method 29 for Testing (adapted from Pablo Maiz Larralde)

Method 1 (USEPA Reference):
<ul style="list-style-type: none"> • Testing for Cyclonic Flow • Treatment for rectangular stacks
Method 2 & 2C (USEPA Reference):
<ul style="list-style-type: none"> • Calibration/Verification of Pitot Tubes • Use of appropriate Differential Pressure Gauges • Calibration/Verification of Stack Temperature Gauge • Barometric Pressure appropriate estimation • Pitot Tube – Differential Pressure Gauge system leak check procedures
Method 3, 3A & 3B (USEPA Reference):
<ul style="list-style-type: none"> • Performance Specifications for Instrumental Measurement Systems • Global Gas Composition acceptable variations • Appropriate use of Fuel Factors • Leak check procedures
Method 4 and 5 (USEPA Reference):
<ul style="list-style-type: none"> • Dry Gas Meter (DGM) Calibration • Calibration/Verification of DGM Temperature Gauges • Nozzle real diameter measurement
Method 29 (USEPA Reference):
<ul style="list-style-type: none"> • Use of Blank data • Use of Audit Samples

He concluded by emphasizing that mercury monitoring and testing are still in their infancy in Mexico and great opportunities for improvement exist. Although Mexico is attempting to follow source sampling Method 29 of the USEPA there are many deviations from the method, as appropriate technology is not yet available. As well, there are many quality assurance and quality control issues that must be dealt with, and standards are not yet implemented. The uncertainty of measurements is not being determined and there are no data presently available for ambient mercury levels. To correct these matters, training programs must be implemented, inter-laboratory comparisons conducted, and technological transfer realized. Larralde outlined the key sectors where improvements are needed, which are presented in **Table 7**.

3.4 Summary of Panel Discussion: Status of Source Inventories and Monitoring

The first day of the workshop concluded with a panel discussion during which participants reviewed the salient

TABLE 7. Key developments needed for monitoring mercury in Mexico (adapted from Larralde)

Regulatory Aspects
<ul style="list-style-type: none"> · More regulation toward emitting sources · Establish complete compliance testing protocols
Testing Firms and Laboratories
<ul style="list-style-type: none"> · Follow standardized reference methods · Obtain complete traceability in measurement · Reduce and estimate uncertainties · Conduct training programs · Develop accreditation · Participate in proficiency testing and interlaboratory comparison programs

issues pertaining to source inventories and monitoring of mercury. The facilitated group discussion focused on the following questions:

- In order to improve monitoring, what specific needs should be addressed?
- What elements in the source inventories need attention and enhancement?
- Are source inventories and monitoring activities responding to the needs of the modelers?
- How can policy makers be encouraged to invest in monitoring activities and improvement of inventories?

Participants and panelists agreed that speciated monitoring was imperative if a full understanding of mercury's behavior in the atmosphere and the larger environment was to be achieved. It was noted that, while a mercury wet deposition network was in place, there was no systematic network of speciated monitoring sites, despite speciated measurements at CAMNet stations and other discrete locations. Along with

speciated monitoring, further understanding of the kinetics of chemical reactions involving mercury should be developed with increased involvement of laboratories.

Monitoring of mercury dry deposition was also seen as a salient issue. Mercury dry and wet deposition are distinctly different processes. The results of several studies indicate that the loading of mercury species associated with dry deposition could be three times that of the wet deposition as typically determined by the Mercury Deposition Network (MDN). For example, a recent study conducted in the United Kingdom by Dr. David Fowler confirmed this estimated distinction in loading. These results suggest that careful consideration be given to methods used to determine the extent of the contribution of dry deposition to the total mercury loading to the ecosystem.

The participants also stressed the need for the monitoring of other pollutants in order to establish possible links and correlations. Further studies on other metals in anthropogenic emissions and in deposition are necessary to determine the significance of their possible interaction with mercury. Further monitoring programs sustained over 10 or 15 years will be crucial to the determination of the overall loading of mercury associated with the atmospheric transport and deposition. Standardization and enhancement of quality assurance and quality control (QA/QC) procedures were emphasized when extending both short and long-term monitoring efforts.

Specific monitoring needs were outlined for Mexico; the need to build analytical capacity for monitoring and assessment, improved QA/QC, increasing the reliability of the data generated with standardized reference methods, providing training and accreditation programs and designing regulatory policies. While the last item can be considered an internal process, involving Mexican politicians and legislators, it was acknowledged that increasing monitoring capacity and reliability needed to be initiated in partnership with the United States and Canada. Sharing technology, analytical capability and results were considered indispensable to obtaining a holistic view of mercury emission and deposition in North America. The efforts of the CEC to date were acknowledged but it was agreed that more was needed.

Most workshop presenters stressed the need to improve and enhance current emission inventories for mercury. Modelers were particularly adamant on this issue since accuracy of inventory data is crucial to the accuracy of their models. It was noted that standards and QA/QC exist for measurements but these are not so apparent in the development of inventories. Source inventories appeared to lack traceability and inconsistencies often exist between national inventories and state or provincial ones. It was put forward that meta analysis, independent review of inventories and a more rigorous methodology should be put in place. Also, an ac-

curate update of basic information from major facilities (e.g. operating life of the facility, last update in pollution control equipment) should form part of emissions inventories. The USEPA is currently investigating the source categories in each state and macro tools were being installed in order to evaluate inventories further but more efforts are needed.

Modelers emphasized the importance of knowing the range of emissions, i.e. the degrees of freedom when statistical data on emissions were compiled. This would allow the use of different boundaries in models, leading to a series of scenarios that would take into account the uncertainties present in the source inventories.

The economic implications of the aforementioned needs were also discussed. It was agreed that long-term efforts are needed with funding of monitoring and the examination of cycling of mercury in the ecosystems. The scientific community needs to convey the importance of these activities using emerging and potential impacts on the health of humans and wildlife as ultimate endpoints. The emphasis should be placed on long-term activities by putting forward the returns on investment of each type of research and project versus the initial cost. Currently intensive efforts are being

funded in different areas but there seems to be no long-term commitment to allow sustained holistic monitoring of speciated and ambient mercury.

The panel discussion summarized the salient issues as:

- Speciated monitoring is required in order to better comprehend deposition patterns.
- Enhanced monitoring of dry deposition is essential for a complete understanding of mercury loading in the environment.
- Monitoring other pollutants and their interaction with mercury is needed.
- Standardization of measurements is critical. This could be achieved as part of the improvement of quality assurance and quality control.
- Mercury monitoring improvement in Mexico is not only a local issue but is crucial to monitoring efforts in North America.
- Source inventories require a more transparent methodology and increased rigor.
- The human health issue is the major incentive for interaction with policy makers.

4.0 MODELING MERCURY EMISSIONS

Analysis of the impact of air pollutant emissions requires determination of the spatial distribution and magnitude of sources and sinks, and an understanding and quantifying of the processes governing pollutant fluxes between the atmosphere and land or water surfaces in order to predict scenarios of deposition based on emission levels. A number of modeling techniques for the study of atmospheric mercury processes have been developed in the last decade. Currently four types of models are largely in use: (1) Lagrangian, (2) Eulerian, (3) mass balance, and (4) receptor.

- (1) Lagrangian models are usually formulated under assumptions of simplified turbulent diffusion, no convergent flows and no wind shear. In these models only first-order chemical reactions can be treated rigorously. Their advantage lies in the fact that they require less computational resources and can facilitate an understanding of problems that do not require descriptions of interactive non-linear processes (*Ref. 36*).
- (2) Eulerian models employ extensive gas and aqueous chemical mechanisms and explicitly track concentrations of numerous species. Usually these models contain modules designed to calculate explicitly the chemical interactions that move gas-phase species into and among the various aqueous phases within clouds while determining the aqueous-phase chemical transformations that occur within clouds and precipitation droplets (*Ref. 37*).
- (3) Mass balance models are mathematical descriptions of the environment used to gain a quantitative and qualitative understanding of the behavior of mercury species throughout different media (e.g. air, soil, water). These models subdivide the environment into compartments or boxes, which are frequently assumed to have homogenous environmental characteristics and concentrations. The models then calculate how mercury species are distributed within that simplified system.
- (4) Hybrid receptor-deposition modeling techniques combine Lagrangian models with physical and/or empirical process models in order to assess the relative contribution of atmospheric sources to air masses crossing a particular monitoring site. They are suitable for regulatory purposes when continuous and relatively spatially dense monitoring network data are available (*Ref. 38*).

Air quality models have been developed on the global, regional and local scale. Local scale models are used to predict concentrations and deposition fluxes downwind of point sources. Regional and global scale models allow the simulation of long-range transport and atmospheric fate of

mercury. This permits the establishment of source-receptor relationships over some distance, up to a continental basis.

4.1 Modeling Mercury on a Global and Continental Basis

4.1.1 Global/Regional Atmospheric Heavy Metals Model (GRAHM)

Elemental mercury has been shown to have a one to two year residence time in the atmosphere, making mercury a global pollutant. A global scale model thus seems appropriate to address questions such as mercury budgets, long-range transport, transboundary exchanges and polar pollution related to mercury in the atmosphere. The Meteorological Service of Canada (MSC) has in the past years developed a high resolution Global/Regional Atmospheric Heavy Metal model (GRAHM). GRAHM is a eulerian multiscale model used to investigate atmospheric mercury at a global scale.

Dr. Ashu Dastoor from the Meteorological Services branch of Environment Canada presented the outputs from this model during the workshop. The model solves dynamic equations for all meteorological and physio-chemical processes for mercury species. Global anthropogenic emissions of mercury for the year 1990 available from Global Emission Inventory Activity (GEIA) provide the source component. The model is also integrated for multi years to respond to some of the questions related to mercury cycling in the atmosphere, and incorporates both atmospheric and mercury emissions data. After two and a half years of running this model, a seasonal cycle of mercury deposition is observed. The main features of the cycle are the high concentrations observed over land areas during the winter and over water masses in the summer. However, the scope of this conclusion is limited because volatilization is not taken into consideration by the model and the data are for northern sites only. Precipitation and boundary layers differ from season to season as well. In **Figure 17**, low surface air concentrations of mercury are observed over the Atlantic Ocean during the winter. In **Figure 18**, higher concentrations are detected in the summer, with a decrease over certain land areas.

The model provides source-receptor schemes, allowing the identification of major contributors for specific regions. Year 1990 data determined that, during the winter months, the Arctic is primarily affected by the emissions of total mercury from European sources, while during the summer months, the contribution from the Chinese mainland predominated. The vertical profiles over North America are considered indicative of source regions; at higher levels in the atmosphere, the contribution comes from European and Chinese sources, at the lower levels the sources are local and regional (**Figure 19**).

Dr. Dastoor also presented figures depicting source region contributors of wet and dry deposition of elemental mercury

(Figures 20 and 21). For Canada, total wet deposition of mercury in the year 1997 was mainly from Europe and Japan (51 percent), with only 28 percent from North America. For the United States, North America is the main contributor of total wet deposition (52 percent), with Europe and Asia (largely China and Japan) accounting for 32 percent. For dry deposition a similar trend can be observed; the United States appears more affected by sources in North America (77 percent) than is Canada (44 percent).

This model is limited, as it currently does not consider photochemistry and lacks the capability to predict concentrations in the Arctic. The lack of kinetics research and the uncertainty associated with the natural and recycled emission data contribute to difficulties in predictions in the Arctic and elsewhere. However, the model is a useful tool for sensitivity analysis, raising important issues and inferences and placing bounds on possible policy applications.

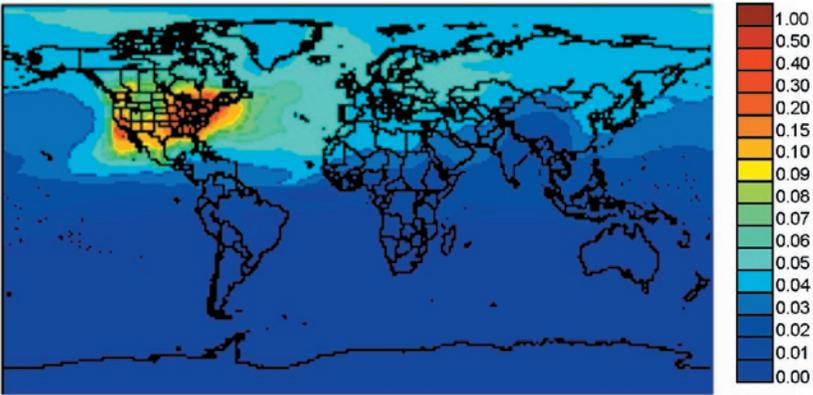
4.1.2 Chemical Transport Model (CTM-3D)

Dr. Christian Seigneur from Atmospheric and Environmental Research Inc. (AER) presented the Chemical Transport Model (CTM), a modeling exercise undertaken in collaboration with the Electric Power Research Institute (EPRI). The model makes distinctions between two types of mercury—elemental mercury and reactive gaseous mercury (RGM). Elemental mercury, given its widespread transport, is dispersed globally; for Hg(II) the local point sources are more dominant and the deposition of this form of mercury appears to be largely local. However, transformation mechanisms allow for elemental mercury to become RGM. The model uses parameters similar to those used in the model presented by Ashu Dastoor and is used for computation schemes at a global, continental and sub-continental scale.

4.1.2.1 Global Scale

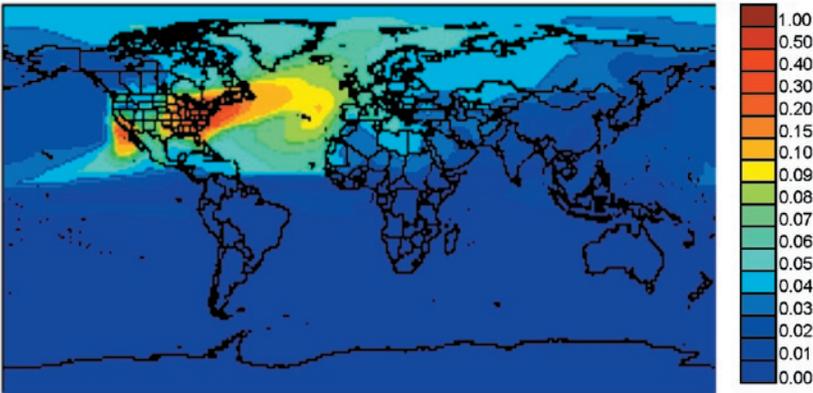
Dr. Seigneur’s 3-D global chemical transport model uses meteorology from National Aeronautics & Space Administration Goddard Institute for Space Studies (NASA GISS) general circulation model and data on emissions of mercury species developed by At-

FIGURE 17. Elemental mercury average surface air concentration (ng/m³) contributions from North America for January 1997



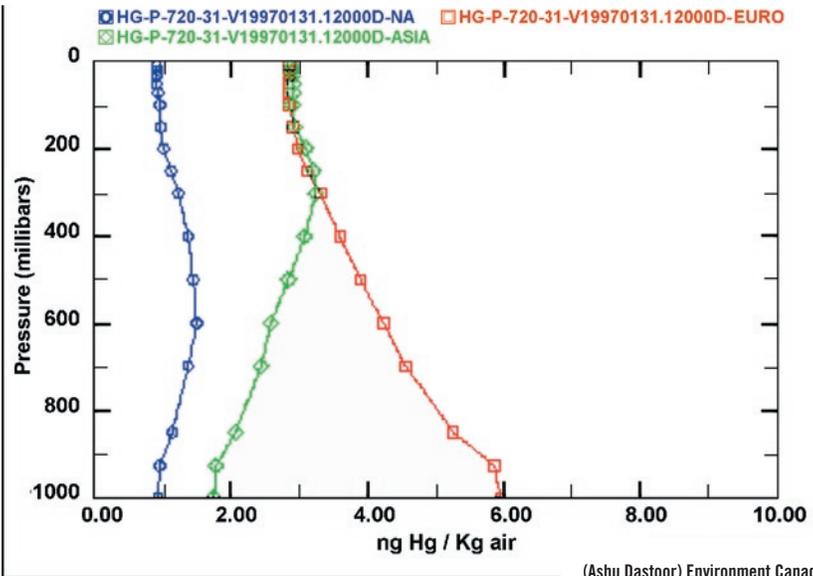
(Ashu Dastoor) Environment Canada

FIGURE 18. Elemental mercury average surface air concentration (ng/m³) contributions from North America for July 1997



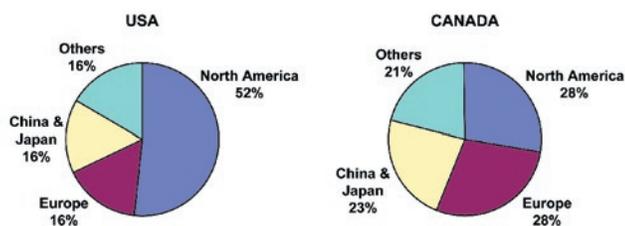
(Ashu Dastoor) Environment Canada

FIGURE 19. Vertical Profile of Mercury over North America, January 1997



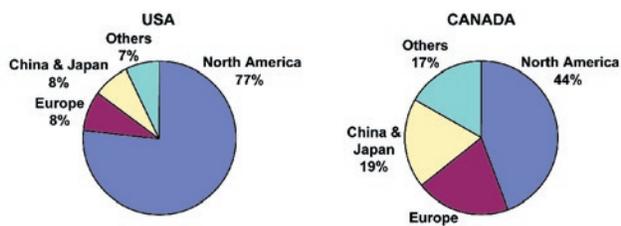
(Ashu Dastoor) Environment Canada

FIGURE 20. Total wet deposition of mercury - Contribution from various source regions, 1997



(Ashu Dastoor) Environment Canada

FIGURE 21. Total dry deposition of mercury - Contribution from various source regions, 1997



(Ashu Dastoor) Environment Canada

ospheric and Environmental Research Inc (AER) based on Global Emission Inventory Activity (GEIA) and Pacyna et al (year 2001). Reactant concentrations are obtained from CTM outputs. In **Figure 22**, global modeling of elemental mercury concentration in the surface layer (ng/m^3) is represented and in **Figure 23** concentrations from global modeling of divalent mercury ($\text{Hg}(\text{II})$) are given in pg/m^3 . For elemental mercury a south to north gradient was observed, with a range of concentrations oscillating between 1.2 and 1.6 ng/m^3 in the south and 1.6 to 1.7 ng/m^3 in the northern hemisphere. For divalent mercury, the pattern is different since this form is deposited relatively close to the sources. Spatial gradients are much stronger, with the northeastern United States, South Africa, Europe and Asia having significantly higher concentrations than those over the ocean.

FIGURE 22. Global modeling of mercury (Hg^0) concentration

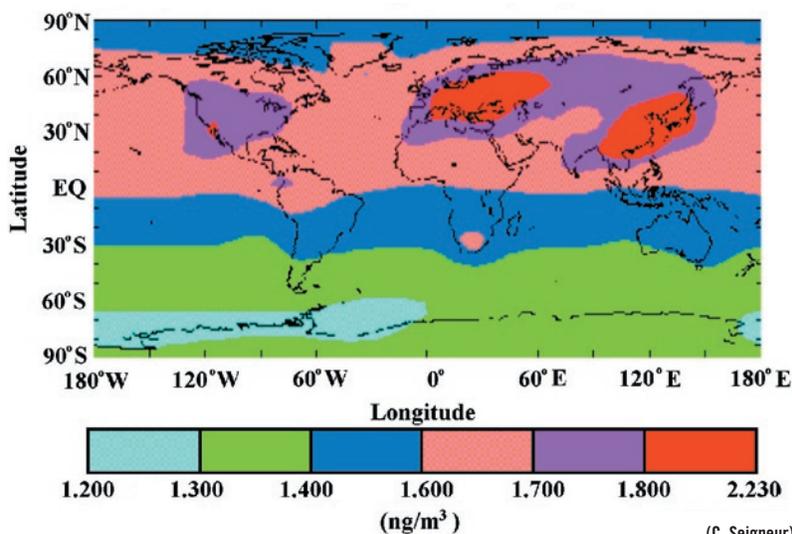
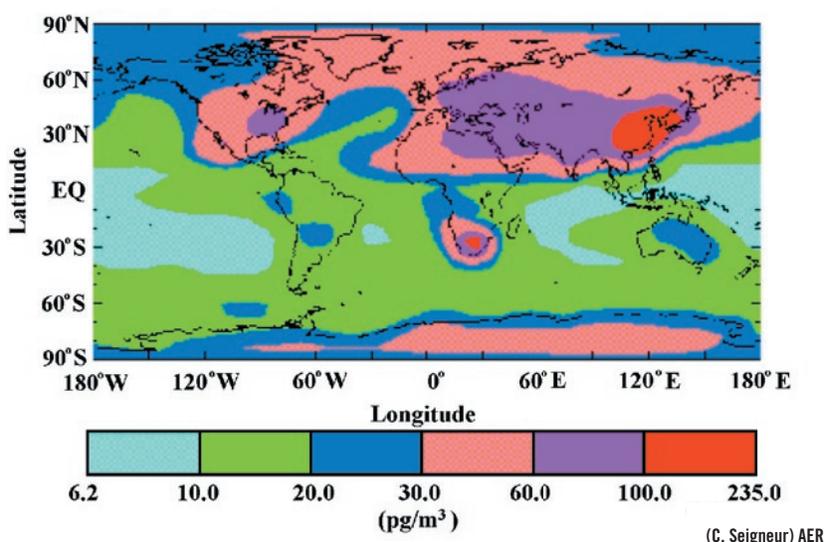


FIGURE 23. Global modeling of mercury ($\text{Hg}(\text{II})$) concentrations



When predictions were compared to measurements made on the west coast of Ireland in the year 1995, elemental mercury was in good agreement with observed data (1.6 ng/m^3 observed for 1.4 ng/m^3 predicted on average). Divalent mercury predictions were comparable, despite being at the low end of the range of measurements. Particulate mercury was also calculated, with the model predicting five (5) pg/m^3 , and the observed data averaging four (4) pg/m^3 . Seigneur pointed out that comparing the model output and measured data was important because output of global predictions was used as a domain for the continental modeling exercises.

4.1.2.2 Continental Scale

The 3-D Continental/Regional Model used a CTM with a 100 km (~ 62 mi) horizontal resolution. The meteorology parameters were based on NOAA Nested Grid Model (NGM) and precipitation data from the National Center for Atmospheric Research (NCAR), NADP/MDN and

the Meteorological Service of Canada for the year 1998. The use of actual measurement data for precipitation in place of a precipitation model eliminates uncertainties associated with summer convective precipitation. Emissions of mercury species data are those developed by AER for the United States, Canada and Mexico (Power plant emissions are those from

EPRI based on the year 1999 USEPA’s Information Collection Request (ICR)). The reactant concentrations were obtained from global CTM outputs and available data, and the boundary conditions from the global model.

The Continental model showed that most emissions were in the eastern part of the United States and not in Canada. The prevailing westerly winds were crucial to the anticipated results of increased dry deposition occurring downwind of the source area. In the context of mercury II, the highly reactive form, dry deposition occurs close to the source area. Thus the model predicted increased dry deposition over the northeastern United States and almost no dry deposition over the western United States. As one moves east from Minnesota and Wisconsin, dry deposition is predicted to increase (Figure 24). Because no dry deposition network exists, the model’s predictions could not be compared to actual measurements.

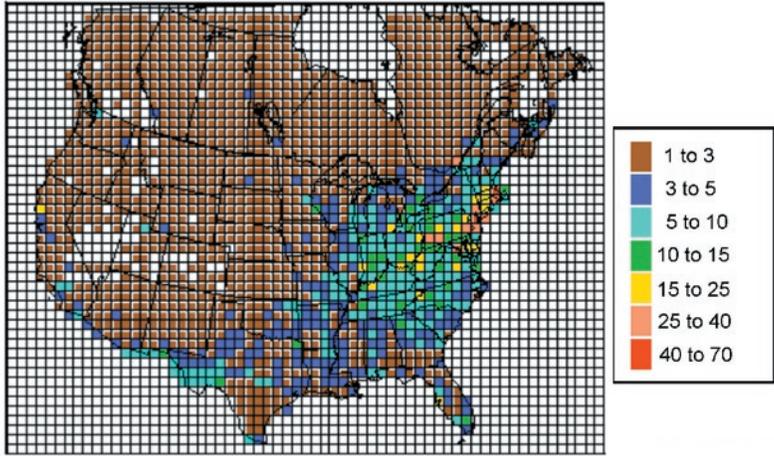
The pattern is different for wet deposition, largely due to oxidation of elemental mercury to reactive mercury, as well as some reduction reactions in droplets; thus wet deposition is impacted by cloud effects, precipitation effects and mercury chemistry. Mercury wet deposition is driven by precipitation, which is high throughout the northeastern and northwestern United States. As a result, spatial gradients for wet deposition are not as pronounced as for dry deposition, without the clustering presented in the dry deposition. Nevertheless, values in the northeastern United States were somewhat elevated due to a combination of high precipitation events and proximity to sources. Elevated values in Florida were also observed as a result of high precipitation during the summer.

When compared to MDN measurements, model predictions for wet deposition fluxes (Figure 25) were mostly in agreement. However, for the states of Wisconsin, Minnesota and Pennsylvania, the range of observed concentration values was 8-9 ng/m³ compared to a 5-10 ng/m³ range for predicted values. In order to understand these discrepancies, a finer resolution was used in a subcontinental simulation.

4.1.2.3 Subcontinental Modeling of Mercury

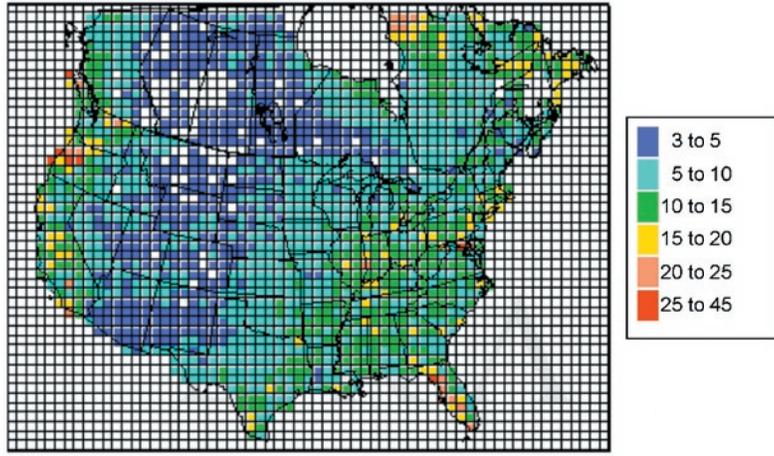
Subcontinental modeling was done with a 3-D regional CTM with 20 km (~ 12.5 mi) resolution. The meteorological, emission and chemistry inputs are identical to the continental model. The boundary is again the same as the

FIGURE 24. Continental modeling of mercury dry deposition (µg/m² yr)



(C. Seigneur) AER

FIGURE 25. Continental modeling of mercury wet deposition (µg/m² yr)



(C. Seigneur) AER

continental model. The continental model is of relatively low resolution, i.e. the deposit in the northeastern United States was shown to be the same on average, but the model predicted larger variations. With subcontinental modeling patterns, the results appeared to be more credible. Minnesota and Wisconsin values were in agreement with measurements but Pennsylvania values were higher by a factor of two when compared to observed data. Thus, when the resolution is increased, the performance of the model also improves but some discrepancies still remain. In the case of Pennsylvania, the model picks up emissions from the Ohio Valley, where coal-fired utilities operate, emitting divalent mercury (~50 percent of total emissions), and predicts deposition downwind in the state. However, MDN measurements do not correlate with these predictions, their values being lower. Seigneur put forward three hypotheses to explain this discrepancy: the measurement could be wrong, or either emission speciation or the chemistry in the model were incorrect. Assuming the monitoring data were correct, emissions of divalent mercury in the model were divided by four. This reduced the calculated values, but they still did not correspond

with the MDN measurements. Seigneur suggested that some mechanism might be taking place between source and receptor converting some of the Hg(II) into Hg(0), but this cannot be confirmed since the knowledge of the chemistry and kinetics of mercury is currently inadequate.

The global and continental models of atmospheric mercury provide good reproductions of the major spatial patterns of mercury concentrations and deposition fluxes. However, the subcontinental modeling suggests that the finer spatial gradients of mercury deposition are not reproduced correctly. Inspection of MDN data suggests a hypothesis that some mercury II emissions are reduced to elemental mercury before being removed by precipitation. Seigneur emphasized that the lack of information on mercury chemistry results in serious data gaps at the regional and local levels (as shown by the Pennsylvania case). If qualitative statements can be made, for instance, determining the importance of Asian emissions in North America, as presented by Dr. Dastoor, no quantitative statements can be made yet with certainty. Thus, more funding is required to better understand mercury kinetics chemistry.

4.1.3 Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model - Great Lakes

Mercury contamination of the Great Lakes basin remains a serious environmental concern, and atmospheric deposition appears to be a significant loading pathway. However, information regarding the relative importance of different source sectors and source regions has not generally been available (Ref. 39). Dr. Mark Cohen from the NOAA Air Resources Laboratory presented a review of the Atmospheric Transport and Deposition of Mercury to the Great Lakes project.

The overall goal of the project is the development of atmospheric mercury source-receptor information for each of the Great Lakes, the Gulf of Maine, and other selected receptors, capable of responding to the following questions:

- What are the relative contributions of different source *regions* (e.g. local, regional, national, continental, global...) to atmospheric deposition to any given locale?
- What are the relative contributions of different source *categories* (e.g. coal combustion, waste incineration, metallurgical processes) to atmospheric deposition at any given receptor?

To accomplish this task, the NOAA HYSPLIT model has been modified to simulate the fate and transport of mercury emitted to the atmosphere from sources in the United States and Canada. The analysis year chosen is 1996, although additional years could be analyzed in future work. The HYSPLIT NOAA model uses a bilateral emissions database developed

from the principal national databases of the USEPA and Environment Canada, augmented to the extent possible with Mexican data.

In comparison with Canada, per capita emissions are much higher in the United States but there is some uncertainty in the estimates. There are distinctly different emission estimates in the "Other" coal combustion category in the EPRI (Electrical Power Research Institute) data as compared to the USEPA data. This discrepancy needs to be resolved. In some cases, estimated speciation information has been added to the inventory, in order to attempt to develop reasonable estimates necessary for the modeling exercise. Meteorological data computed by an external model (e.g. National Centers for Environmental Predictions (NCEPs) NGM model) are used to drive the dispersion simulation. The modeling analysis is evaluated by comparison against available ambient monitoring data. Data from several locations within the modeling domain primarily for mercury in wet deposition (Ref. 40) are available for this evaluation. Dr. Cohen summarized the project methodology:

- begin with atmospheric mercury emissions inventory,
- perform atmospheric fate and transport modeling of these emissions using a modified version of NOAA's HYSPLIT model,
- track source-receptor information during the modeling,
- evaluate the modeling by comparison of the simulation's predictions against ambient monitoring data. If the model performs satisfactorily, report source-receptor linkages from the simulations (similar to earlier modeling work with dioxin and atrazine).

The HYSPLIT model uses puff motion tracking techniques to produce outputs. Theoretical puffs are released every seven hours or so and tracked throughout the year. Multiple puffs are released from one source, and the technique is then extended to several sources. Thus there are different track puffs through the atmosphere dispersed along a center line, taking into consideration all chemical mechanisms and dry and wet deposition. The advantage of such a technique is that source and receptor information is more easily compiled; however, there is no capacity to accommodate non-linear chemistry. The model uses a full chemical scheme but there is a good deal of uncertainty in the characterization of many of the reactions. More studies of gas phase reactions and thermal chemistry are needed.

The model can be used to estimate the impact of each source sector, and, in some cases large individual sources, on any given receptor and indicates that deposition frequently appears associated with long-range sources and is thus a regional phenomenon, not just due to sources adjacent to any particular Great Lake. The size of the region of influence differs for each lake. For example, the bulk of mercury deposition to Lake Superior (see Figure 26) is due to sources

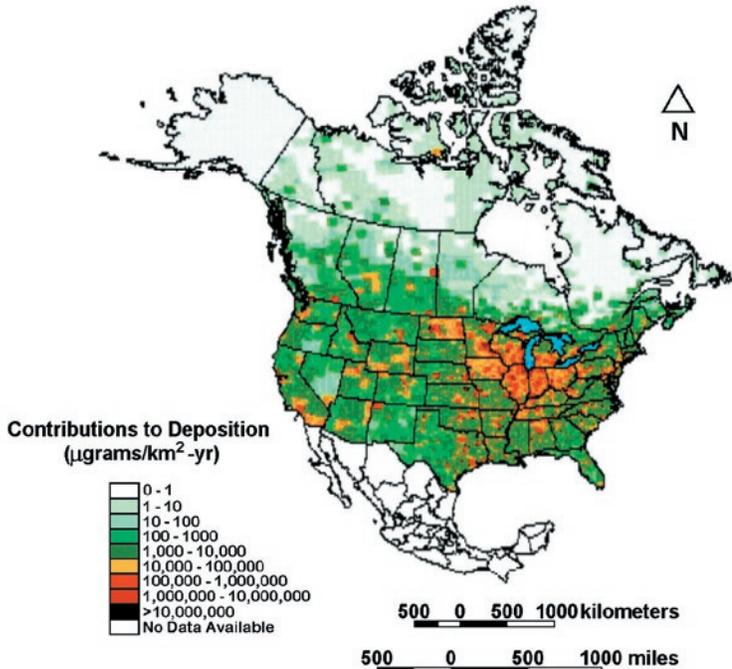
400 km (~ 249 mi) away, whereas for Lake Michigan, (see Figure 27) a great majority of the significant sources to the lake are less than 100 km (~ 62 mi) away.

It is also possible to comment on the relative importance of each source or source sector (coal-fired utilities, metallurgical processing, etc.) and although this differs again for each lake, fuel combustion sources are important to deposition in all lakes.

The results from this model are considered to be preliminary for a variety of reasons. Concerns remain about the accuracy of the U.S. and Canadian emissions inventories used. Additional source regions must be considered, particularly in Mexico, and the global contribution must also be estimated. Extensive model evaluation must continue, including verification against ambient mercury measurements. An additional effort is necessary to ensure that the model is accurately simulating the behavior of atmospheric mercury (wet and dry deposition algorithms; chemical reaction mechanisms and rates; atmospheric phase partitioning). Extensive sensitivity analyses must be performed to evaluate the influence of different modeling uncertainties. For this initial demonstration, only a limited number (28) of “standard source locations” were used; many more will be utilized in the more complete analysis to come. Nevertheless, these preliminary results show that the analysis appears to be consistent with available data and the output can be considered *an initial assessment of the mercury source-receptor relationships* for atmospheric deposition to the Great Lakes and the Gulf of Maine

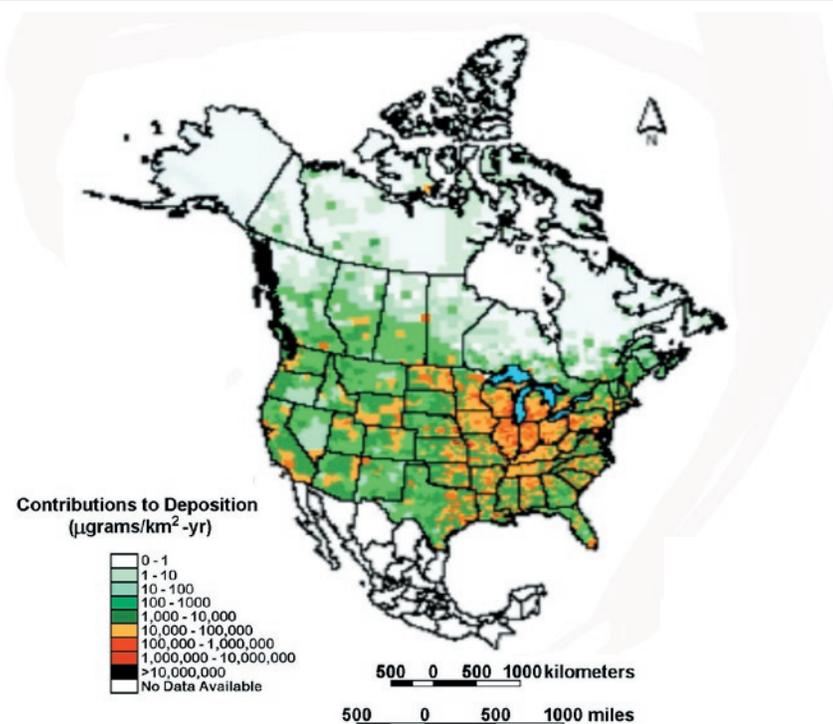
When asked about incorporation of information from Mexico into the model, Dr. Cohen replied that further source information had been received and was presently being processed for inclusion in the next iteration for the model. Dr. Cohen also addressed concerns related to the linearity of the Lagrangian technique. The model is predicated on treatment of the many mercury reactions in the atmosphere as first order reactions giving rise to linear relationships. This assumption may prove to be inadequate in addressing the actual nature of the reactions. The complexities of the atmospheric chemistry, including the presence of other

FIGURE 26. Estimated contribution to the atmospheric deposition of mercury to Lake Superior ($\mu\text{g}/\text{km}^2\text{-yr}$)



(M. Cohen) NOAA

FIGURE 27. Estimated contribution to the atmospheric deposition of mercury to Lake Michigan ($\mu\text{g}/\text{km}^2\text{-yr}$)



(M. Cohen) NOAA

chemicals, may not be adequately captured by first order linearity. The possible interaction between sources is also being considered, but that interaction will likely prove insignificant.

4.1.4 Acid Deposition Oxidation Model (ADOM) - European Application

Dr. P.K. Misra of the Ontario Ministry of the Environment (OMOE) presented the Acid Deposition Oxidation Model (ADOM), sharing the results of the different modeling simulations the OMOE undertook. The ADOM Eulerian model, which was originally designed for transport and deposition of acidifying pollutants and photochemical oxidants, has been modified to examine the physico-chemical processes of mercury in the atmosphere, especially for cloud mixing, scavenging, aqueous phase chemistry, transport and wet deposition. The model includes elemental mercury, particulate mercury and divalent mercury species. Dr. Misra outlined the critical elements of the model; these include an emissions inventory detailing speciation, and natural and re-volatilization emissions which are a function of temperature. The chemistry, dry deposition rates for elemental mercury, and regional models of background concentration are also crucial. The mass transfer, chemistry and adsorption component of the model is illustrated in **Figure 28**.

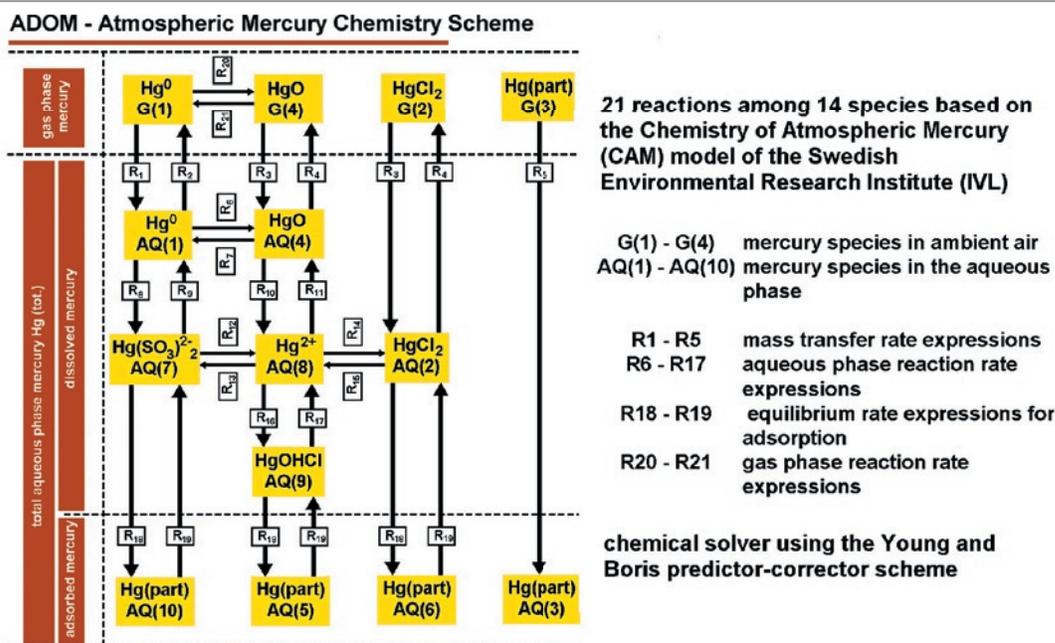
This model has been applied in some preliminary model simulations over eastern North America using the best available information on scavenging and dry deposition processes for elemental, divalent and particulate mercury. Dr. Misra explored the uncertainties involved with each species in terms of wet and dry deposition. Uncertainties exist in

terms of dry deposition rates and heterogeneous chemistry for elemental mercury. For mercury chloride, uncertainties include emission speciation and the extent to which mercury chloride is attached to particles in the stack plume, resulting in particle bound mercury. Finally, for particulate mercury, emission speciation and plume chemistry remain unclear.

The model was run for a 31 day period in August 1988. Modeled concentrations of elemental mercury and wet deposition were in the range of observed data, as were those for mercury chloride. However, particulate mercury was not in the range of observed values when compared to available data. Although the results from the North American simulation show discrepancies with the observed data, the deposition patterns are still well represented. Dr. Misra mentioned that, with the availability of a new inventory, the OMOE intended to run additional simulations in the future.

In collaboration with Germany, under the Canada-Germany Science and Technology Cooperation Agreement, the model was also applied to Europe. The database for anthropogenic emissions was drawn from emission inventories for the years 1990 and 1995, compiled respectively by the Umweltbundesamt in Germany (Year 1994) and the Norwegian Institute for Air Research (Pacyna et. al, NILU 2000). In the year 1990, total European emissions of mercury were significantly higher than those in the year 1995 (463 tonnes (~ 510 tons) per year compared to 327 tonnes (~ 360 tons) per year); however, the year 1995 mercury chloride emissions were four times higher than those for the year 1990. Particulate mercury emission data are comparable between the two inventories.

FIGURE 28. Mass transfer, chemistry and adsorption component of the ADOM model



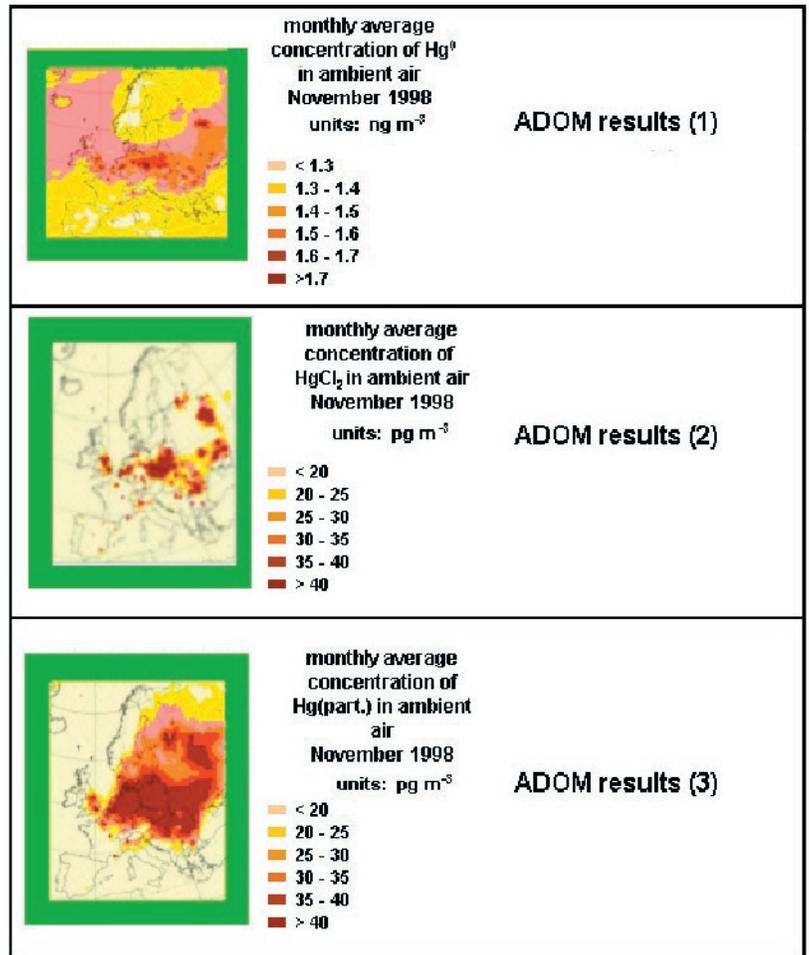
The output of the model showed calculated elemental, divalent and particulate mercury concentration and deposition patterns over the month of November 1998 (Figure 29 (1), (2), and (3)). Concentration values for elemental mercury appeared to be slightly higher than the background values. Mercury chloride values were reasonable, in the same range of the measurements performed in North America, and particulate mercury values seemed credible.

When compared to values observed in Europe, the elemental mercury model predictions appeared to be more accurate when using year 1990 inventory rather than year 1995, especially for a specific period of time (Figure 30).

This highlights the fact that the accuracy of emission inventories plays an important role in model simulation, affecting results.

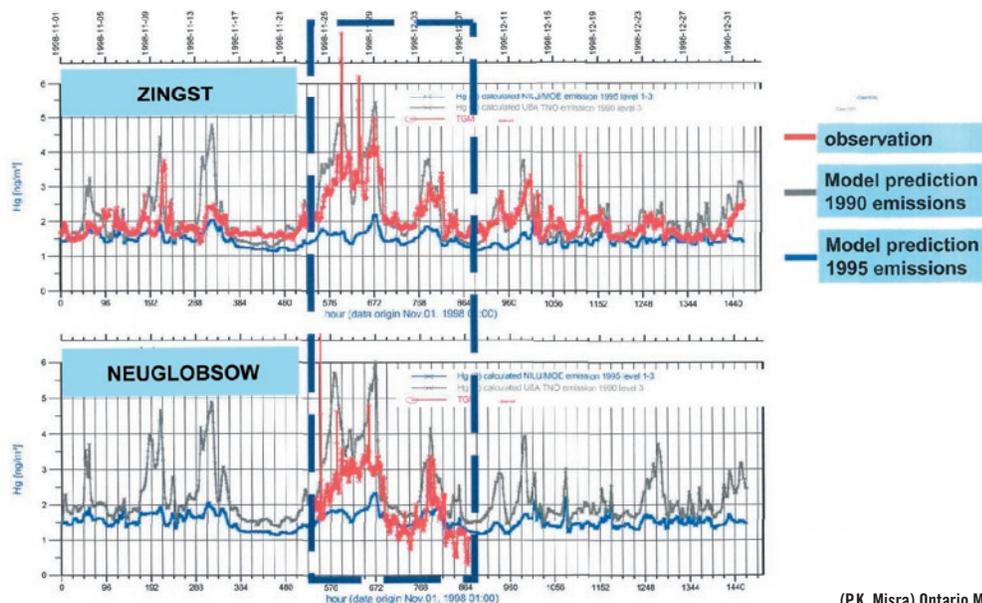
Dr. Misra also noted some discrepancies between model predictions and observed mercury concentrations in the atmosphere at the German monitoring stations of Zingst and Neuglobsow and Swedish sites of Aspverten and Rorvick (Figures 31 a and b). For elemental and particulate mercury the model predicted and the observed hourly averages are in reasonable agreement. However, predictions and observations for reactive gaseous mercury show discrepancies, with a severe under-prediction for the Zingst,

FIGURE 29. ADOM simulation results concentration for: (1) Hg⁰; (2) HgCl₂; (3) particulate Hg. Monthly averages in ambient air, November 1998



(P.K. Misra) Ontario Ministry of the Environment

FIGURE 30. ADOM results 2 (European version) (Comparison of observed and model predicted data)



(P.K. Misra) Ontario Ministry of the Environment

FIGURE 31a. Campaign averages of model predicted and observed reactive gaseous mercury (RGM) concentrations in ambient air at four measurement sites in Germany MOE 1- MOE 4. Units: pg/m³

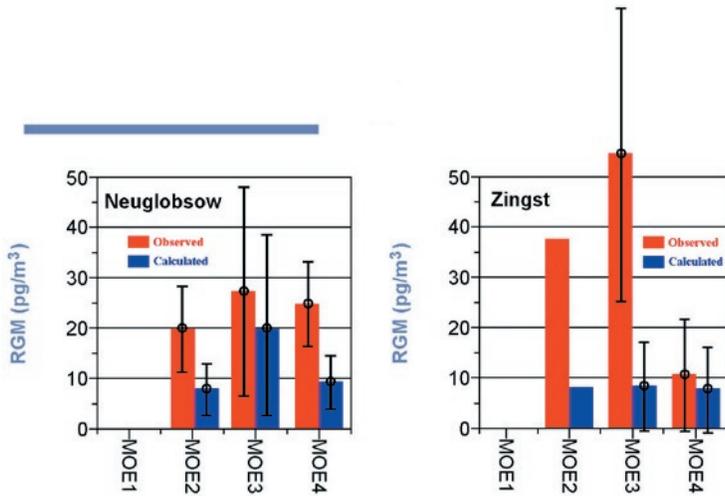
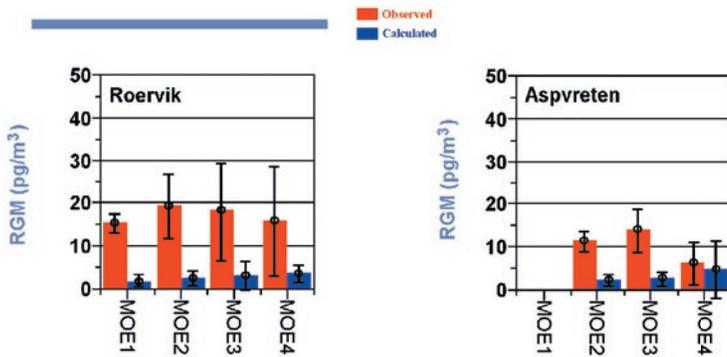


FIGURE 31b. Campaign averages of model predicted and observed reactive gaseous mercury (RGM) concentrations in ambient air at four measurement sites in Sweden MOE 1 - MOE 4. Units: pg/m³



(P.K. Misra) Ontario Ministry of the Environment

Aspveten and Rorvick sites. This difference in values could be attributed to the coastal location of these sites; the ocean may have an impact on the measurements. The measurements may also be inherently faulty; however, the limited data do not allow drawing of any firm conclusions at present.

Dr. Misra uncovered many questions about mercury speciation and models. Particulate mercury predictions were close to the observed emissions and similar to previous inventories. Reactive mercury predictions appeared to be reasonable near the source region; however, at distant coastal sites, predictions were much lower than observed measurements. Again, there is no clear rationale for this outcome; possibilities include volatilization from coastal waters, significant background concentrations of reactive mercury and the uncertainty of measurements. The mercury in precipitation predictions averaged over a month corresponded well with observed values.

Dr. Misra concluded his presentation by highlighting what he thought were the policy implications of the issues raised. Those are presented in Table 8.

4.2 Other Modeling of Mercury on a Sub-Continental Basis

4.2.1 Community Multi-scale Air Quality (CMAQ) Model

The USEPA is currently engaged in the development of several atmospheric mercury modeling systems. From the years 1993 to 1999 the Regional Lagrangian Model of Air

TABLE 8: ADOM Modeling: Possible Implications on Policy

Issue	Possible Implications
- Emissions of mercury chloride and particulate mercury are deposited locally and regionally	- Reducing these emissions in North America will reduce deposition - Uncertainties in emission speciation result in deposition uncertainty
- Emissions of elemental mercury add to the global atmospheric pool and could be deposited anywhere around the globe	- Emission reductions on a global scale are required - Where could the most cost effective reductions be achieved?
- Natural/re-volatilization emissions could be as large or larger than anthropogenic emissions	- Reductions in mercury deposition would be much less than reductions in emissions - Large reductions in anthropogenic emissions might be required to show in measured deposition data

(P.K. Misra) Ontario Ministry of the Environment

Pollution (RELMAP) was adapted to simulate emission, transport, dispersion, atmospheric chemistry and deposition of mercury across the continental United States. This model was used for USEPA's Mercury Study Report to Congress in the year 1997 and some subsequent investigations.

From the year 1999 to the present the Community Multi-scale Air Quality (CMAQ) model, originally applied to ozone, sulfur, nitrogen and particulate matter, has been adapted to the modeling of mercury and its various interacting physical and chemical reactions. The Models-3 CMAQ modeling system was created to integrate major tropospheric air pollutants in a multi-scale "one atmosphere" structure to be accessible to both scientific and air quality management communities. The CMAQ mercury model is a simplified single-volume version meant to simulate gas/liquid partitioning, cloud water chemistry and adsorption of mercury complexes to soot particles suspended in cloud water (*Ref. 41*).

Now, in addition to pre-existing CMAQ model chemistry, the CMAQ mercury model simulates two gas-phase reactions, eight aqueous-phase reactions, and six dissociation equilibria for mercury and mercury compounds. Anthropogenic emissions of mercury for the CMAQ-mercury model are based on an emission inventory developed at the USEPA's Office for Air Quality Planning and Standards and chemical/physical emissions speciation assumptions developed at the National Exposure Research Laboratory (NERL) (*Ref. 42*).

Russ Bullock from the NERL presented the CMAQ-mercury modeling system during the workshop, highlighting the simulation process modules present in the models:

- Horizontal and vertical advection
- Mass conservation adjustments for advection processes
- Horizontal and vertical diffusion

- Gas-phase chemical reaction solver
- Aqueous-phase chemical reactions and deposition
- Aerosol dynamics and size distributions
- Gas and aerosol dry deposition velocity estimation
- Plume chemistry effects

Mr. Bullock underlined the primary factors affecting the transport range of mercury emissions in the atmosphere, which include the chemical and physical forms of the emissions, surface dry deposition characteristics, emission plume characteristics, and chemical and physical reactions in clouds. The latter two require a high resolution eulerian framework for estimation.

The CMAQ-mercury model has been included in the International Mercury Model Intercomparison organized by the Meteorological Synthesizing Centre – East (MSC-East) under the sponsorship of the EMEP (European Monitoring and Evaluation Program). The goal of phase one of this model intercomparison was to review the nature and responsiveness of the various modules for physico-chemical transformations of mercury species in a cloud/fog environment with prescribed initial mercury concentrations in ambient air and other physical and chemical parameters relevant for atmospheric mercury transformations. A primary objective was to compare model results to gauge the levels of uncertainty and sensitivity and to assess if cloud-water concentrations were within the measured range of rain concentrations.

Modeling was executed under different test conditions, resulting in five cases. These test simulations show that a day/night oscillation of the aqueous mercury concentration occurs, driven mostly by the reaction of elemental mercury with chlorine (oxidation) during nighttime and reaction of mercury II with HO₂ (reduction) during daytime (*Ref. 43*). It was observed that if a significant amount of mercury is already present in a given cloud, additional mercury from

TABLE 9. CMAQ-Hg Model Intercomparison Initial Concentrations (Bullock)

	Case 1	Case 2	Case 3	Case 4	Case 5
Gaseous oxidized mercury in air (10 ⁻⁹ g/m ³)	0	0	0	0.01	0.01
Particulate mercury in air (10 ⁻⁹ g/m ³)	0	0	0.04	0	0.04
Aerosol soot concentration in air (10 ⁻⁴ g/m ³)	0	0.5	0	0	0.5
Elemental mercury in air	1.7E-9 g/m ³				
Elemental mercury in water	0				
Oxidized mercury in water	0				
Mercury absorbed on soot particles in water	0				
Sulfur dioxide in air, SO ₂	1.0E-6 g/m ³				
Sulfite ion in water, SO ₃	0				
Ozone in air, O ₃	7.0E-5 g/m ³				
Ozone in water, O ₃	0				
Hydrogen peroxide in air, H ₂ O ₂	100 ppt				
Hydrogen peroxide in water, H ₂ O ₂	0				
pH of cloud water	4.5				

emissions may not increase the concentration of mercury already present in the cloud in a linear manner, suggesting that the kinematics of the associated reactions may not be linear. This was seen in Case 3 where mercury concentrations were higher at the beginning of the testing (Table 9).

By the end of the simulation, Case 3 results appeared to be developing an oscillation of dissolved mercury concentration similar to Case 1. Russ Bullock suggested that if a significant amount of mercury is already present in the cloud water, additional mercury does not significantly increase the mercury concentration in the cloud water. Both Case 1 and Case 3 have no soot and mercury concentration oscillates between 20 ng/L at sunrise and 5 ng/L at sunset. When soot is present (Case 2 and Case 5), there seems to be a moderate reduction of mercury during daytime and no significant effect on oxidation of elemental mercury during nighttime, leading to greater concentration of mercury in cloud water (Figure 32).

From the MSC-E work, the cloud water model agrees with other similar models within a factor of two, but questions remain about the realism of the strong diel cycle.

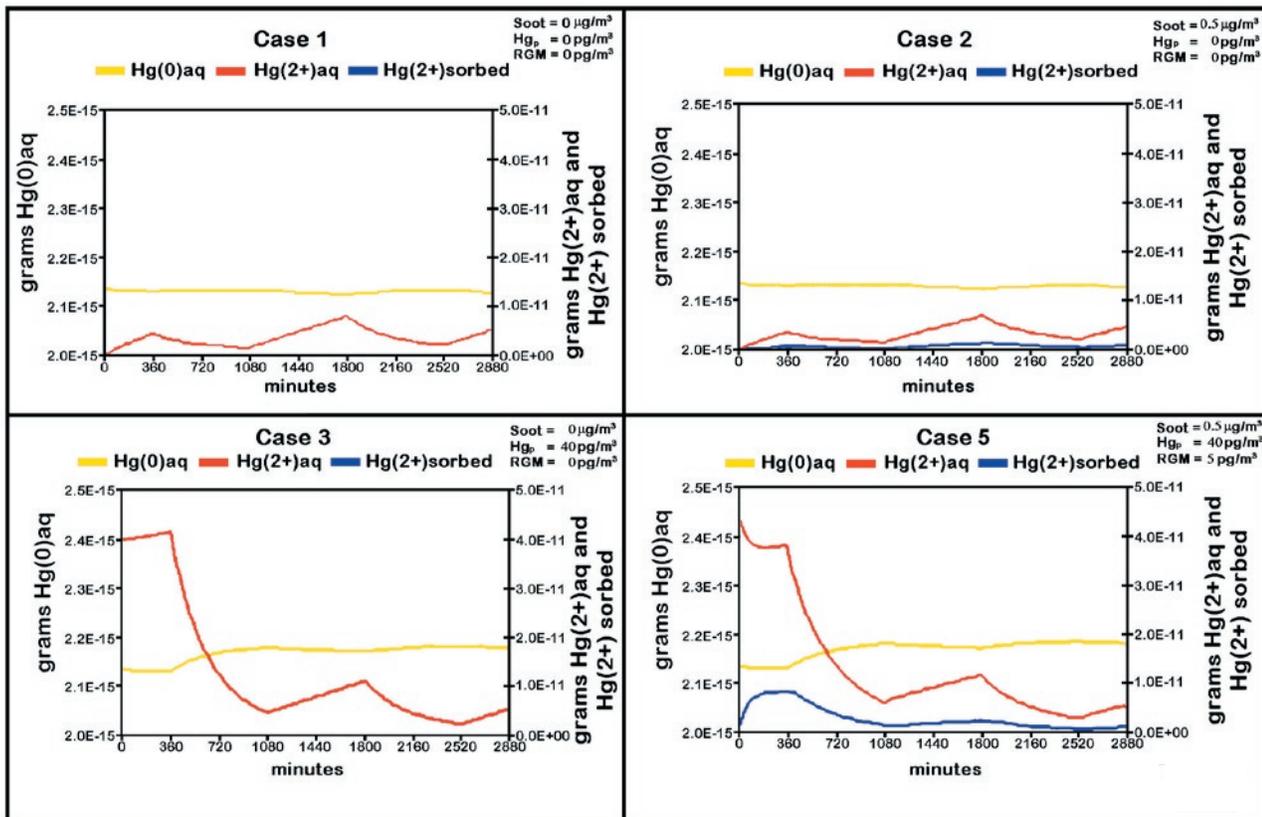
The full-scale CMAQ model was also evaluated with simulations performed for two four-week test periods in the year 1995 (April 4 – May 2 and June 20 – July 18). The model resolution was 36 km (~ 22 mi) horizontally and 21

layers vertically. Derived meteorological inputs were already available along with ozone, sulfur, nitrogen, and particulate inputs. Mercury emissions data were taken from the 1997 U.S. Mercury Study Report to Congress and the domain was the southeast United States. Simulated wet deposition of mercury was compared to weekly observations from the Mercury Deposition Network (MDN). The model estimates and the wet deposition data compared reasonably well during the spring but appear poorly related in the summer (Figure 33). In the summer, precipitation events were not accurately predicted, due to the occurrence of small spotty thunderstorms. It was concluded that this model is not suitable for the 36 km (~ 22 mi) range application during the summer.

Russ Bullock synthesized the implications of the intercomparison with the MSC-E exercise and the comparison with MDN data. They are presented in Table 10.

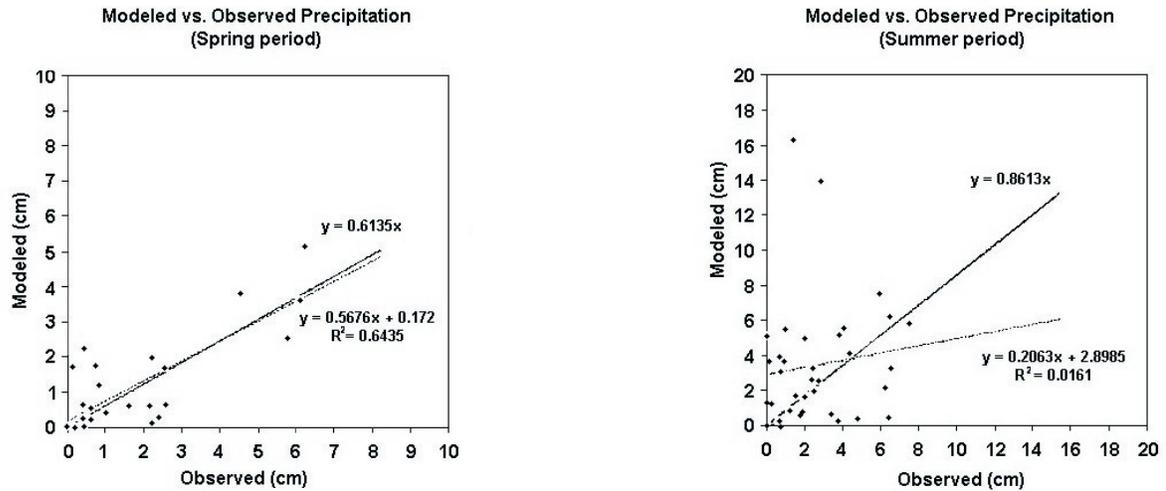
In addition, Russ Bullock pointed out that these modeling exercises produce many policy considerations. Natural emissions are not yet included in source inventories used for simulation modeling. Evaluation of the mercury dry deposition sub-model is not yet possible due to a lack of observational data. Thus, closure of the sink or receptor terms is not possible. With no complete closure on either the source or sink terms, model evaluations are based largely on conjecture. Such closure within the domain of

FIGURE 32. Simulation results for Cases 1, 2, 3 and 5



(R. Bullock) USEPA

FIGURE 33. CMAQ estimated wet deposition compared to MDN data for the spring and summer periods.



(R. Bullock) USEPA

TABLE 10. Implications of CMAQ-Hg Test Results

- From MSC-E work, cloud water model agrees with other similar models within a factor of two, but there remain questions about the realism of the strong diel cycle.
- Cloud water model produces total mercury concentrations that are within the range of observed precipitation values, but observational data on Hg(0) and Hg²⁺ species in cloud water under various conditions are needed to appraise model accuracy.
- Full-scale model results for wet deposition are strongly dependent on the validity of the precipitation definition.
- Model accuracy for mercury wet deposition is comparable to that seen in early NAPAP (Acid Precitation) sulfur modeling. Moderate accuracy in cool seasons, but less accuracy in warm seasons where convective precipitation dominates.
- More comprehensive field testing is certainly needed.

comprehensive field studies is needed, either through source and sink definition or boundary flux definition.

4.3 Modeling Mercury on a Regional Basis

4.3.1 Emission and Regional Scale Dispersion of Mercury in Eastern United States

Dr. Chris Walcek, from the State University of New York (SUNY) Albany, shared the initial results of a project funded by the New York State Energy and Research Development Authority (NYSERDA). Before presenting the project, Dr. Walcek began by identifying the current limitations in the science of modeling atmospheric mercury. When Air Quality Models (AQM) were developed for the study of acid rain and ozone depletion, they were considering a situation where the residence time of these pollutants in the atmosphere was limited compared to that of some forms of mercury. Elemental mercury, due to its residence time of up to a year in the atmosphere, poses a unique challenge for the modeling community by requiring higher and longer

term resolution, stretching the boundaries of models to a global scale.

Construction of a credible scientific atmospheric mercury model involves compromise. Indeed, the complexity of the modules and the amount of parameter data used to achieve accuracy are occasionally reduced to avoid computational difficulties. For example, in order to achieve global scale models, high resolution can be sacrificed. To avoid these shortcomings Dr. Walcek advocated the development of new models: because mercury poses new challenges, new AQMs should be designed to address them.

The NYSERDA-funded project has a number of objectives. Among them are the quantification of the effects of poor treatment of various parameters on calculations, especially when discrepancies between observed and predicted data are evident, as well as the evaluation of cloud micro-physics schemes in models. Investigating annual aggregation techniques applied to parameter data in order to assess the adequacy of statistical sampling of a number of events versus compiling them, and evaluating the accuracy of measurements are also among the goals. A detailed list of the objectives is given below:

1. **Test and evaluate a wide range of atmospheric mercury modeling approaches**
 - SKIRON/ETA Meteorological System
 - Regional Atmospheric Model System (RAMS-HG)
 - SUNY regional-scale-HG
 - Lagrangian approaches (HYSPLIT, etc.)
2. **Quantify the effects of “poor” or “parameterized” treatment of:**
 - resolution - coarse (global-scale) vs. fine resolution

- ozone, full radical chemistry vs. tabulated, seasonal chemistry
- alternate clouds and micro-physics schemes

3. Investigate annual aggregation techniques

- statistical sampling vs. brute-force annual simulation

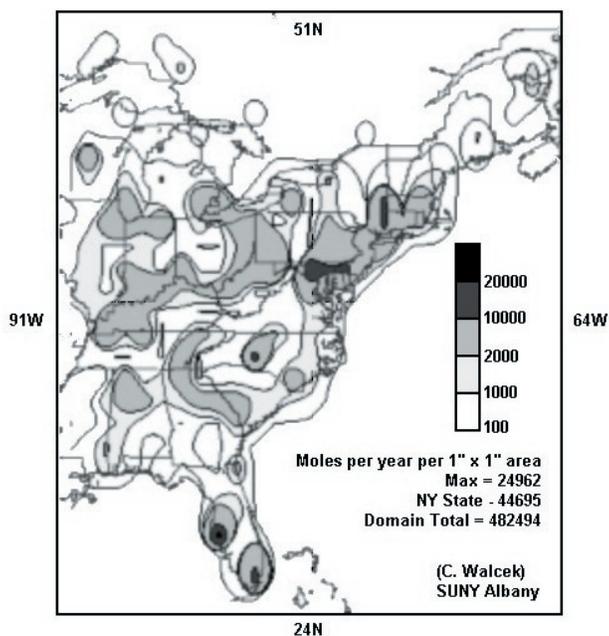
4. Evaluation and validation against available concentration and deposition measurements

At the time of the workshop, the project had only been operational for six months. Dr. Walcek's first activity had been the assembly of a new inventory for the northeastern United States from the USEPA data base, and simulations using two European regional scale models within a North American domain. Emissions data for the anthropogenic speciated mercury emission inventory were collected from numerous sources: USEPA National Toxics Inventory (NTI), the Canadian source inventory and EPRI estimates, all for the base year of 1996. A total of 5341 point sources were considered along with quality assurance/quality control stack or point source information, which included some missing and inconsistent parameters. Canadian and border state point sources were used in the model. A total of 1634 county reports were collected, and county centroids were used to locate emissions, which were included in the areal source estimate. The source categories were profiled for "representative" speciation using the following ratios:

Ratio of Hg(0):Hg(II gas):Hg(II particulate) – 48:35:17

Ratio of Hg(0):Hg((II gas) + (II particulate)) – 48:52

FIGURE 34. NYDEC-compiled Mercury Emission Inventory (prepared 2001; assesses 1996-1997 emissions)



(C. Walcek) SUNY Albany

Figures 34 and 35 show the emission map for the eastern United States based on a new inventory compiled by Walcek and an emission map compiled with the Global Emission Inventory Activity (GEIA) for the year of 1990. Significant discrepancies are apparent between the two inventories. With the new inventory, emissions are widely distributed, with highs in Washington D.C., the Hudson River in New York, the Ohio River Valley and in Florida. With the GEIA inventory, high emissions are concentrated in urban areas; there are no high levels observed in the Ohio River Valley and Florida is less prominent.

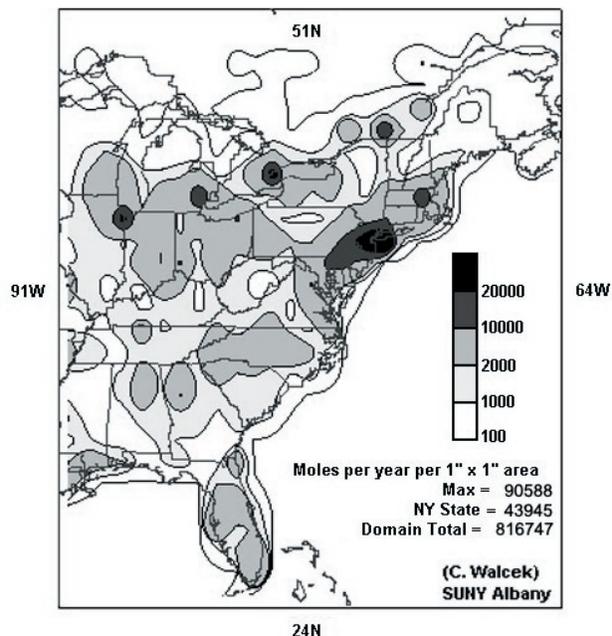
When a statistical analysis is performed (Figure 36), the new inventory emissions are only moderately correlated with the GEIA inventory. Several factors could explain such dissimilarity.

- Perhaps real changes in emissions occurred between early 1990s to the year 1996 (pollution control technologies).
- Most Canadian sources are missing from new inventory because of successful reduction since the year 1990.
- Maybe the largest source areas are not coincident.

However, the significant difference in geographical patterns is difficult to explain, even using demographic variations as a possible rationale. These discrepancies can have a significant impact on model's calculation, and their occurrence must be investigated if that impact is to be mitigated.

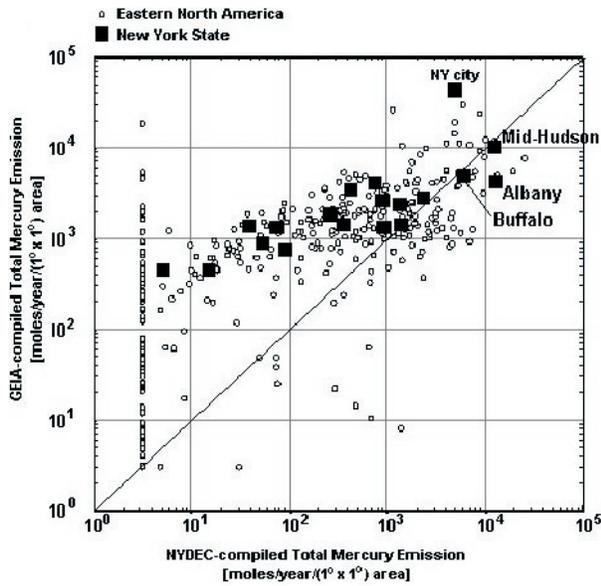
Dr. Walcek also presented a preliminary simulation done with two European regional-scale models within a North American domain by George Kallos, A. Voudouri, O. Kalkaliagou, J. Pytharoulis, E. Mavromatidis and P. Katsafados

FIGURE 35. GEIA Mercury Emission Inventory (prepared ~ 1996; assesses 1990 emissions)



(C. Walcek) SUNY Albany

FIGURE 36. Comparison of GEIA and NYDEC Mercury Emissions Inventories



(C. Walcek) SUNY Albany

of the University of Athens. The models used were two meteorological-dispersion models, the Regional Atmospheric Modeling System (RAMS) and the SKIRON/ETA meteorological system.

RAMS is a eulerian model developed jointly by Colorado State University and Mission Research Inc/ASTeR Division. It is a merger of a non-hydrostatic cloud model and a hydrostatic mesoscale model. The complexity of cloud micro-physics is recognized, allowing a focused examination of individual episodes. The SKIRON system consumes less computational time in comparison to RAMS because the treatment of cloud micro-physics is simplified. Developed at the University of Athens, this model is based on the ETA/NMC model, which permits a better consideration of step-like terrain because of a vertical coordinate alternate terrain treatment. The model is particularly suitable for longer term (annual) simulations.

The dual simulation was done for a 15-day episode for August 1997, with similar inputs for each model. The resulting RGM prediction maps show significant discrepancies: the RAMS model output concentrations range from 10 pg/m^3 to 30 or 40 pg/m^3 whereas the SKIRON/ETA model yields concentrations up to 100 pg/m^3 . For elemental mercury, the SKIRON/ETA model shows concentrations in urban areas three times higher than RAMS (up to 3.60 ng/m^3 compared to a maximum of 2.00 ng/m^3). Hg(II) wet deposition appeared to be within the range of the observed concentrations. These results are preliminary and Dr. Walcek pointed out that full treatment of the model outputs was still in the early stages. The significant differences between models could probably be explained by alternate physics modules in the models

(clouds, precipitation, chemistry). Nevertheless, this dual simulation indicates that further comparison of models, with a better understanding of the effects of the management of various parameters on calculations, is necessary.

Dr. Walcek concluded by presenting a summary of his findings to date, given in Figure 37.

FIGURE 37. Summary of Findings - Dr. C. Walcek

- Long atmospheric residence time forces compromises in model formulation
 - global scale vs. high resolution
 - episodic vs. annual average
 - O_3 , full radical chemistry vs. tabulated/simplified chemistry
 - clouds and microphysics
- Assembled and aggregated an eastern North America speciated Hg(0), Hg(II)g; Hg(II)p) mercury emission inventory
- Stack information nearly “useless”
 - (Stated information for 80% of point sources (~4000 srcs) grossly inconsistent)
- New inventory only moderately correlated with GEIA-Global emissions inventory
 - (overall emissions about 60% of GEIA-reported; NY State lower by 50%)
 - (maybe real changes early 1990s to year 1996?)
 - (some Canadian sources missing from new inventory?)
 - (largest source areas not coincident)
- “Dual” Simulation of North American mercury show “qualitative” agreement but appreciable discrepancies
 - SKIRON/ETA vs. RAMS-Hg Model Outputs
 - measurements inadequate to distinguish superior performance
 - alternate physics (clouds, precip-chemistry) probably dominant reason

4.3.2 Mercury Budget for Québec

Dr. Laurier Poissant, from the Meteorological Service of Environment Canada, Québec Region, presented his studies on total gaseous mercury (TGM), on processes of water-air and soil-air exchange in Québec, and mercury depletion events near the Arctic, giving an overview of the mercury budget for Québec.

In the year 1998, TGM concentrations were measured at four stations along the St. Lawrence River. The stations

FIGURE 38. The Canadian Atmospheric Mercury Network (CAMNet)



(L. Poissant) Environment Canada

were at St. Anicet and L'Assomption, located in rural areas within 100 km (~ 62 mi) of Montréal, and Villeroy and Mingan respectively, rural and remote areas further north (Figure 38).

The median TGM concentrations throughout the network varied from 1.62 to 1.79 ng/m³ (Table 11).

Station	TGM (ng/m ³)		
	Average	Min.	Max.
St. Anicet	1.77	0.91	10.9
L'Assomption	1.88	1.2	33.81
Villeroy	1.61	0.63	3.92
Mingan	1.61	0.33	2.87

Measurements confirmed that southern Québec was a source region for mercury, with high TGM concentrations compared to background concentrations. Going downwind, the concentrations decreased, indicating a sink of mercury. The TGM time series measured in rural and remote sites showed a significant decreasing spatial trend from St Anicet to Mingan by 11.5 percent (means difference of 0.18 ng/m³), illustrating this pattern. This large spatial variation in air and precipitation concentrations demonstrates that air masses located close to industrial and urban areas are burdened with larger mercury concentrations.

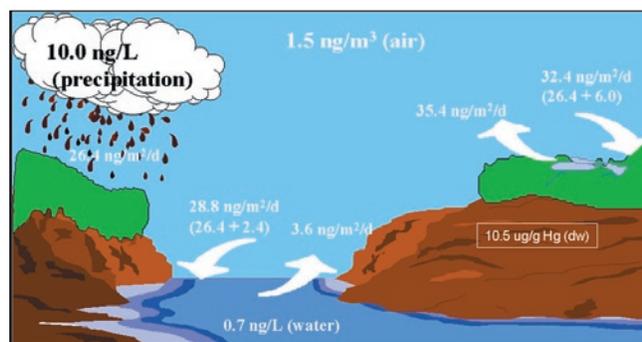
As mentioned in an earlier presentation by Dr. Pierrette Blanchard, Dr. Poissant also observed temporal patterns in St. Anicet and Mingan for mercury concentrations. There was a seasonal distribution, with highs in the spring and lows in the winter, most likely due to inefficient scavenging processes associated with cloud and droplet physics.

In his study on the processes of water-air and soil-air exchange in Québec, Poissant undertook an intensive field measurement campaign at two sites (land and water) in southern Québec. The terrestrial surface site location and the water surface site (on a quay) were both at St. Anicet, on the shores of the St. Lawrence River. Estimation of the regional mass balance of mercury in the upper St. Lawrence River showed that total mercury deposition over land is more or less in equilibrium with the evasive flux of mercury. On the river surface this equilibrium is not observed, suggesting that once mercury enters the water column it becomes available for biota or sediment uptake or downstream transport, and to a lesser extent, re-emission. These differences between air-water and air-soil exchanges are illustrated in Figure 39.

During the winter, mercury gas deposition is lower and the re-emission term is nearly equal to the gas deposition term (Figure 40).

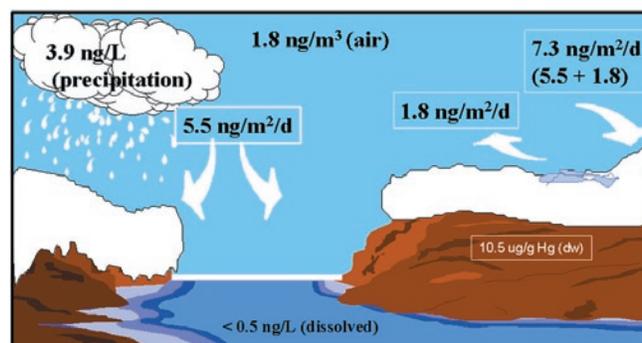
Poissant then presented measurements of total gaseous mercury and ozone concentrations achieved in Kuujjuarapik, in the lower Arctic region along the Hudson Bay (Québec). Two scenarios were tested: Case A, where complete scavenging/depletion events occurred, and Case B where ozone and mercury concentrations are average. Using a Global Ozone Monitoring Experiment (GOME) satellite, vertical column concentration of bromine monoxide (BrO) were measured. Relatively large mercury depletion events (MDEs) were observed during the presence of bromine monoxide clouds (Case A).

FIGURE 39. Mercury balance in the Upper St. Lawrence River Valley (Summertime)



(L. Poissant) Environment Canada

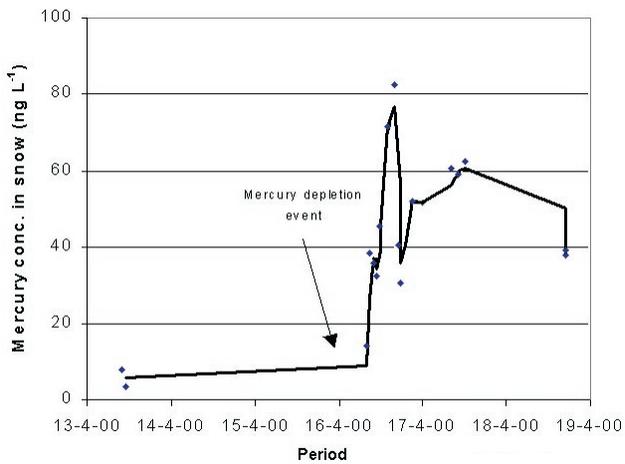
FIGURE 40. Mercury balance in the Upper St. Lawrence River Valley (Wintertime)



(L. Poissant) Environment Canada

Measurements also showed that mercury concentrations in snow increased in parallel to MDEs. Poissant presented the times series of mercury concentrations in snow during the spring of year 2000. Following depletion events, mercury concentrations in snow increased by about 16 fold within 24 hours. However, loss of mercury from snow pack following MDEs is important, with a removal rate of up to 50 percent in 12 hours (Figure 41).

FIGURE 41. Time Series of Mercury Concentrations in Snow at Kuujjuarapik (Spring 2000)



(L. Poissant) Environment Canada

It was also observed that total gaseous mercury flux increases by four times after depletion events suggesting that mercury fallout during depletion is very reactive and, when time series of mercury fluxes and net solar radiation are compared, photo-sensitive. Hence, a large portion of the mercury formerly deposited appears to be re-emitted back to the atmosphere by volatilization due to solar radiation. Air temperatures below 0° Celsius (32° F) and sunlight seemed to favour this process (Figure 42). According to Poissant, air temperature also plays a role in the removal

of mercury from snow surface by melting water. After one TGM depletion event, air temperature was warm (>0°C (>32° F)), suggesting that cloudy and warm conditions seemed to favour a loss of a large portion of the deposited mercury with melting water.

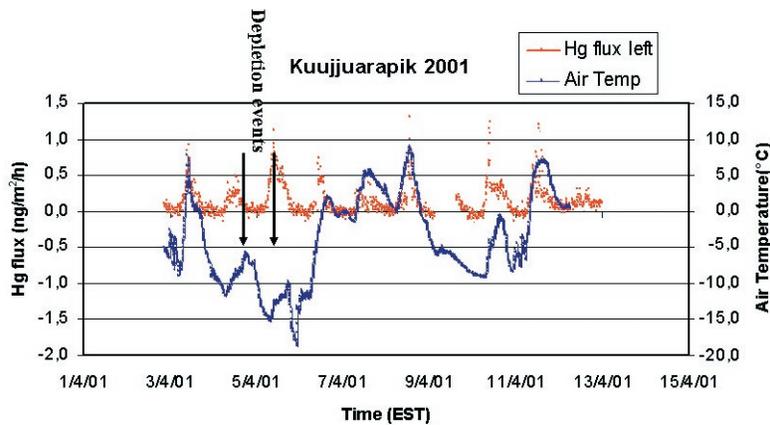
In light of this, Poissant concluded that mercury is removed from snow surface either through:

- re-emission (photo-reduction - air temperature below 0° Celsius (32° F) and sunlight); and/or
- with melting water (air temperature above 0° Celsius (32° F) and darkness).

Poissant concluded his presentation by outlining the main issues raised:

- There are anthropogenic sources impacting the Québec region (both regional and long range);
- A large spatial variation in air and precipitation concentrations within 5° parallel (45° N to 50° N) showed that air masses located close to industrial and urban areas have larger mercury burdens;
- Mercury processes are very complex (e.g. chemistry, gas exchanges) and further research is needed;
- It is important to have an integrated modeling scheme to address the complexity of mercury in the environment.

FIGURE 42. Time Series of Hg Fluxes and Air Temperature at Kuujjuarapik (Spring 2001)



(L. Poissant) Environment Canada

5.0 SCIENCE AND POLICY

5.1 Public and Other Interests: Invited Presentations

This session was intended to give an opportunity to members of the public outside the scientific community to examine the interaction between scientists and policy experts on the issue of mercury and recommend possible improvements to this interaction.

The speakers represented a wide range of stakeholders: Bruce Lourie from Pollution Probe, a Canadian environmental Non Governmental Organization (NGO); Dr. Leonard Levin, from the Electric Power Research Institute (EPRI), a non-profit utility consortium; Dra. Christina Cortinas de Nava, currently a consultant in Mexico, and previously senior staff member of the Instituto Nacional de Ecología of Mexico, and Michael Bender, executive director of the Mercury Policy project, a U.S. based NGO dedicated to the promotion of policies advocating the reduction or elimination of mercury.

Pollution Probe was created thirty years ago to promote research, education and advocacy of issues related to environmental pollution. Their mercury program was established in the year 1996; its goal is the protection of human and ecosystem health through reduction or elimination of the use and release of mercury from anthropogenic sources to the environment. Among their initial activities was the compilation of the first comprehensive mercury inventory in Canada. Since then they have been part of several actions aimed at reducing mercury pollution in Canada and North America. Bruce Lourie, Pollution Probe's mercury program director, gave an overview of these projects, which is presented below.

- First comprehensive mercury inventory in Canada '96
- First NGO led voluntary Memorandum of Understanding with Hospitals '97
- Member of CEC NARAP-Mercury Task Force '98 – '99
- Electrical Products Research '98 – '00
- Mercury Elimination and Reduction Challenge (MERC) Switch-Out '01
- Publication of the Mercury Primer '02
- Participant in the United States/Canada Binational Toxics Strategy (BTS)
- Involved in the Canada Wide Standards processes

Lourie emphasized that science should function as the catalyst for policy making, laying the foundation for the design of appropriate policy frameworks. He described the development of policy frameworks in response to scientific research results, outlining two major stages in the reaction

of decision makers to scientific developments pertaining to mercury.

In the first stage, the early evidence from acute mercury exposure (Iraq, Minamata, Japan) led to the reduction of gross emitters and established an initial policy framework leading to the creation of specific initiatives and involvement of organizations such as the IJC, CEC, the BTS, United Nation Economic Commission for Europe (UNECE) and the New England Governors and Eastern Canadian Premiers (NEG/ECP). Inventories of mercury releases were developed and target sources and source sectors were identified. Most, if not all, of the immediately apparent opportunities for emission reduction were realized at the federal, provincial/state and local levels (i.e. phase out of mercury in paints and batteries, development of primary stage control and containment technologies, deployment of alternate technologies).

Following this initial response, typically new inventories are compiled, highlighting the remaining significant sources and uses for consideration of further control. However, at this stage the cost of reduction appears higher to policy makers and, before initializing reduction strategies, a re-evaluation of science frequently occurs. In this second phase, questions are raised about the relative contributions from different sources and regions, speciation of mercury emissions, the impact of natural sources, and possible ecosystem response to additional reduction.

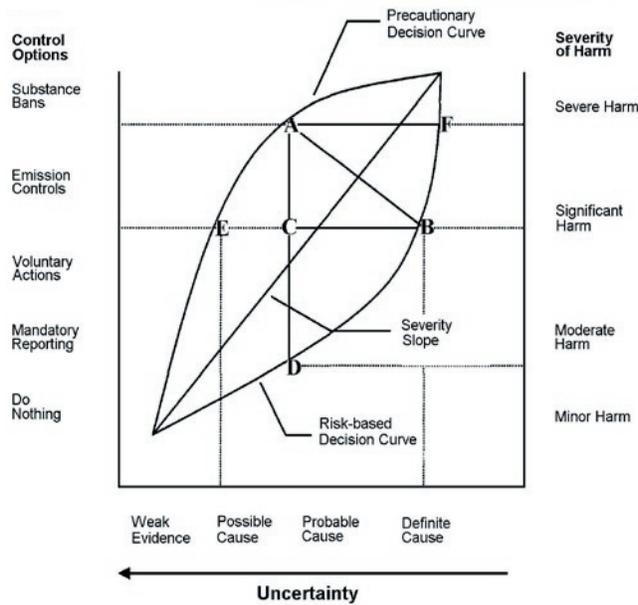
This second phase is viewed as critical by Pollution Probe and other NGOs, particularly the retention of the precautionary principle while science is re-evaluated. At the 1992 Rio de Janeiro Environmental Summit participating countries adopted Principle 15 of the Rio Declaration, which defines the precautionary approach:

“In order to protect the environment, the precautionary approach should be widely applied by states according to their capabilities. When there are threats of serious or irreversible damage, *lack of full scientific certainty shall not be used for postponing cost-effective measures to prevent environmental degradation?* (emphasis added)

Lourie pointed out that actions toward mercury emission reduction should not be delayed in the face of uncertainty. Decision-making pertaining to emission reduction should be rationalized: risk-based models for policy makers should be developed and a decision framework implemented. Policy makers evaluate risk and the possible severity of a situation if actions are not implemented, as they explore alternatives. The design of models is necessary to assess when actions may be invoked and what response might follow. Lourie presented a possible model, shown in **Figure 43**.

After presenting the genesis of policy frameworks, Lourie identified three situations where policy makers responded

FIGURE 43. Decision Framework



to the issues raised by science: (1) policy response to human health risk related to mercury exposure and the advancement of science in this area, (2) incorporation of science into the policy decision making process, and (3) advice from policy makers to the scientific community.

- (1) Lourie stressed that human health risks related to mercury remain the most significant stimuli toward the further engagement of policy makers. In recent years several reports and researchers have captured the attention of the governments. The Minamata and Iraq incidents were the major developments leading to earlier emission reduction policies. In the year 2000 a report by the U.S. National Academy of Science (NAS) stated that *“Chronic, low-dose prenatal methylmercury exposure from maternal consumption of fish has been associated with poor performance on neurobehavioral tests, particularly attention, fine-motor function, language, visual-spatial abilities, and verbal memory.”* (NAS 2000). This raised the issue of mercury to prominence again, obliging policy makers to articulate and respond to the rising concerns surrounding mercury uses, emissions, and deposition.

The source-receptor studies, speciation research, and inventories of global and North American source contributions highlight the issue of local versus global deposition. This development in mercury science has significant implications in decision making. Policy makers are now questioning the necessity or adequacy of action at the domestic level, given the global aspects of mercury pollution. The possible synergistic effects of O₃, SO₂ and greenhouse gases on mercury methylation might also influence the policies pertaining to these

pollutants, requiring a comprehensive approach to air pollution from policy makers.

- (2) Lourie pointed out that policy makers are eager to apply science within the policy frameworks and priorities that exist, but too often these priorities are not well synchronized with the research agendas. Policy is always made within a context of uncertainty but science should consistently support the narrowing of this uncertainty. Scientists should continually consider that policy makers must apply science in recognition of the evolving and sometimes contradictory findings, operating on a weight-of-evidence approach. Their goal is to serve the best interests of the public through the application of science that best shapes the desired policy outcomes.

As examples of how science is being incorporated in the design of policies, as part of the debate on the timing of the government response to the issue of mercury, questions are being raised about how much knowledge of the nature and extent of natural versus anthropogenic contributions is needed prior to setting standards for reductions. Here science could play a critical role in determining the response of the governments.

With regard to the relationship between domestic efforts and global emissions estimates, policy makers may ask if a national strategy is cost effective. Here science could assist in the determination of what course of action should be taken.

- (3) Policy makers can provide advice to the scientific community so that scientific knowledge is fully reflected in the policy making process. Scientists should be realistic regarding their expectations of policy makers' responses. Science programs should be designed with careful consideration of the needs and capabilities of policy makers. The scientific community should also attempt further integration of complex and inter-related ecosystemic interactions to give policy makers a holistic approach to the issue at hand. Finally, events like this workshop, where scientists and policy makers can come together and exchange ideas, are commendable initiatives.

Leonard Levin, manager of the research program on air toxics and mercury at the Electric Power Research Institute (EPRI) also presented some issues related to research and its implications for policy. First, he outlined the complexities of mercury cycling through the environment and the difficulty in tracing this pollutant through its different pathways. He stated that there is still uncertainty about the precise relationship between atmospheric deposition and mercury concentration in fish. Levin pointed out that, at present, models are the best available method to assess the fate of mercury from source emissions, through its atmospheric transport and deposition, and its ultimate arrival into fish.

EPRI, in collaboration with the Mercury Deposition Network (MDN), is undertaking studies to attempt to trace the sources of atmospheric mercury. A series of aircraft measurements have demonstrated that forestfires may be a significant pathway by which mercury contained in vegetation and surface deposits enters the atmosphere. EPRI is also supporting joint studies in northwestern Ontario with Canadian and U.S. agencies who are depositing mercury tracers onto the surface of a lake basin and attempting to follow them through a lake watershed to determine the timing and extent of arrival to fish. This METAALICUS (Mercury Experiment to Assess Atmospheric Loading in Canada and the United States) project should allow an estimation of the time required for mercury reductions to be reflected in fish body burdens.

Another area of uncertainty is the issue of “legacy” emissions. Legacy emissions refer to mercury issuing from soil due to past human activities (tailings at old mine sites etc.) and from geological formations. Mr. Levin stressed that the uncertainty surrounding these emissions needed to be addressed before control strategies are designed and adapted. The influence of non-U.S. sources and background sources on the national source inventory should also be taken into account. Mr. Levin argued that if Asian emissions were reduced by a factor of two, it would have a significant effect on mercury deposition to the United States west coast. Indeed, recent indications are that up to 25 percent of the global total of mercury emissions may emanate from mainland Asia and these may play a significant role in U.S. mercury deposition. However, estimates for Asian mercury emissions are poor and Levin stressed that further research in this area was needed.

Levin also noted that information about mercury transformation in emission plumes is not well understood, impeding a clear understanding of the linkage between the sources of mercury species and their subsequent deposition. More research is needed on plume speciation from a variety of sources; many studies have been done on coal-fired utilities but other sources such as chlor-alkali plants or municipal waste incinerators should be considered. He also pointed out that further research was needed to evaluate the links between specific source types and fish consumption advisories. Without targeting the appropriate sources, any potential change in U.S. industrial emissions may leave the overall goal largely unachieved.

Michael Bender, executive director of the Mercury Policy Project, addressed issues similar to those of previous speakers, but presented them from a more global perspective. The Mercury Policy Project is affiliated with the Tides Center and was formed in the year 1998 to raise awareness about the threat of mercury contamination. It has recently assisted in the formation of the international Ban Mercury Working Group (or “Ban Hg-Wg”), a global network of public interest non-profit organizations working to phase out mercury, ban

international commerce of the metal, minimize exposure and permanently “lock up” surplus mercury. Its primary objective at this time is to influence the United Nations Environment Program’s Global Mercury Assessment to achieve the rapid elimination of mercury uses and releases globally.

As part of this initiative the Mercury Policy Project has advocated formation of a committee of various stakeholders to assess research needs, study mercury emission and exposure comprehensively in order to find a global solution, and prioritize information and data gaps before global strategies and international treaties are drafted. Bender pointed out that current priorities should also be further evaluated by soliciting input from concerned local communities. Issues pertaining to mercury should be put in the context of the reduction of impacts on wildlife and humans.

Mr. Bender was also concerned with the lack of data on the mercury content of marine fish. While freshwater fish are widely tested for mercury in the United States and Canada, the same testing effort is not apparent for marine fish, despite the fact that approximately three quarters of the fish consumed in the United States is marine. Widening current testing to other forms of seafood is vital because fish is just one form of this significant source of protein in the human diet. According to Mr. Bender, the mercury problem cannot be adequately addressed or projections of future trends made if data of this nature are lacking.

Dra. Christina Cortinas de Nava, currently a consultant in Mexico and previously senior staff member of the Instituto Nacional de Ecología of Mexico, presented the policy challenges faced by Mexico. She first stressed that Mexico should be included in any efforts made in the United States and Canada regarding mercury research. The CEC efforts on this contaminant were praised, but more was felt necessary.

Dra. Cortinas de Nava then pointed out that, in Mexico, public awareness of the impact of mercury on ecosystems and human health was very limited and consequently pressures on the government for remedial action were low, resulting in lack of a regulatory framework regarding mercury emissions.

In the Zacatecas region, where mine tailings are ubiquitous, the population was aware of the possible impacts of mercury on human health and pressured the government to act. This social concern created some political will to deal with the issue, but a clear relationship between health effects and mercury pollution was not established, resulting in a lack of follow through. Indeed, in the absence of a quantified human health risk, public pressure alone does not appear to be a sufficient motivator for political action.

This experience convinced Dra. Cortinas de Nava that mercury fish contamination was the best enticement for the

government to take action. The region of Veracruz, locale of the largest oil refineries in Mexico, has a population largely reliant on fish in their diet. Because of pollution, not only by mercury but also by lead and cadmium, this region might be one where awareness in the Mexican government might be raised and have a subsequent impact on policy makers.

Dra. Cortinas de Nava pointed out that, while some monitoring activity is present in Mexico, more specifically for water quality, there is no systematic multi-media monitoring and no data on the exposure effects for the population. The lack of capacities and infrastructures and the absence of a good source inventory impedes detailed consideration of policies. Stressing the possible health risks for the Mexican population can motivate capacity building and monitoring, but political will is not the only obstacle in Mexico; lack of funds is also an issue. Increased cooperation with the United States and Canada in the development of joint projects and information sharing between the countries would assist greatly in overcoming such obstacles.

Dra. Cortinas de Nava concluded that the scientific community must present projects to policy makers in a practical, innovative manner. The case of Mexico highlights the fact that mercury policies and strategies can be implemented only if the human health issue is put forward strongly both to the decision makers and also to the concerned population. She also challenged the modeling community by proposing modeling exercises using Mexico as a domain.

To conclude the session, Luke Trip outlined the main issues raised in the different presentations:

- Human health aspects and the reaction of a concerned population are major catalysts for policy makers to take action.
- Pollution prevention and the precautionary principle approach are important touchstones in the development of mercury policy.
- The lack of data regarding background sources should be remedied; in the meantime it should be taken into account when policies are designed.
- Global vs domestic contributions should be weighted when strategies for emission reduction are put forward.
- An ecosystem approach should be used when findings are presented to policy makers.
- Contamination of marine fish is an issue lacking analysis and understanding—policy makers and the scientific community may need to assess this pathway to humans more carefully.
- Mexican capacity for monitoring must be increased through cooperation; Mexican data must be improved and included in modeling exercises.

In the question and answer period that followed, several issues were raised. The absence of any discussion on the impact on wildlife was noted; it was pointed out that the damage done by DDT to the bird population and other wildlife played a major role in the responses of various governments. With mercury, there has been little focus on the wildlife impact or how this issue might generate interest in the public and political spheres (one exception is the media attention on the effects of mercury on loons in the Maritimes). A participant responded that a shift in the political mind set occurred recently, with the political and public realms more focused on human health issues, particularly in Canada. Health issues are now central to political and public debates and policy makers might be more responsive to data and findings that can be related in some way to human health.

5.2 Policy Discussion

This segment of the workshop began with a presentation by representatives of the three countries (Mexico, Canada, and the United States) on the current and future situation with regard to the evolution of mercury capacity and policy.

5.2.1 Current Status

5.2.1.1 Mexico

Rocio Alatorre Eden Winter of the Instituto Nacional de Ecología (INE) outlined the main goal of the INE to provide information to the decision makers. Presently in Mexico a rudimentary emissions inventory for mercury is being assembled and private laboratories are developing the capacity to analyze for this contaminant. In the near future, Mexico should have two wet deposition monitors, as well as a bank of human mercury samples and some quality assurance and quality control programs. As well, Mexico would continue to replace mercury thermometers, develop risk communication programs, begin the recycling of mercury with dentists and develop new regulations for nonessential uses of the metal.

5.2.1.2 Canada

Luke Trip of the Environmental Protection Service, Environment Canada, described the current involvement of his department in many initiatives and programs to better understand and control mercury. The policy direction in the Environmental Protection Act is initially a precautionary approach, leading to the virtual elimination of sources of anthropogenic mercury. The program plan for the years 2001/2002 includes:

- a socioeconomic study on mercury retirement in relation to market forces to determine if mercury remains a viable commodity

- international initiatives with United Nations Environment Programme (UNEP) and UNECE focused on best available prevention techniques.

Environment Canada and Health Canada are cooperating on the following joint initiatives:

- Dental Amalgam Waste Memorandum of Understanding (MOU)
- other devices under the Medical Devices Act and the Hazardous Products Act

Activity continues on the further control of mercury emissions from metallurgical processing sources and the development of a multi-pollutant (including mercury) strategy for the electric utility sector.

Beyond the year 2002, activities include:

- Continued implementation of control strategies;
- Long-term storage and management of any mercury exceeding Canada's needs;
- A communications strategy involving a mercury knowledge base on the Internet (joint project of the USEPA and Environment Canada);
- Research partnership with other federal departments and the coal-fired utilities sector; and
- Continued support for the UNEP Mercury Initiative

5.2.1.3 United States

Sally Shaver, Director, Emissions Standards Division of USEPA presented a series of slides. The first dealt with needs which included:

- improved emissions data
- better information on;
 - new sources of data
 - transport and fate of mercury
 - measurement capability
 - human health exposures
 - health effects
 - control methods and their economic feasibility

Application of science in the following fields:

- development of a mercury action plan
- continuation of reductions in emissions from MWI (Municipal Waste Incinerators) and medical waste incinerators and introduction of programs to lower emissions from the electric utility sector
- pursuit of pollution prevention in preference to regulation
- increased emphasis on global studies and international strategies
- increased communications and outreach

Advice from policy makers to scientists:

- focus on priorities - gaps, long-term and short-term solutions
- look to specific areas or sectors where emission reductions could be best achieved
- increase ability to measure mercury species at source and in the environment
- evaluate exposure of high risk populations in a realistic manner
- continue evaluation of global mercury issues

The United States has reaffirmed its commitment to reduce mercury emissions. Currently it is developing regulations for the utilities sector with the following timeline:

- a proposal by December 15, 2003
- promulgation by December 15, 2004
- compliance by December 15, 2007

5.2.2 Discussion

The subsequent discussion centered on the three workshop questions:

- What implications are emerging from the science that could affect policy?
- How do policy makers best apply the results or tools available from the science?
- What advice can policy makers provide to better focus future scientific work?

Before the questions could be considered, several broad issues were discussed. As observed in other workshops where policy and scientific interests have interacted, a communication gap was quite apparent between the two interests. This gap could impede full appreciation of the deleterious impact of mercury deposition and subsequent development of an appropriate control policy. The need for bridging mechanisms was apparent. Workshops were considered to be one mechanism; others should be sought to provide more opportunity for open discussion between scientists and policy makers in place of frequently formal segregated presentations.

Among the suggestions from the policy community was a commitment by scientists to better understand the milieu of the policy maker. John Arseneau of Environment Canada noted that decision makers have certain characteristics. They are motivated, committed, empowered individuals who are action oriented and frequently called upon to choose the least imperfect option from a selection of not wholly desirable outcomes. Often the challenge is the selection of the least harmful or unbalancing option rather than the ideal or best solution.

Scientists must realize that not only is policy based on fact and knowledge but it is an attempt to balance the interests of many parties, both private and public. Ongoing dialogue and exchange between scientists and policy makers are crucial. What are the policy makers' priorities? They want:

- a common and sound database
- researchers to address policy makers' questions, including:
 1. what is the problem?
 2. where is the best point of influence for the problem?
 3. what instruments can be developed to affect the influence point for the problem?

Sally Shaver (USEPA) and John Arseneau agreed that, in terms of developing policy, it is more important that scientists, in responding to queries from policy makers, bring forward, to the extent possible a consensus as their response rather than attempting to perfect an exact and precise answer. The uncertainties behind any consensus should also be made clear. Scientists must communicate a clear and relatively broad understanding of the science at any given point in time to allow policy makers to respond to the "So what?" question.

Links between and among research groups are often not very strong, and should be improved to allow more supportive research. To the extent possible, the scientific community needs to organize in a manner that leads to the development of these consensus statements.

In response, participating scientists explained their difficulty in answering policy questions. Frequently, they perceive such questions as vague and fuzzy, and resistant to a scientific response. While the processing of these questions may appear to be simple to the policy makers posing them, they often require consideration of complex information. Frequently, answers are attempted through the use of models; however, any model is only as good as the data and information it is based on.

In the case of mercury, its behavior in the environment is very complex and hence difficult to model. A high quality speciated emissions inventory, an understanding of the physical and chemical processes, and accurate ambient measurements are all required if model output is to have an established value. To the extent possible, simplistic responses must be avoided. It is important that policy makers allow scientists to facilitate and guide scientific work.

The scientific community was also concerned that science that reflects directly on policy often does not appear in the peer-reviewed literature. As publication is a crucial means of communication and recognition in the scientific field, it was suggested that an incentive be put

in place within agencies rewarding this type of work and supporting such publications.

The science used to develop policy plays a crucial role in the process. John Arseneau indicated that science informs policy makers on the nature of the problem through identification and characterization. Scientists should attempt to indicate clearly what is currently known about the problem and identify where or by what means the problem can best be influenced. Policy makers depend on scientists to provide a larger or broader view, as they are perceived as objective and independent and not as driven by immediate issues.

As well, scientists must generate realistic expectations for the policy makers regarding timelines and impacts. Short-term tangible results are often necessary to support implementation of any long-term solution. These may take the form of simple answers frequently most desired by policy makers, but they must be based on reasonable science and explicit consideration of their limitations.

Assessment of progress is also critical, while recognizing that such work often is seen as unattractive in the scientific community. For example, although development, and particularly, operation of ambient air quality networks may not be viewed as desirable work by scientists, the role of such networks in the evaluation of the effectiveness of any applied policy and subsequent allocation of resources remains crucial.

The contribution of science to policy making can be seen through the recent influence it has had on policy in the United States. The recent improved emissions data developed by the United States in response to Congress's amendment of the Clean Air Act in the year 1990, where 188 hazardous air pollutants, including mercury, were codified, reinforced the need for more information on the coal-fired utilities as a mercury source. The Toxic Release Inventory revealed additional sources that may require regulation, including coal mines.

To develop new regulations, more information is needed from science regarding the transport and fate of mercury. The continuing determination of source-receptor relationships should make a significant contribution to the development of an effective control strategy.

At this workshop, policy makers also gave advice on how to improve the focus of scientific research. Mercury topics suggested for future examination included the further development of source control methods and technologies, and the evaluation of exposure in terms of realistic consequences or outcomes, with a focus on levels and impacts among high risk populations. While agreeing with scientists that better modeling would require improved data and databases, it was also noted that scientists need to consider the questions

confronting policy makers and explore the problem more holistically, perhaps beyond the confines of their individual research, to “connect a few more dots.”

Finally, it was suggested that scientists need to broaden their horizons and regard mercury as a continental issue affecting all of North America and, ultimately, a global issue. It was suggested that a committee of interested stakeholders at the global level be convened to identify research needs and assess possible initiatives to ensure that funds are allocated to areas where a good return for the investment would be anticipated. The committee should solicit input from stakeholders and in particular, those at highest risk.

The communication gap exists not only between the policy makers and scientists but also with the general public. It is important that policy makers and scientists participate in a coherent way in making the public aware of the issues and educating them regarding the sources and presence of mercury and the realistic dangers associated with it.

In conclusion, Luke Trip offered some summary comments on the three questions raised at the workshop.

1. What implications are emerging from the science that could affect policy?

Observations:

- There are too many unknowns in the dry deposition data.

- Human health impacts need better delineation
- Mercury emission inventories (anthropogenic and natural) and global contributions all need better determination and characterization (speciation).
- Some level of established certainty will be necessary to drive policy.

2. How do policy makers best apply the results or tools available from the science?

The results or tools could be useful for:

- communication in the public realm;
- communication in their own organizations, and the bridging between policy and science;
- global studies to see what influences us.

3. What advice can policy makers provide to better focus future scientific work?

- A better consensus among scientists is needed.
- All of North America, and indeed the global influence, need consideration.
- To the extent that the next important issue (the smoking gun) can be anticipated, it is likely to be in the human health area.

6.0 FINDINGS AND RECOMMENDATIONS

6.1 Findings

In addition to the positions put forward by individual presenters at the workshop and those arising from the panel discussions, the International Air Quality Advisory Board of the IJC and the Mercury Task Force, Sound Management of Chemicals of the CEC have developed the following findings.

6.1.1 Sources

- **Given the ability of its elemental form to remain suspended in the atmosphere for over a year, with subsequent transport over long distances, mercury is a global pollutant. While significant fluctuations in the global emissions inventories for mercury were not apparent in recent years, changes were observed in their dominant sources and geographic locations.**

Although the global inventories developed by Pacyna & Pacyna suggest no significant change in total mercury released annually from anthropogenic sources to the atmosphere between the years 1990 and 1995, estimated to be in the vicinity of 2000 tonnes (~ 2200 tons) per year, alterations in the nature of dominant sources and their geographic locations were apparent. In the more recent inventory (the year 1995), North American and European sources showed decreasing emissions. However, estimates of the Asian contribution to total emissions have increased by over 25 percent; the majority of emission are largely the result of increased coal combustion in China.

- **While recognizing that uncertainty in the definition and estimation of ‘natural’ emissions remains to be addressed, this should not be a rationale for inaction toward further reductions of mercury emissions from anthropogenic sources. Anthropogenic sources will continue to account for a most substantial portion of total emissions, even given a generous and broad definition of natural sources, and reduction of mercury emissions from anthropogenic sources should continue to be aggressively pursued.**

A number of presenters observed that the contribution of background or ‘natural’ sources to the global mercury pool might represent as much as 50 percent of total emissions, and advocated a better accounting of these sources, both in terms of quantity and species. However, the initial need was determined to be an elegant definition and differentiation of these emissions, as what currently might be characterized as ‘natural’ reflects, in some cases, an original anthropogenic

release from several decades (or centuries) ago. Action toward further reductions in anthropogenic mercury emissions, through both voluntary co-operative activities and effective regulation, should be accelerated while such issues are addressed.

- **The methodology used to develop source inventories of emissions, particularly in addressing the comprehensive collection of all necessary parameters and quality assurance of collected data, appears to lack scope, rigour and transparency.**

The need to improve and enhance current mercury emission inventories was emphasized by several presenters. Modelers were particularly adamant on this issue as the accuracy of inventory data bears directly on the accuracy of their modeled deposition estimates. While sampling protocols and quality assurance and quality control (QA/QC) procedures are routinely documented and available for ambient measurements, such information is frequently not provided for emission estimates. Validated modeling is dependent upon improvements to current coordination mechanisms to further require nations, states and provinces (frequently the primary sources of such data) to provide all necessary information required in a comprehensive and comparable form as input for models. Also, the extent to which mercury is volatilized from waste disposal sites and other area sources to form a part of the urban plume, particularly in warmer weather, should be investigated further.

- **Further progress under the Mexican regulatory process should provide additional information on significant sources of mercury in the near future.**

In Mexico, limited official information is currently available on mercury emissions and mercury content in feedstock or waste streams, due in part to the embryonic nature of pollutant release and transfer (PRTR) reporting regulations there. At the time of the workshop two source categories, cement production and burning of waste fuels, were regulated; standards for incineration, hazardous waste, and pharmaceuticals sectors were proposed but were not yet promulgated. Adequate continental modeling scenarios involving Mexico cannot be improved until further reporting requirements are in place.

6.1.2 Transport and Deposition

- **Mercury atmospheric and deposition processes are very complex; further research is needed to better understand mercury kinetics and chemistry, the interaction with other pollutants and species, and the subsequent impact on transport and deposition processes. For example, while data showing rapid cycling of mercury between the surface and the**

atmosphere during polar sunrise undergo further scrutiny, the precise nature of mercury cycling in the Arctic region has yet to be determined. Until issues regarding kinetics and chemistry are better resolved in this locale and elsewhere, the output of mercury deposition models will continue to be marked by significant uncertainties.

Mercury chemistry is complex, with many drivers and possible factors influencing behaviour during emission, transport, and wet and dry deposition. Several presentations emphasized the need for a more sophisticated knowledge of the chemistry of mercury species in various media if the behaviour of mercury in those media, including biota, is to be properly understood.

- **The lack of knowledge regarding mercury deposition pathways and fluxes in water bodies must be addressed as part of the development and implementation of appropriate abatement strategies. A mass balance/whole-ecosystem approach is necessary to better understand the sources and movement of mercury throughout the ecosystem, including its entry into and further concentration within the food chain.**

Some participants argued that interactions between mercury currently resident in soils and ongoing mercury deposition are not well characterized, inhibiting estimation of the speed and extent of the response of mercury content in fish to emission reductions. Large pools of mercury are present in watersheds, and they may modulate the response to any changes in deposition. The existing pools may also be susceptible to mobilization/re-volatilization and ultimately further contamination of fish. The METAALICUS study in the Experimental Lakes Area in northwestern Ontario, where the relationship between the atmospheric loading of mercury to watersheds and the mercury concentration in the fish population is being examined, should provide valuable guidance on these issues and should be tracked closely.

6.1.3 Monitoring

- **Ambient air mercury data of sufficiently high quality are essential to the determination of the burden on the ecosystem. Establishment of core monitoring sites, with measurement of all relevant chemical species in all relevant phases, thorough spatial coverage, further standardization in sampling and analytical protocols and in the selection and application of meteorological data, and an enhancement in overall quality assurance and control are all necessary activities. These activities are crucial to the accurate determination of backgrounds, loadings, and significant source regions on a continental and global basis, as well as**

the verification of model outputs and formulation of effective enhanced controls.

Workshop participants concurred with the importance of additional investments in high quality monitoring programs to better determine loadings of mercury to the environment while increasing confidence in the delineation of source-receptor relationships and the verification of deposition models.

- **Enhanced and sustained monitoring of the parameters necessary to estimate dry deposition is essential for a complete understanding of mercury loading in the environment.**

Dry deposition should not be underestimated when reflecting on the mercury cycle in the environment. The relative lack of measurement of the parameters necessary to determine long-term dry deposition inhibits an improved estimation of mercury loading and verification of mercury deposition models.

- **Monitoring activities, including aircraft based sampling programs, designed to determine the atmospheric concentrations of various species of mercury and their interaction, are required to better comprehend deposition patterns, residence time, and atmospheric chemistry associated with the various species present.**

Participants agreed that speciated monitoring from various platforms, including aircraft, was imperative if a full understanding of the behaviour of mercury in the atmosphere and the larger environment was to be achieved. While a mercury wet deposition network is in place and speciated measurements are made at CAMNet stations and other discrete locations, there was no systematic comprehensive network of speciated monitoring sites, nor a program to ensure comprehensive measurement via aircraft of airborne species over relevant time scales.

- **Near-source monitoring is necessary to achieve a better understanding of emitted plume chemistry.**

A lack of adequate monitoring near anthropogenic sources was noted as an impediment to an enhanced understanding of plume chemistry and further improvements to modeling and source/receptor determinations. Future studies of mercury wet and dry deposition with high resolution sampling and meteorological support data should be undertaken adjacent to major mercury sources.

- **There is a need for increased spatial and temporal coverage in monitoring activities, especially in and over water and downwind of urban areas.**

Spatial coverage of current monitoring programs was not considered adequate to allow a confident determination of representative loading estimates, particularly in areas with a higher concentration of emission sources. Temporal coverage was also an issue; increased temporal monitoring would further the understanding of factors such as mercury methylation rates and the subsequent presence of mercury in fish.

- **Current programs in Canada and the United States to determine the mercury level in freshwater fish should be sustained and consideration should be given to more extensive monitoring of marine fish and other seafood. Mexico should initiate a national program to measure the mercury content of freshwater and marine fish and seafood species in locales where these are a significant food source to humans.**

Consumption of freshwater and marine fish and other seafood accounts for much of the elevated mercury concentrations observed in humans, a fact reflected in the large number of Canadian and American freshwater lakes under fish consumption advisories due to the mercury content of certain species of fish. As modeling, monitoring, and assessment activities continue and further reductions in anthropogenic releases are put in place, initiating, sustaining and enhancing the determination of the mercury content in freshwater fish will remain a core activity. Workshop participants also noted the need for initial and enhanced monitoring of marine fish and other seafood throughout North America to allow estimation of the possible daily intake of mercury by humans from both freshwater and marine foods.

- **Mercury monitoring and testing are still in their developmental stage in Mexico and great opportunities for improvement exist through supportive technological transfer.**

In Mexico, neither routine atmospheric deposition nor well established ambient atmospheric concentration monitoring networks exist. Continual baseline monitoring of the mercury content of freshwater and marine fish species and other seafood is also lacking. A strengthening of basic source and ambient test methods, including QA/QC procedures, is also needed. Technological transfer among the United States, Canada and Mexico offers a rapid and effective means to improve on these circumstances.

6.1.4 Modeling

- **The North American continent appears to be significantly impacted by global mercury emissions,**

particularly those arriving via trans-Pacific and trans-Arctic pathways. Work to date suggests that the global contribution to domestic mercury pools must be further considered in the development of continental and large regional scale models.

Global models of atmospheric mercury transport indicate that the contribution of external sources to mercury loading within the North American continent could be significant. While substantial levels of uncertainty in model prediction preclude quantitative statements, the contributions of Asian emissions to the North American budget are a factor to be considered by modelers.

- **Comparison of models, with a better understanding of the effects of methods used to determine various parameters on calculations, is needed.**

Some modelers emphasized the importance of model inter-comparison exercises for calibration of models. Even if such comparisons show appreciable discrepancies, a better understanding of the effect on model precision and accuracy caused by the variation in input parameters used to represent physical and chemical mechanisms, and the differences in other entry data sets, would result.

6.1.5 Policy

- **As mercury exhibits adverse health effects on humans even at very low levels, the examination of human health risks associated with mercury will continue to be the most effective stimulus for appropriate control initiatives for mercury emissions locally, regionally and globally.**

The impact of relatively low levels of mercury on cardiovascular, immunological, and neurological well-being in humans and other such effects is now central to political and public debates regarding mercury emission control programs. Continued focus on and examination of these issues are crucial to determining appropriate further advances in voluntary and regulatory control programs.

- **Policy makers must function in a context of uncertainty and scientists should consistently attempt to narrow this uncertainty and, to the extent possible, reach some “precautionary approach” consensus prior to discussions with their policy counterparts.**

Mercury emission inventories (anthropogenic and natural) and global contributions, wet and dry deposition mechanisms, are among the many issues that need better determination and characterization. The unknowns surrounding these issues could hinder further policy efforts; some level

of consensus on these and other issues among the scientific community would be most helpful to policy development.

- **Further focused interaction between the scientific community, especially modelers, and policy makers is necessary to effectively address the mercury issue. Regular dialogue between the scientific and policy elements within environmental agencies is crucial to scientific work and the design of policy that is relevant and responsive.**
- **Global emissions require further consideration in the evolution of strategies for emission reduction.**

Evidence presented during this workshop showed that the global contribution to the North American mercury pool is significant and needs to be better quantified.

6.2 Recommendations

The International Air Quality Advisory Board and the Mercury Task Force of the Commission for Environmental Cooperation recommend the following:

1. **A continued focus in the three countries on further reductions in emissions of mercury from anthropogenic sources through an effective combination of voluntary and regulatory programs.**
2. To reduce the uncertainty associated with anthropogenic emission and ambient concentration and loading estimates, **development of a long-term commitment of enhanced and stabilized funding to improve the quality, comparability and scope of mercury source and ambient measurements, including levels in selected biota, the availability of appropriate meteorological data, and to support associated modeling efforts.**
3. Comprehensive programs to link atmospheric deposition and other mercury pathways with bioaccumulation in fish be enhanced. **Canadian and American programs to measure the mercury content in freshwater fish consumed by humans should be continued and current measurements of marine food species should be enhanced. Mexico should be supported in the initiation and maintenance of such programs.**
4. **The modeling community** is encouraged to develop a comprehensive description of mercury measurement needs central to the evaluation and further improvement of models, **while moving to account for global loading in their estimates as appropriate**, especially the contribution via trans-Pacific and trans-Arctic pathways to the North American mercury pool.
5. With the current movement toward additional voluntary usage reduction programs and consideration of further controls on anthropogenic sources, **available resources should be augmented in the areas outlined above so the outcome of these control measures can be adequately predicted and subsequently determined.**
6. **Appropriate government agencies should be encouraged to increase dialogue between the policy and scientific arms of their organization** to ensure that policy evolves from the most current and robust science.
7. **Investigation of further possible effects of mercury on human health must be sustained**, along with interactions among health experts and the monitoring, modeling and policy community to ensure that the most current and relevant information on human health effects is available while considering further reductions in anthropogenic mercury releases.
8. **Canada, the United States and Mexico should continue and enhance their co-ordinated approach, with joint technical programs where possible, in all aspects of mercury research and policy development.**
9. Recognizing mercury as a global pollutant which must ultimately be addressed at a level beyond the continental, **the dialogue between the two Commissions (the International Joint Commission and the Commission for Environmental Cooperation) on the mercury issue be maintained and opportunities for interaction with other International and Intergovernmental organizations be acted upon.** As an example, North America should participate fully in the UNEP global assessment of mercury currently underway.

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ACRONYMS

ADOM	Acid Deposition Oxidation Model
AER	Atmospheric and Environmental Research Inc.
AQM(s)	Air Quality Model(s)
ARET	Accelerated Reduction/Elimination of Toxics
ARQP	Air Quality Processes Research Division
BTS	Binational Toxics Strategy
CAC	Criteria Air Contaminants
CAM	Chemistry of Atmospheric Mercury
CAMNet	Canadian Atmospheric Mercury Network
CEC	Commission for Environmental Cooperation
CEPA	Canadian Environmental Protection Act
CMAQ	Community Multi-scale Air Quality Model
CNA	Comisión Nacional del Agua
CTM	Chemical Transport Model
CWS	Canada Wide Standard
DDT	Dichloro-Diphenyl-Trichloroethane
EMEP	European Monitoring and Evaluation Program
EPA	Environmental Protection Agency
EPRI	Electric Power Research Institute
FEDDS	Florida Everglades Dry Deposition Study
FIPS	Federal Information Processing Standards
GEIA	Global Emission Inventory Activity
GLWQA	Great Lakes Water Quality Agreement
GOME	Global Ozone Monitoring Experiment
GRAHM	Global/Regional Atmospheric Heavy Metal
HAL	Hg Analytical Laboratory
HAP(s)	Hazardous Air Pollutant(s)
HYSPLIT	Hybrid Single Particle Lagrangian Integrated Trajectory
IAQAB	International Air Quality Advisory Board
ICR	Information Collection Request
IJC	International Joint Commission
INE	Instituto Nacional de Ecología
MDE(s)	Mercury Depletion Event(s)
MDN	Mercury Deposition Network
MeHg	Methylmercury
MERC	Mercury Reduction & Elimination Challenge
MERS	Multi-Pollutant Emission Reduction Strategies
METAALICUS	Mercury Experiment to Assess Atmospheric Loading in Canada and the United States
MOU	Memorandum of Understanding
MSC	Meteorological Service of Canada
MSC-E	Meteorological Synthesizing Center - East
MWI	Municipal Waste Incinerators
NADP	National Atmospheric Deposition Program
NARAP(s)	North American Regional Action Plan(s)

NAS	National Academy of Science
NASA GISS	National Aeronautics & Space Administration Goddard Institute for Space Studies
NCAR	National Center for Atmospheric Research
NCEP	National Centers for Environmental Prediction
NCP	Northern Contaminants Program
NEG/ECF	New England Governors and Eastern Canadian Premiers
NEI	National Emission Inventory
NERL	National Exposure Research Laboratory
NGM	Nested Grid Model
NGO	Non-Governmental Organization
NILU	Norsk Institutt for Luftforskning (Norwegian Institute for Air Research)
NOAA	National Oceanic and Atmospheric Administration
NPRI	National Pollutant Release Inventory
NTI	National Toxic Inventory
NYSDERDA	New York State Energy and Research Development Authority
OMOE	Ontario Ministry of the Environment
QA/QC	Quality Assurance/Quality Control
RAMS	Regional Atmospheric Modeling System
RELMAP	Regional Lagrangian Model of Air Pollution
RGM	Reactive Gaseous Mercury
SEARCH	SouthEastern Aerosol Research and Characterization
SMOC	Sound Management of Chemicals
SPDC	Static plume dilution chamber
SRW	Simulated rainwater
SUNY	State University of New York
TGM	Total gaseous mercury
TRI	Toxics Release Inventory
U.S. EPA or USEPA	United States Environmental Protection Agency
UNECE	United Nations Economic Commission for Europe
UNEP	United Nations Environment Programme

SYNOPSIS OF PRESENTATIONS

A brief overview of the content of the presentations at the workshop and contact information for presenters is provided below.

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Decision makers exhibit characteristics that scientists ought to be aware of as an initial effort to better understand the policymaker. Scientists need to understand that policy is generally based upon the notion of balancing interests of many parties. Ongoing dialogue and exchange between scientists and policymakers are crucial in future appropriate policy advancements.

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With the assistance of the Mercury Policy Project, a multinational perspective developed in response to mercury contamination, the International Ban Mercury Working Group has been formed. Further, the Mercury Policy Project advocates the formation of a committee of distinct stakeholders to assess issues such as research needs, emissions inventories and human exposures in order to develop a global strategy and prioritize information and data gaps. The lack of data on the mercury content in marine fish is emphasized in the presentation.

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The Canadian Atmospheric Mercury Network (CAMNet)

The Canadian Atmospheric Mercury Network (CAMNet) was created to improve understanding of mercury trends and processes in the environment. Monitoring data are collected by the Air Quality Processes Research Division (ARQP) of Environment Canada. The initial focus of CAMNet was to ensure that measurements of total gaseous mercury were of high quality and comparable across the nation. The mandate has since been expanded to share knowledge and coordinate measurements of mercury in precipitation through MDN involvement. Reactive gaseous mercury has been included in the measurement and future initiatives may also include particulate mercury.

Monitoring of atmospheric mercury in Atlantic Canada has included continuous analysis of total gaseous mercury in Nova Scotia and New Brunswick. Data derived from these areas suggest that, in addition to the links to anthropogenic sources, natural mercury sources also contribute to spatial and temporal variability in total gaseous mercury. Further studies suggest that TGM may be affected by local and long range sources and seasonal patterns, with increased concentrations during the summer months.

Ontario has two gaseous mercury monitors in place to further temporal and spatial analysis at the IADN (Integrated Atmospheric Deposition Network) sites at Point Petre and the satellite site at Egbert. Québec has two monitoring sites that cover the entrance and exit of the main St. Lawrence River wind corridors. Studies at these locations are comparable to those in the Atlantic region, reflecting a seasonal pattern with increased mercury concentrations during the summer.

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U.S.EPA and NOAA Resources Lab

Eulerian-type Atmospheric Mercury Model Development at the U.S. EPA/ORD

The Community Multi-scale Air Quality (CMAQ) model presently in use by the U.S.EPA has been adapted to mercury and its associated interacting physicochemical reactions to integrate major tropospheric air pollutants in a multi-scale "one atmosphere" structure. Several simulated processes are included in the modules: horizontal and vertical advection; mass conservation adjustments for advection processes; horizontal and vertical diffusion; gas-phase chemical reaction solver; aqueous-phase chemical reactions and deposition; aerosol dynamics and size distributions; gas and aerosol dry deposition velocity estimation; and plume chemistry effects. Primary factors such as chemical and physical forms of emission and chemical and physical reactions in clouds affect the transport range of mercury emissions to air.

By comparing cloud chemistry model simulations, it is found that mercury cloud water chemistry is significantly affected by chlorine (Cl_2) and hydroperoxyl radical (HO_2) concentrations in the interstitial air between cloud droplets and thereby exhibits strong day/night oscillations in cloud water mercury concentration. Mercury concentrations already existing in the clouds may also affect the increase, or lack thereof, in concentration should additional mercury be added.

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NOAA Air Resources Laboratory

Modeling Mercury Transport and Deposition to the Great Lakes Basin and New England

The overall goal of the Atmospheric Transport and Deposition of Mercury to the Great Lakes project at the NOAA Air Resources Laboratory is the development of atmospheric mercury source-receptor information for each of the Great Lakes, the Gulf of Maine, and other selected receptors. The relative contributions of *different source regions* and *different source categories* to atmospheric deposition to each receptor are estimated using a specially-configured version of the HYSPLIT_4 model. The model incorporates important fate and transport processes for atmospheric mercury, including dispersion, phase-partitioning, chemical equilibria and reactions, and wet and dry deposition. A binational emissions inventory derived from U.S. EPA and Environment Canada is used as a preliminary step in assessing mercury source-receptor information. The model is being evaluated by comparing its predictions with ambient measurements and with independent estimates of deposition to various receptors (e.g., estimated mercury deposition to Lake Michigan during the Lake Michigan Mass Balance Study). It is hoped that resources will be available in the future to extend the modeling domain and emissions inventory to include Mexico and other parts of the globe.

An initial assessment of the mercury source-receptor relationships for atmospheric deposition to the Great Lakes and the Gulf of Maine is presented. The presentation also addresses some of the uncertainties inherent in the current version of the model.

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Consultant

The Mexican public is largely unaware of the effects of mercury on human and ecosystem health, and thus, government response on this issue is limited. However, in the Zacatecas region, where the public was aware of the possible impacts of mercury, pressure was placed on governments to act. Unfortunately, the lack of a precise acknowledged relationship between mercury and health has resulted in inaction. It is believed that fish contamination is the best trigger for government action. Mexico lacks a systematic fish monitoring program, so further improvements to these measurements as well as enhancement of emissions inventories, and examination of the quality of data so generated are necessary. An investigation into the toxicity levels of mercury in fish in the area of Veracruz, where oil refineries and chloralkali plants are located in close proximity to the river is recommended.

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A Global Model of Mercury Transport

The Global/Regional Atmospheric Heavy Metal model (GRAHM) is an Eulerian Multiscale model used to investigate atmospheric mercury on a global scale. This model can assess relative contributions of mercury from major anthropogenic source regions of the world. The model estimates mercury deposition concentrations on a seasonal cycle, mainly the highs over land during the winter and over water masses in the summer, in addition to seasonal source contributions. Presentations include maps and pie charts on mercury deposition and emission concentrations and sources illustrating contributions from source regions. Limitations to this model are the exclusion of volatilization; data limited to northern sites; lack of accounting for photochemistry; and the current inability to predict concentrations in the Arctic.

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Sources of Mercury in Canada

The Canadian mercury emission inventory is assembled using data from a variety of sources, including the National Pollutant Release Inventory (NPRI), Criteria Air Contaminants emissions inventory (CAC), Accelerated Reduction/Elimination of Toxic Program (ARET), industry supplied estimates and further consultation with the industry. There have been recent changes to the NPRI which allow for better data for specific facilities.

Mercury emissions have been reduced by 90% in Canada since the 1970 estimate. A comparison with the U.S. revealed that the dominant emission reductions were from mining and smelting in Canada, whereas decreases in the U.S. have been largely attributed to enhanced controls of municipal and medical waste incineration. Further reductions of mercury emissions are envisioned through the Canada Wide Standard (CWS) and the Multi-Pollutant Emission Reduction Strategies (MERS) developed by the federal and provincial governments.

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The Florida Everglades Dry Deposition Study (FEDDS) improves the understanding of dry mercury deposition processes by providing information to better represent and parameterize dry deposition in atmospheric models and to provide deposition estimates as validation checks on models application. Although measurements were taken at significantly distant locales from anthropogenic mercury sources, high RGM levels and diurnal cycling behaviour were observed. The disappearance of RGM in dew was also observed. Further RGM analysis was conducted near Ann Arbor, Michigan with similar observations to the FEDDS. However, when compared with data gathered for other locations, it was evident that RGM behaviour varies between Arctic, Coastal and rural areas.

The importance of environmental monitoring and the inadequacy of current national efforts to properly assess the trends in ambient mercury or the dry deposition of mercury were emphasized. In addition, further spatially and temporally resolved data as well as the extension of monitoring to marine environments are needed for calibration of regional and continental models.

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Mercury Speciation Network and Aircraft Measurement Campaign

Transport characteristics of mercury vary substantially according to species. Long range transport is associated with elemental mercury, whereas regional and local transport is correlated to reactive gaseous and particulate mercury. Studies on mercury transport include an aircraft measurement campaign in Coral Springs, Florida and mercury speciation experiments in Point Barrow Alaska.

In Coral Springs, high levels of mercury are found in fish living in the Everglades. The objective of this project is to determine sources of mercury. Through sampling efforts it was concluded that the ocean was not seen to be a source of mercury; however data suggest that long range transport of mercury combined with oxidation mechanisms in the atmosphere contribute to high levels of reactive gaseous mercury in the Everglades.

The Point Barrow Alaska study revealed high concentrations of particulate mercury during the night and high gaseous mercury during the day. In addition, aircraft profiles demonstrate a surface phenomenon whereas, at higher elevations, concentrations of mercury are low or non-existent.

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Measurement of Mercury Emissions from Stationary Sources in Mexico

There is currently no atmospheric deposition or atmospheric concentration monitoring for mercury in Mexico, as all monitoring efforts have been focused on soil and water contamination. Initial mercury tests began in the 1990's for incineration facilities and cement kilns burning hazardous waste. Other sources such as combustion facilities and users of mercury are not regulated and monitored. Regulation of monitored sources is based on permits, which lack well defined reference methods and testing protocols.

New standards for incineration facilities incorporating reference methods equivalent to the USEPA Reference Method 29 are under development, although they deviate strongly from the sampling QA/QC requirements of the method. Such deviations are partially due to a lack of reliable basic source test methods. Compliance testing protocols have yet to be developed.

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Mercury cycling through the environment can be complex and the tracing of mercury through different pathways can be difficult. These obstacles result in uncertainty when determining the precise relationship among emissions, atmospheric deposition and mercury concentration in fish. EPRI projects include those that assess the fate of mercury from source emissions leading to its ultimate arrival in fish. One such project is METAALICUS.

In addition to the cycling and transport of mercury, spatial factors (particularly non-U.S. sources) and the issue of legacy mercury emissions are other areas of uncertainty that need to be addressed before control strategies are designed and adapted. It was also noted that further research is needed to evaluate the links between specific source types and fish consumption advisories, as well as global contributions of mercury.

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Polar Sunrise: A Short Circuit in the Global Mercury Cycle

Mercury depletion events (MDE) may be a means for mercury from the global pool to accumulate in the Arctic. Studies undertaken in the year 2000 revealed the production of reactive gaseous mercury (RGM) during MDEs through the oxidation of mercury, resulting in high levels of RGM more typical of those seen near major point sources. It is suspected that reactive halogen chemistry may contribute significantly to MDEs. Additionally, sunlight and frozen surfaces also seem to be critical to these events (S. E. Lindberg, Brooks, S.B., C-J. Lin,

K. J. Scott, M. S. Landis, R. K. Stevens, M. Goodsite, and A. Richter. 2002. The Dynamic Oxidation of Gaseous Mercury in the Arctic Atmosphere at Polar Sunrise, *Environ. Sci. & Technol.* 36: 1245-1256).

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Pollution Probe

Science-Policy Interface in Mercury Reduction Standard-Setting

An overview of Pollution Probe projects aimed at reducing mercury pollution in Canada and North America include the first Canadian comprehensive mercury inventory (1996) and publication of the Mercury Primer (2002), among many others. Emphasis is placed upon science acting as a catalyst for policy making, laying the foundation for the design of appropriate policy frameworks. Policy frameworks developed in response to scientific research pertaining to mercury evolved in two distinct stages. In the first phase, federal, provincial/state governments took advantage of the immediately apparent opportunities for emission reduction, while cooperating and involving organizations such as the IJC and CEC, and developing enhanced emission inventories, highlighting the significant sources and uses available for possible further control.

The second phase raises many questions regarding the relative contributions from different sources of mercury, speciation of mercury emissions, the impact of natural sources and possible ecosystem response to additional reduction. All of these issues are viewed as critical by Pollution Probe. However, in light of these several uncertainties, the precautionary principle should be adopted. Importance of actions toward mercury emission reduction should not be delayed; rather, with the aid of models, assessment of when actions may be invoked, and what response might follow could be determined. Supporting evidence of similar situations where policy makers responded to the issues raised by science were presented.

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Environmental and Human Considerations Regarding Mercury: The Amazonian Project and the Canadian COMERN Initiative

Effects of mercury on human health as determined by studies on fish consuming populations living adjacent to portions of the Amazon River were reviewed. Subjects were selected based on comparisons of mercury concentrations in the hair of indigenous peoples whose diet was composed largely of fish. Methylmercury has a high affinity with the human brain and nervous system. Human health impacts correlated with relatively high mercury concentrations include a reduction in some characteristics of vision, particularly a decreasing ability to discern colours, as well as impaired dexterity.

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Putting Deposition in Context with other Sources and Pathways

The main route of human exposure to methylmercury is through fish consumption. Watersheds contain large pools of mercury which have accumulated over time. These watersheds directly or indirectly contribute to the accumulation of methylmercury in fish. The relationship between dissolved organic matter and mercury in stream water has been determined to be one factor in the control of mercury release from watersheds. Further, because these humic substances are associated with wetlands, it was suggested that the release of mercury would also be associated with wetlands. Additional discussion on the origin of mercury in watersheds revealed high concentrations of mercury on soil surfaces. It was noted however, that the interactions between mercury in soils and reductions in emissions and subsequent deposition and ultimate reduction of mercury in fish are quite uncertain and would require more investments in research and continued emission reductions.

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Atmospheric Quality & Meteorology Section

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Modeling Mercury on a Regional Basis

Critical elements of the regional mercury model used by the Ontario Ministry of Environment, the Acid Deposition Oxidation Model (ADOM) a Eulerian Model, encompass an emissions inventory, detailing speciation and natural emissions and re-volatilization. Other crucial elements include chemical schemes, dry deposition for elemental mercury and regional models of background concentrations. This model has been applied to Europe resulting in varying predictions of deposition of elemental and particulate mercury and mercury chloride.

Uncertainties exist among the individual species in terms of wet and dry deposition. As well, predictions using this model varied between the species. Predictions using the 1995 Norwegian Institute for Air Research emissions data for particulate mercury and reactive mercury were observed to be similar and lower than that derived with the 1990 emissions inventory data. However, elemental mercury predictions were more accurate using this preceding inventory data. Explanations for the variations are unclear.

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A Mercury Budget for Québec

Total Gaseous Mercury (TGM) studies, in addition to information on processes of water-air and soil-air exchange in Québec and mercury depletion events near the Arctic, provide an overview of the mercury budget in Québec.

TGM concentrations and related measurements determined at stations along the St. Lawrence River revealed that Southern Québec is a source region for mercury. Further analysis indicates the existence of temporal and spatial variations in air and precipitation concentrations. Water-air and soil-air exchange in Québec studies and estimations of the regional mass balance of mercury in the upper St. Lawrence River showed total mercury deposition over land is more or less in equilibrium with evasive flux of mercury; however river surface concentrations did not coincide with such estimates, suggesting mercury uptake, downstream transport or re-emission of mercury from biota or sediment.

Further presentation material focused on TGM and ozone concentrations measured in Kuujuaupik, in the lower Arctic region along the Hudson Bay (Québec).

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The 1996 and 1999 inventories of mercury sources in the United States draw on data from the National Emission Inventory (NEI). Emphasis was placed on the methodology behind the emissions estimates. The current national situation reveals a decrease in emission of mercury, largely due to a decrease in municipal waste combustion and medical waste incineration between years 1990 and 1999. On the other hand, certain U.S. county emission maps continue to show high mercury levels due to continued municipal waste combustion.

Another aspect of the inventory was addressed: mercury speciation. Inadequate data exist for estimation of the emissions of the three species of mercury and further improvements in speciation measurements are needed.

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Atmospheric Mercury Observations and Implications for Policy

In the United States, measurements of mercury deposition is carried out by the Mercury Deposition Network (MDN) and the National Atmospheric Deposition Program (NADP). The MDN, which has a focus on wet deposition, is one of three networks within the NADP. The MDN and NADP share similar characteristics which include Regional/National/International scope and uniform sampling and analysis procedures.

A monitoring study of mercury emissions in combustion plumes from power plants and municipal waste incinerators examining the physical and chemical transformations of mercury in such plumes in order to determine the species and volumes of released mercury is also discussed. Mercury monitoring observations in close proximity to sources are recommended to provide field data to support static plume dilution chamber studies, to better estimate mercury deposition immediately adjacent to known mercury emission sources.

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Acosta y Asociados

Atmospheric Mercury Emissions in Mexico

Mercury emission inventories in Mexico have been previously compiled by the Electric Power Research Institute (EPRI), and more recently by the Instituto Nacional de Ecología (INE). In 2000, the INE identified major sources of mercury and developed an approach to estimate usage, disposal and emission.

In the absence of emissions estimations for Mexico, EPRI relies on estimates based on emission factors of the EPA and Parcom-Atmos from the Netherlands. Estimates provided by Acosta y Asociados used two approaches which are drawn from the EPRI, INE and Parcom-Atmos inventories. These estimates suggest the largest mercury emission source is attributed to gold mining and refining. It was also noted that there is limited information on mercury emissions, which is partially due to the lack of regulatory processes in Mexico.

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Subcontinental Mercury Modeling - United States and Canada

Subcontinental Mercury Modeling, based on the 3-D regional Chemical Transport Model (CTM) TEAM, provides finer spatial resolution when compared with the continental model. Parameters used by the subcontinental model, i.e. meteorological, emission and chemistry inputs, are equivalent to those used by the continental model.

Mercury emissions are more important in the Eastern part of the U.S. and less so in Canada. Large variations are predicted in wet and dry deposition of mercury; this is due to factors such as wind direction, mercury chemistry and, in the case of wet deposition, cloud and precipitation effects.

The subcontinental model appears to be more accurate than the continental model for some sites (e.g., Minnesota and Wisconsin) but not so for others (e.g., Pennsylvania).

Uncertainty in mercury deposition in certain areas such as Pennsylvania, which is downwind of the Ohio valley (an area with large mercury emissions) may result from incorrect predictions of mercury chemical transformations. However, conversion processes that may occur between the emission sources and the receptor areas currently remain unknown. Thus, the subcontinental model does suggest that the finer spatial gradients of mercury deposition are not reproduced correctly at this time in global and continental models.

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The future of mercury reduction will rely on improved emissions data, in addition to better information in areas such as transport and fate of mercury and human health impacts. Further application of science in the fields of global studies and international strategies, pollution prevention, development of a mercury action plan, emission reductions from electrical utilities and further reductions from municipal and medical waste incinerators, and communication and outreach programs should also be considered. The U.S. has reaffirmed its commitment to reduce mercury emissions and policymakers have provided advice to scientists in response to this commitment.

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Global Source Overview

Estimates prepared by J.M. Pacyna and E.G. Pacyna were used to provide an overview of global emission sources. Since 1990, there has not been a significant change in total mercury released from anthropogenic sources to the atmosphere. Pacyna and Pacyna acknowledge that the 1995 data does not contain mercury emissions for gold production although it has been suggested that gold production is a significant contributor to mercury emissions. Despite the observation that total emissions did not significantly change, there was a variation in dominant sources and their geographic locations.

Decreased mercury emissions in Europe and North America could be attributed to prevention activities, installation of control equipment and procedures, and decreases in emissions from combustion sources. On the contrary, Asia has increased their mercury emissions, apparently largely due to coal combustion in China.

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Emission and Regional Scale Dispersion of Mercury in Eastern United States

Current limitations in the science of modeling atmospheric mercury include the inadequate consideration of prolonged mercury atmospheric residence time as compared with that associated with acid rain and ozone. Development of new models was suggested by the speaker.

A new inventory for the North Eastern United States is currently being assembled, drawing on several databases, and developing simulations using two European regional scale models - RAMS and SKIRON ETA. Results revealed significant discrepancies between the two models, which may be attributed to, among other factors, differences in the physics modules used in each model.

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Mercury in Mexico

Mexico is presently assembling an emissions inventory to further develop a capacity to analyze mercury. The installation of two wet deposition monitors, as well as a bank of human mercury samples is anticipated in the near future. Furthermore, the implementation of some quality assurance and quality control programs appears to be a possibility. Mexico will continue its present efforts in reducing mercury through remedial actions such as developing risk communication programs and appropriate recycling of mercury.

