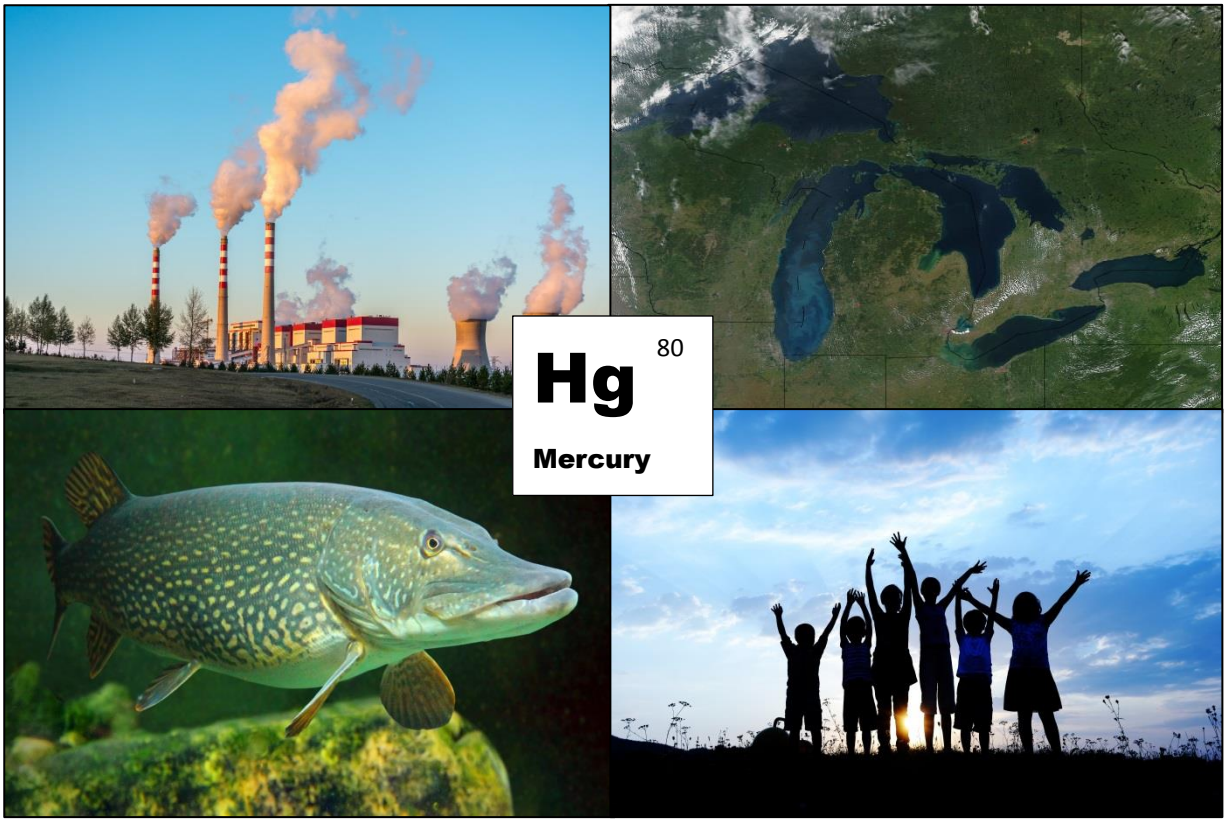
ATMOSPHERIC DEPOSITION OF MERCURY IN THE GREAT LAKES BASIN



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Executive Summary

After several decades of effective action by Canada and the United States to address sources of mercury within the Great Lakes Basin, the need to address atmospheric deposition of this toxic substance from out-of-Basin regional and global sources is increasingly evident. Achieving the general objective of the Great Lakes Water Quality Agreement that human consumption of fish should be unrestricted by concerns due to harmful pollutants will require reductions in atmospheric loadings of mercury from distant as well as regional and local sources. Continued strong efforts by Canada and the U.S. are needed to coordinate action at the international level, supported by sustained monitoring efforts within the Great Lakes basin to determine the effectiveness of such action. Monitoring mercury pollution is a critical need in light of persistent mercury contamination of Great Lakes fish, particularly as concern rises that emissions outside of the U.S. and Canada could, through long-range atmospheric transport, diminish or offset progress made by the two nations in reducing domestic emissions.

Atmospheric deposition has been recognized as a major pathway of persistent, bioaccumulative substances (PBTs) entering the Great Lakes since the 1970s. Canada and the United States acted on this knowledge by providing for monitoring of atmospheric deposition of these substances in Annex 15 of the Great Lakes Water Quality Agreement (Agreement) revised by protocol in 1987. The bi-national Integrated Atmospheric Deposition Network (IADN) that resulted from Annex 15 has provided valuable information on loadings and trends of PBTs entering the Great Lakes from the air since 1990.

IADN, however, does not routinely measure mercury deposition. The Great Lakes region lacks an adequately and sustainably-funded, geographically diverse and consistently maintained mercury atmospheric deposition monitoring network. Meanwhile, mercury levels in some fish and wildlife species in some locations of the Great Lakes Basin region are increasing. Although there have been dramatic reductions in local and regional air emissions of mercury and further reductions are expected, increased mercury loadings from global sources are being deposited in the Great Lakes region. This could offset a substantial amount of the local and regional emissions reductions. Tracking sources and concentrations of mercury associated with atmospheric deposition in the Great Lakes region will be vital to crafting effective mercury reduction strategies.

The Commission is charged under the Agreement with advising the Parties on "approaches and options that the Parties may consider to improve effectiveness in achieving the purpose and objectives" of the Agreement and to advise on "research and monitoring of the Waters of the Great Lakes, including recommendations for specific research and monitoring priorities." The Commission wishes to underscore the need for continued vigilance regarding mercury in the Great Lakes Basin, particularly in fish, and associated human health risks. Such vigilance requires recognition of the important role of out-of-Basin and global air transport of these substances.

Therefore, the Commission:

- Recommends that governments increase and provide sustainable funding for an optimized binational monitoring network to track atmospheric deposition of mercury in the Great Lakes Basin as well as funding for modeling to allow for source attribution.

- Commends governments for their positive action with respect to pursuing global mercury reduction policies, including support for the mercury-focused Minamata Convention.

- Recommends that Canada and the U.S. support and advocate international actions, including additional multilateral global agreements to reduce loadings of persistent bioaccumulative toxic substances (PBTs) in addition to mercury that reach the Great Lakes Basin through the atmosphere from other continents. Such actions should supplement current domestic programs that are reducing local and regional atmospheric transport of these substances.

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The Role of Atmospheric Deposition in Pollution of the Great Lakes

Atmospheric deposition is a major pathway of persistent, bioaccumulative toxic substances (PBTs) to the Great Lakes.[1] The discovery of two PBTs, PCBs and DDT, in fish in Siskiwit Lake on Lake Superior's Isle Royale in the 1970s was a significant turning point in understanding the role of atmospheric transport and deposition of toxic substances (Swain 1978). Pollutants were clearly not being discharged directly by factories or sewage plants into this remote lake on an island in a national park, and other sources were ruled out, leaving the atmospheric deposition as the most probable pathway of contaminant delivery.

Understanding of the science of atmospheric deposition processes has grown considerably since then. It is now well established that pollutants can move from the atmosphere directly into surface water and indirectly via the landscape through wet (pollutant absorption into precipitation) and dry (pollutant adsorption onto falling particles, and gaseous air-ground surface exchange) deposition processes. Since persistent pollutants break down slowly, they can travel long distances in ambient air before depositing onto land or water.

Some PBTs, including mercury, that deposit to the ground or water may revolatilize back into the atmosphere and this cycle can repeat, with the chemicals traveling to cooler places due to global air mass transport patterns. This is sometimes called the "grasshopper effect" or global distillation. As a result, high levels of some PBTs can be found in the Arctic, far away from cities and factories (EPA 2013a). The Great Lakes themselves can thus become sources of atmospheric pollution, as contaminants revolatilize from the Lakes.

On land, atmospheric PBTs can enter the ecosystem after being adsorbed onto vegetation (e.g., growing leaves or leaf litter) which then decomposes on the forest floor and can be ingested by insects and songbirds.

The Great Lakes and many inland water bodies across the Great Lakes region continue to suffer impairments due to atmospheric inputs (GLC 2006). About 90% (i.e., 53,300 of 60,500 square miles) of Great Lakes surface area in the U.S. is reported to be impaired under Clean Water Act standards at least in part because of atmospheric deposition of mercury and other pollutants (U.S. GAO 2013).

In the U.S., Total Maximum Daily Load (TMDL) plans are required to be developed when pollutants in a water body exceed U.S. Clean Water Act water quality standards. The plans allocate reductions in pollution loadings to various sources to meet the limit of pollution that would bring the water body into conformance with water quality standards. While historically plans were developed for individual water bodies, in the past decade, some states have been pursuing regional TMDLs, in particular for pollutants with a significant atmospheric contribution. For example, Michigan's Department of Environmental Quality prepared draft Statewide Total Maximum Daily Load (TMDL) plans for dealing with atmospheric sources of mercury and PCBs (MDEQ 2013a and 2013b).

[1] Persistent, bioaccumulative and toxic pollutants (PBTs) are long-lasting substances that can build up in the food chain to levels that are harmful to human and ecosystem health. Because of their persistence and bioaccumulative properties, they do not break down easily and are particularly difficult to remove from the environment.

The Michigan mercury inland lakes TMDL document observes that atmospheric deposition is by far the largest contributor to current mercury loadings in the state and that in-state sources, such as permitted emissions from coal-fired power plants, make up only 7.8% of the state's atmospheric mercury load, while out of state sources make up the remaining 92.2% (MDEQ 2013a). Further, 75.3% of atmospheric mercury deposition to Michigan originates from background sources (natural sources as well as anthropogenic sources outside of North America). To meet the TMDL, the State's contribution to emissions must be reduced by 82%, along with the same degree of reduction in out of state anthropogenic sources contributing to Michigan deposition. Yet Michigan cannot directly influence an 82% reduction in out-of-state mercury emissions.

Long-distance atmospheric transport and deposition of mercury affects Canada in similar ways. An estimated 95% of man-made mercury deposited in Canada comes from foreign sources (Environment Canada 2013c).[2]

Given the significant role atmospheric deposition plays in mercury pollution of the Great Lakes and inland waters in the Basin, it is clear that attainment of water quality standards for mercury will require policies and actions at the regional, binational and global levels to curb emissions and atmospheric transport of these pollutants.

Mercury Contamination

After many years of declining mercury levels in fish and other Great Lakes biota, concentrations have generally leveled off or slowly increased in some species in some locations.

Mercury is ubiquitous in the Great Lakes environment. It is of primary concern because of its effect on human health, with elevated human exposure usually resulting from consumption of contaminated fish. For fetuses, infants, and children, the primary health effect of methylmercury (an organic form of mercury that is most bioavailable) is impaired neurological development. Methylmercury exposure in the womb, which can result from a mother's consumption of fish that contain methylmercury, can adversely affect a baby's growing brain and nervous system. Impacts on cognitive thinking, memory, attention, language, and fine motor and visual spatial skills have been seen in children exposed to methylmercury in the womb (EPA 2013b).

Domestic Canadian and U.S. efforts to control mercury began in earnest in the 1960s after the outbreak of so-called Minamata disease in Minamata, Japan in 1956 and the pollution of the English-Wabigoon River system in Canada in 1969. Minamata residents who ingested fish and shellfish contaminated by methylmercury discharged in waste water from a chemical plant suffered severe neurological symptoms and some women who consumed the contaminated fish gave birth to children with birth defects (Ministry of the Environment, Japan, 2002). Similarly, in 1969, a pulp and paper mill near Dryden, Ontario, discharged mercury to the English-

[2] Mercury contamination has also been recognized by the Commission in the transboundary Lake of the Woods-Rainy River Basin immediately to the west of the Lake Superior Basin. In its Plan of Study submitted to the Canadian and U.S. governments, the International Lake of the Woods Basin Water Quality Study Team (covering the Lake of the Woods-Rainy River Basin) included a project calling for the assessment of loading (or flux) and bioaccumulation of mercury and methylmercury in key border waters in the Basin.

Wabigoon River, polluting the fish and making them unfit to eat. The White Dog and Grassy Narrows First Nations people depended on the fish, and experienced high levels of mercury in their blood and hair. Levels of mercury in the region remained elevated for decades (Parks and Hamilton, 1987).

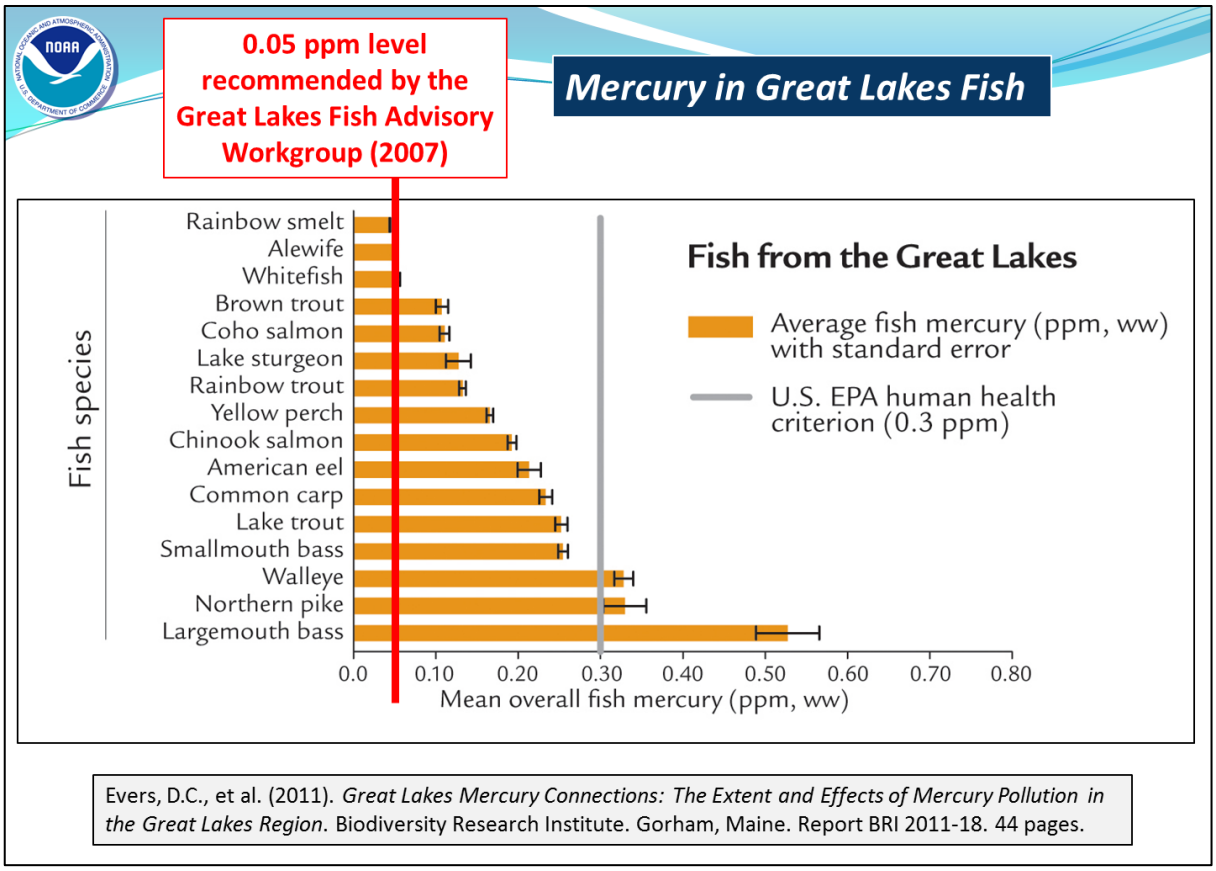

Figure 1. *Credit: NOAA Air Resources Laboratory*

Mercury is responsible for fish contaminant advisories for portions of all five Great Lakes (EPA 2015a). In Ontario's Great Lakes waters, mercury accounts for 25% of advisories for Lake Superior, 21% for Lake Huron, 40% for Lake St. Clair and the St. Clair River, 11% for Lake Erie and 12% for Lake Ontario (OMNR 2015).

In addition, mercury is responsible for statewide fish contamination advisories urging limited consumption of some species for virtually all inland lakes in Michigan and Minnesota (MDCH 2015), as well as some inland waters in each of the other Great Lakes states. Some 189 tested water bodies in Michigan exceed water quality standards for mercury (Drevnick 2015).

Mercury exposure remains a particular concern for infants, children and subsistence anglers in the Great Lakes Basin. A study conducted by the Minnesota Department of Health from 2007 to 2011 found that 8% of 1,465 tested newborns in three states had mercury levels in blood above the safe dose limit set by U.S. EPA. The agency said, "This means that some pregnant women in the Lake Superior region have mercury exposures that need to be reduced." No Michigan samples were above the U.S. EPA dose limit. However, 3% of the Wisconsin

samples were above the limit, and 10% of the Minnesota samples had elevated levels of mercury. It is hypothesized that locally-caught fish consumption rates are higher in mothers of the newborns tested in Minnesota (Minnesota Department of Health 2013).

Mercury contamination also affects fish and wildlife health. EPA observes, "Effects of exposure on wildlife can include mortality, reduced fertility, slower growth and development and abnormal behavior that affects survival, depending on the level of exposure. In addition, research indicates that the endocrine system of fish, which plays an important role in fish development and reproduction, may be altered by the levels of methylmercury found in the environment" (EPA 2015b).

Mercury emitted from anthropogenic sources, largely as gaseous elemental mercury, can remain in the atmosphere for six months to a year, enabling long-distance transport. Gaseous elemental mercury can be transformed to oxidized inorganic mercury species that are much more likely to be deposited via wet and dry deposition to the earth's surface. Some of the mercury deposited to terrestrial and marine ecosystems can be converted to methylmercury by bacteria, and the mercury then increases in concentration as it moves up the food chain (USGS 2009) (Pirrone et al. 2013). Large predator fish such as walleye and trout can have mercury concentrations one million times or more higher than that of the surrounding water.

Trends

In Canada and the U.S., initial regulatory efforts under water pollution control laws enacted in the 1970s focused on sources of direct discharges of mercury to water, such as chlor-alkali factories and sewage treatment plants. Controls on these and other sources, such as mercury-based paint and mercury used in pesticides, contributed to reductions in levels of mercury in the environment. Coal-fired power plants now contribute over 50% of all human-caused mercury emissions in the U.S. but federal initiatives are underway to reduce domestic mercury emissions from coal combustion, and some older coal-fired power plants are being retired or converted to natural gas.

Nonetheless, there are trends of concern. The Commission's 16th Biennial Report notes that levels of mercury in whole fish caught in the Great Lakes basin have been generally stable or increasing since about 1990 (IJC 2013). Summarizing research presented in a special 2011 issue of *Ecotoxicology*, Evers et al (2011) concluded that "(1) mercury remains a pollutant of major concern in the Great Lakes region, (2) that the scope and intensity of the problem is greater than previously recognized and (3) that after decades of declining mercury levels in fish and wildlife concentrations are now increasing in some wildlife species and areas."

Referring to mercury concentrations in whole body lake trout or walleye collected from each of the Great Lakes, EPA notes studies showing that "generally, the declines in mercury concentrations observed up until approximately 1990 have ceased and that mercury concentrations in fish have started to increase. Environment Canada and U.S. EPA data were used in the analyses of both studies and correspond with their findings. This suggests that concentrations of mercury in top predator fish are atmospherically driven and the recent increases may be a reflection, in part, of increased global mercury emissions." (EPA 2014b).

The Great Lakes ecosystem is the most intensively studied system in Canada with respect to mercury concentrations in the aquatic environment. Mercury concentrations in most biota from the system generally declined from 1972 until the early 1990s. After that time, some biota in the upper Great Lakes (Superior and Huron) continued to show declines in mercury concentrations, such as lake trout and walleye (Bhavsar et al., 2010), presumably in response to a reduction in mercury emissions. However, mercury concentrations in lake trout and walleye collected by Environment Canada and the U.S. EPA showed an increase in concentrations over the same time period (McGoldrick et al., 2012), while biota from the lower Great Lakes (Erie and Ontario) show a levelling out or increase in mercury concentrations (Bhavsar et al., 2010; McGoldrick et al., 2012).

Figure 2. *The Ontario Power Generation's Nanticoke Generating Station (2007) on Lake Erie stopped burning coal on December 31, 2013. Credit: Ontario Power Generation*

Several hypotheses in addition to increased global emissions have been offered, including changing climate, lower water levels and greater exposed shoreline associated with drought, land use changes, acidification that changes the methylation potential of the aquatic ecosystem, and changes in food webs associated with invasive species. For example, authors of one study suggested modifications in Lake Erie's food web due to invasions of mussels and the round goby could be partially responsible for increases in mercury in Lake Erie walleye (Bhavsar et al. 2010). Authors of another paper hypothesized that increases in temperature, rainfall intensity,

runoff, and water-level fluctuations could alter either the methylation of mercury or the mobilization of methylmercury (Wiener et al. 2007).

Stable or increasing concentrations in mercury in Great Lakes Basin fish and wildlife come at a time when domestic mercury emissions have declined. Total mercury emissions to the atmosphere from inventoried anthropogenic sources in the Great Lakes states declined by approximately 50% between 1990 and 2005 (Evers et al. 2011). Total Canadian mercury emissions decreased 85% between 1990 and 2010. U.S. mercury emissions decreased approximately 60% between 1990 and 2005. U.S. EPA's new Mercury and Air Toxics Standards (MATS) are expected to further reduce mercury emissions from coal-fired power plants by about 90% (EPA 2014a). However, a recent U.S. Supreme Court decision has created uncertainty about continued implementation of MATS[3]. On the other hand, the Obama Administration's August 2015 issuance of a Clean Power Plan rule, designed to reduce carbon emissions 32% from 2005 levels by 2013, will have the side benefit of reducing mercury emissions, since it targets coal-fired power plant emissions (EPA 2015c). Meanwhile, Canada-wide standards helped reduce emissions from Canadian coal-fired power plants by about half between 2008 and 2012 alone (Canadian Council of Ministers of the Environment 2014).

But global anthropogenic emissions have begun to climb, with significant increases in Chinese and other Asian emissions offsetting emission declines in North America and Europe (UNEP2013). Asia posted the largest increases in mercury emissions from 1990 to 2010 largely due to expanding energy production from coal-fired power plants, and accounts for almost half of global releases. According to the United Nations Environment Programme, another significant source of global mercury emissions is artisanal gold mining.[4] Estimated emissions from this source have doubled since 2005, in part due to new and better information, but also due to rising gold prices and decreasing mercury prices that are expected to lead to further emission increases (UNEP 2013).

Atmospheric fate and transport models can be used to estimate source-attribution for Great Lakes mercury deposition. One recent study (Cohen et al. 2015) estimated that U.S. anthropogenic sources contributed on the order of 20-25% of 2005 deposition to the Great Lakes basin, using different simulation methodologies. Chinese anthropogenic sources contributed 6-8%, and all other global anthropogenic sources contributed 8-10%. The remainder of the deposition came from oceanic natural emissions and re-emissions of previously deposited mercury (25-35%), terrestrial natural emissions and re-emissions (19-26%), biomass burning (~4%) and geogenic emissions (~4%).[5]

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[3] (Michigan v Environmental Protection Agency, U.S., 135 S.Ct. 702 (Mem), 135 S.Ct. 2699, 192 L.Ed.2d 674, 83 USLW 4620 (2015).).

[4] Artisanal mining refers to activities using rudimentary methods to extract and process minerals and metals on a small scale. Artisanal gold miners combine mercury with gold-carrying silt to form a hardened amalgam that picks up most of the gold metal from the silt. The amalgam is later heated with blowtorches or over an open flame to evaporate the mercury, leaving the gold.

[5] Cohen notes both oceanic and land-based mercury emissions have a strong anthropogenic re-emissions component, but he did not attempt to attribute portions of this to specific countries in his 2015 paper. In a 2011 study that did estimate such re-emissions, Cohen estimated that China (14%) was second to the U.S. (32%) in contributing mercury to the Great Lakes basin. Other countries in decreasing order of mercury contribution were Canada, India,

Source attribution results varied for the different lakes, with Lake Erie showing the highest contribution from U.S. anthropogenic sources and Lake Superior the lowest. These results are generally consistent with other modeling efforts that have attempted to provide source-attribution estimates for atmospheric mercury deposition in North America (e.g., Selin and Jacob, 2008; Lei et al., 2013; Grant et al., 2014). Natural sources of mercury emissions include forest fires, volcanoes, geothermal sources and topsoil enriched in mercury, among others (Pirrone et al 2010). U.S. EPA modeling found that the proportion of mercury deposition in the Great Lakes contributed by non-U.S./Canadian sources is more than 87.5% in Lake Superior, and by non-U.S. sources less than 62.5% in Lake Erie (GLRC 2010).

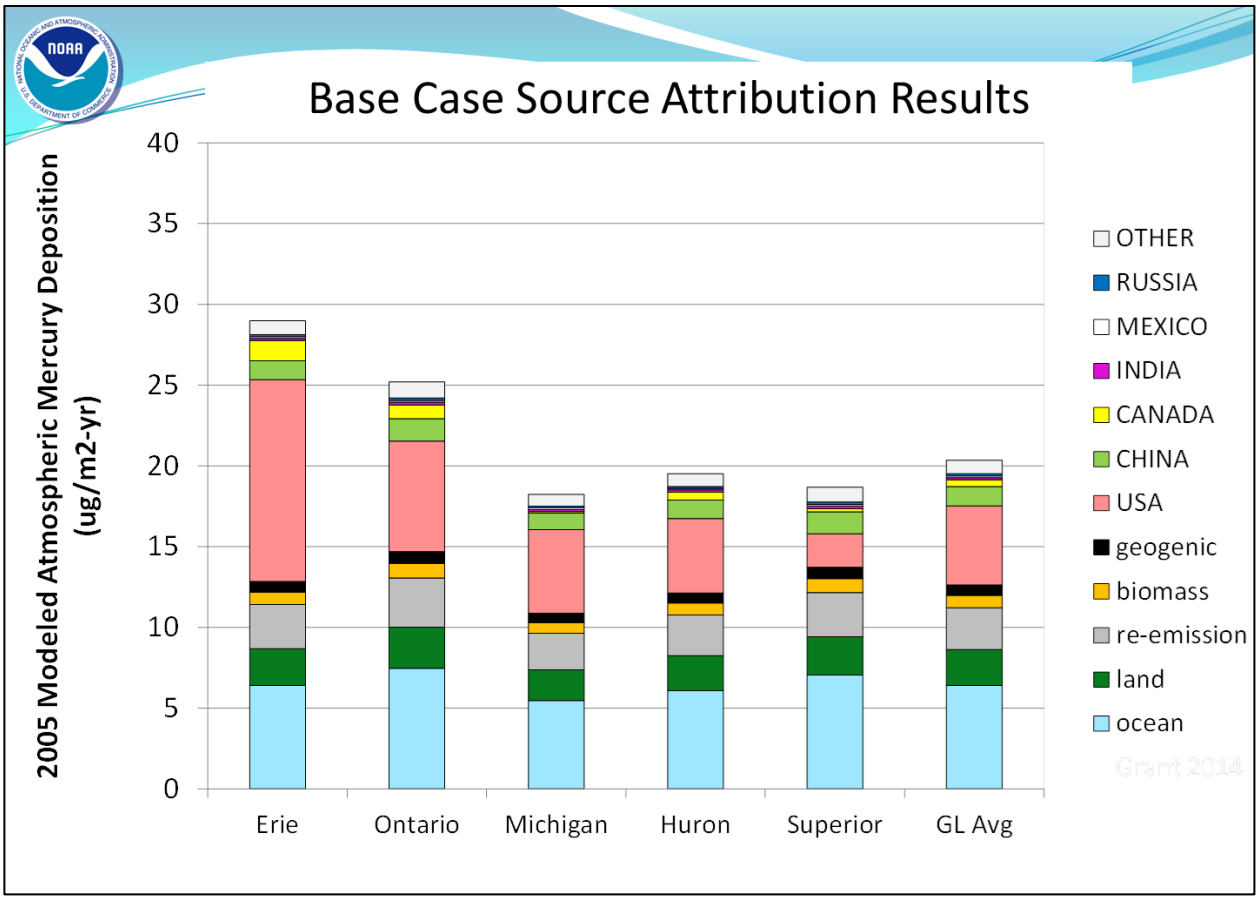

Figure 3. *Credit: NOAA Air Resources Laboratory*

The fact that considerable mercury contaminating the Great Lakes Basin originates off-continent and is transported a great distance through the atmosphere has important policy implications. Some models suggest that reductions in domestic mercury emissions resulting in lowered mercury concentrations in fish tissue could be partially or completely offset by growth in non-U.S. mercury emissions (Vijayaraghavan et al 2014). Therefore, additional efforts to

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Russia, Mexico, Indonesia, South Korea, Japan and Colombia. Together, these other countries accounted for an estimated 15% of the mercury entering the Great Lakes.

reduce mercury emissions around the world are increasingly important to the health of the Great Lakes ecosystem. However, North American anthropogenic source influences have been significant in the past and are still significant – especially in some industrialized regions – today.

In summary:

- The scope and impact of mercury pollution in the Great Lakes watershed is of increasing concern.
- Reductions in domestic mercury emissions will continue to be significant in protecting Great Lakes fish, wildlife and human health.
- Efforts to reduce mercury emissions on a global scale will benefit the Great Lakes.

Mercury Monitoring Networks

Since much of the current mercury loading to the Great Lakes enters the Lakes through atmospheric deposition, it is important to monitor mercury in wet and dry deposition and gaseous phases in ambient air. Modeling is also important in order to fill in spatial and temporal gradients not captured by monitoring and to estimate attributions of various sources.

Long-term, sustained funding for mercury monitoring in the Great Lakes Basin is problematic. Although there has recently been progress in funding for tracking of wet deposition in the U.S., this funding support is assured only in the short term.

The Lake Michigan Air Directors Consortium (LADCO), consisting of air quality program managers from Wisconsin, Illinois, Indiana, Michigan, Minnesota, and Ohio, channeled approximately $330,000 in existing funding in 2014 and 2015 from the Great Lakes Atmospheric Deposition (GLAD) program to a Great Lakes wet-deposition mercury monitoring program consisting of 21 sites. The network consists of 12 already active Mercury Deposition Network (MDN) sites in Illinois, Minnesota, New York, Ontario, Pennsylvania, and Wisconsin, eight existing but formerly inactive MDN sites in Illinois, Indiana, Michigan, and Ohio, and one new MDN site in Ohio. Newly activated monitoring sites began measurements in 2014. The LADCO initiative should generate critical and statistically valid data on wet deposition of mercury and gaseous mercury.

Year-to-year funding has been made available for most of the LADCO-sponsored monitoring for fiscal year 2016. But monitoring will be discontinued at 1 site in Illinois, 1 site in Michigan, and 2 sites in Wisconsin at the end of 2015. Monitoring at 11 sites in Ohio, Indiana, and Minnesota will be continued for fiscal year 2016. LADCO continues to pursue funding to operate all sites that operated in 2014-2015.

The LADCO funding will augment limited mercury monitoring relevant to the Great Lakes in the U.S. and Canada:

Mercury Deposition Network (MDN): MDN is considered the only network providing a long-term record of total mercury concentration and deposition in precipitation across the United States and Canada. The MDN began measuring total mercury in precipitation in 1996. It now includes more than 100 sites, including the LADCO sites mentioned earlier.

As many as 51 mercury wet-deposition monitoring sites from 4 networks were operated in the Great Lakes states and Ontario from 1996 to 2010. By 2013, 20 of those sites were no longer in operation (Risch et al 2014). The number of active mercury monitoring sites in the U.S. portion of the Great Lakes region had diminished by 37 % during the previous 15 years

(Risch et al 2014). Only a single Mercury Deposition Network (MDN) site was operated to represent half the geographic area of the Great Lakes region in Illinois, Indiana, Michigan's Lower Peninsula, and Ohio. The LADCO funding temporarily closes much of that gap.

MDN monitoring sites which have been consistently maintained and have generated long-term data are located in Wisconsin, Minnesota, Illinois and Indiana.

Atmospheric Mercury Network (AMNet): Operating 20 sites in the U.S., AMNet monitors the atmospheric concentrations of speciated mercury fractions -- gaseous elemental HG (GEM), Gaseous Oxidized HG (GOM) and particulate Bound Hg (PBM) -- and supports dry deposition estimates, emission regulatory assessments, model evaluation, and long-term trends. Three AMNet sites are in the Great Lakes Basin.

The Canadian Air and Precipitation Monitoring Network (CAPMoN): As part of a long term monitoring effort, CAPMoN measures the concentration of mercury in precipitation and total gaseous mercury in air at a specific number of sites. In 2010 there were 6 CAPMoN sites collecting 7-day integrated precipitation samples specifically for the purpose of determining regional scale mercury concentrations in precipitation. CAPMoN operates these sites across Canada as part of the Mercury Deposition Network (MDN) described above. CAPMoN uses the same standard operating procedures, equipment and methods to provide a seamless database for mercury wet-deposition fluxes.

In 2010 there were 4 CAPMoN sites conducting continuous measurements of total gaseous mercury reported as hourly averages through the National Atmospheric Chemistry Data Base. Starting in 2010, speciated atmospheric mercury has also been measured continuously at one CAPMoN site (Keijimkujik) by the Atlantic Region of Environment Canada. The speciated mercury site is part of the Atmospheric Mercury Network (AMNet) mentioned above. Long-term speciated mercury measurements are made by Environment Canada at Alert, Nunavit and several sites operated by academic institutes (Environment Canada 2015).

Mercury dry deposition fluxes need to be estimated from monitored speciated atmospheric mercury concentrations and modeled dry deposition velocities. No sites in Canada measuring speciated mercury are in the Great Lakes region.

These networks have attempted to estimate TGM and/or wet deposited mercury. One of the components missing from the networks and the LADCO initiative is the quantification of dry deposited mercury because of lack of instrumentation. Depending upon location and time of year, dry deposited mercury is important to monitor (Zhang et al. 2012). Unfortunately, while some short-term (days to week) intensive studies reporting dry deposition values of mercury have been performed, no long-term data sets exist; thus, there are no monitored assessments of long-term trends in mercury dry deposition for the Great Lakes (F. Marsik, June 10, 2013, personal communication; GLC 2007).

Current atmospheric mercury fate and transport models must make assumptions regarding the rates of dry deposition for various species and phases of mercury, often assuming that the different phases of mercury (gaseous and particulate) act similarly with respect to deposition. Having measurements to compare against these models would be of great benefit to verify or alter modeled mercury dry deposition numbers.

None of the monitoring networks described above are of sufficient resolution or coverage to yield useful trend data for the Great Lakes region. Aware of the gap, the Great Lakes Commission (GLC) has called for a regional Great Lakes mercury monitoring network. A 2010 resolution called for "efforts in the United States and Canada to authorize and fund

comprehensive, collaborative, long-term mercury monitoring and research programs within and outside of the Great Lakes-St. Lawrence River basin" (GLC 2010). Legislation introduced in the U.S. Congress to establish a nationwide network languished.

An optimal Great Lakes mercury monitoring network would consist of at least 21 stations chosen for geographic representativeness, significant mercury trends, or a high concentration of mercury sources or deposition (Risch et al 2014). These sites would be maintained with consistent funding over long periods of time, enabling standardization of data and analysis of temporal trends. The estimated total annual operating costs for 21 sites would be $250,000 US (D. Gay, October 9, 2015, personal communication).

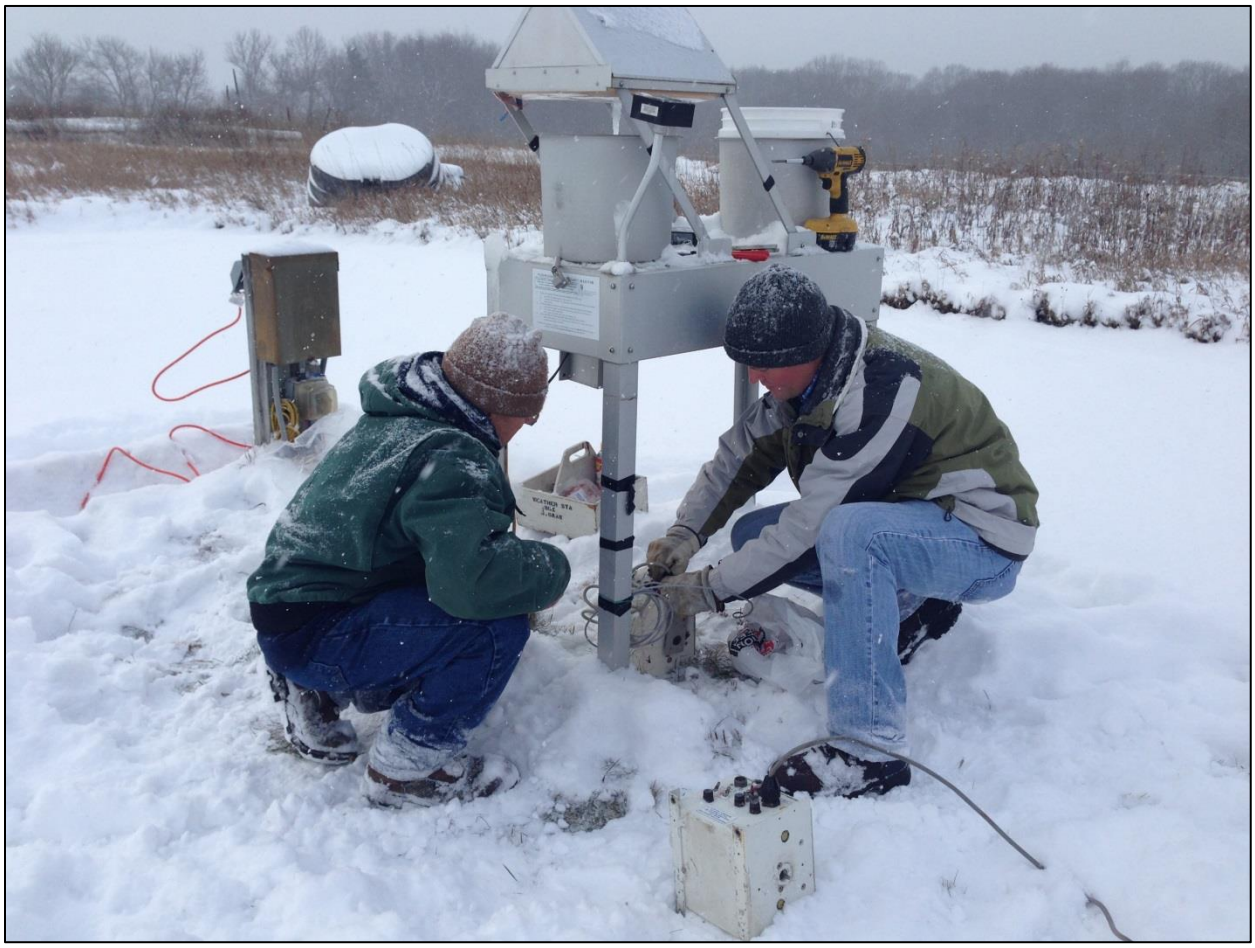

Figure 4. *Workers install a wet deposition sampler to track atmospheric deposition of mercury at the Kellogg Biological Station in Michigan in December 2013.*
Credit: Lake Michigan Air Directors Consortium

In addition to the optimized monitoring network, funding for modeling of atmospheric deposition in the Great Lakes region is needed. Modeling facilitates understanding of atmospheric deposition of mercury, supporting estimates of dry deposition, as well as total deposition drawing from data supplied by monitoring. It also supports source attribution through the use of comprehensive fate and transport models (Cohen et al 2007).

The speciation of mercury emissions from coal-fired power plants into different mercury forms (elemental, reactive gaseous, and particulate) has been discontinued, starting with EPA's 2008 National Emissions Inventory (NEI) (M.D. Cohen, October 5, 2015, personal communication). Speciation of emissions is a key input to mercury models.*

The lack of a systematic evaluation and optimized long-term monitoring design inhibits understanding of trends and sources, and inhibits appropriate public policy responses. Further, without a dedicated Great Lakes mercury monitoring network maintained over time, researchers' ability to test the intricacies of model predictions, including the importance of regional sources, is limited. In summary, gaps in mercury deposition monitoring make it difficult to discern where the contaminant is coming from and the most effective method to control it.

Atmospheric Deposition and the Great Lakes Water Quality Agreement

Although the 2012 renewal of the Agreement does not include an Annex dedicated to atmospheric deposition of toxic substances as its 1987 predecessor did, it does recognize the contribution of atmospheric deposition of pollutants in Annex 3, Chemicals of Mutual Concern, committing the Parties to:

- identify and assess the occurrence, sources, transport and impact of chemicals of mutual concern, including spatial and temporal trends in the atmosphere, in aquatic biota, wildlife, water, and sediments;
- identify and assess loadings of chemicals of mutual concern into the Waters of the Great Lakes from all sources including point sources, non- point sources, tributaries, and the atmosphere.

Further, the Agreement recognizes "that international efforts may contribute to reductions of releases of chemicals of mutual concern from out-of-basin sources that are deposited within the Great Lakes Basin Ecosystem."

The annex is implemented by a subcommittee co-led by Environment Canada and the U.S. EPA. Organizations serving on the subcommittee include the Great Lakes Indian Fish and Wildlife Commission, Indiana Department of Environmental Management, Minnesota Department of Health, Ontario Ministry of the Environment and Climate Change, the U.S. Agency for Toxic Substances and Disease Registry and the Wisconsin Department of Natural Resources.

An Annex 3 task team is evaluating a set of candidate chemicals identified by Canada and the United States using a series of binational considerations established by the Annex 3 Subcommittee and has made recommendations regarding their potential designation, by Canada and the United States, as binational Chemicals of Mutual Concern (CMCs). On May 13, 2015, the task team released summary reports characterizing several candidate CMCs, including mercury. (Great Lakes Executive Committee 2015).

* Errata: replaced text: "Further, the updating of mercury emissions inventories formerly supported by U.S. EPA funding has been largely discontinued and should be reinstated. The inventories are key inputs to mercury modeling (M.A. Cohen, October 5, 2015, personal communication)."

Commission Advice to Governments

Based on the analysis in this report, the Commission offers the following advice to the Canadian and U.S. governments:

Governments should increase and provide sustainable funding for an optimized monitoring network of atmospheric deposition of mercury in the Great Lakes Basin and for modeling to allow for source attribution. As mercury control strategies are implemented within both Canada and the United States, the monitoring of trends in atmospheric deposition of mercury will be critical in the assessment of their success, and can inform international discussions involving Canada and the U.S. on mercury control strategies.

The U.S. and Canadian mercury network should provide reliable funding for monitoring wet and dry mercury deposition and ambient air measurements. Funding should support monitoring at a sufficient number of sites over time to yield statistically valid trend information and to assess the efficacy of both in-Basin and national existing and future mercury controls. The estimated annual cost of operating the monitoring network is $250,000 (US) for 21 sites in Great Lakes states, with additional funding required for Ontario sites.

As a first step, the Commission recommends that a coordinated assessment of basin-wide mercury monitoring be undertaken to identify spatial and temporal gaps in current network capacity and the range of forms of mercury measured. Research should focus on improving mercury budgets for the Great Lakes Basin and each of the Great Lakes towards development of ecological and human health risk assessment frameworks.

The Commission commends governments for their positive action with respect to pursuing global mercury reductions policies, including support for the mercury-focused Minamata Convention.

In January 2013, 140 countries, including Canada and the U.S., reached agreement on a treaty intended to protect human health and the environment from anthropogenic emissions and releases of mercury and mercury compounds. The treaty aims to lead to the global phase-out of mercury in a variety of products and processes over the next decade, eventually ban primary mining of mercury, and promote the use of the latest technologies in controlling emissions and releases from various industrial sources. Officially titled the Minamata Convention on Mercury, the agreement was opened for signature at a diplomatic conference in Japan in October 2013. The convention will enter into force once 50 countries have ratified it (as of October 2015, 19 countries had ratified).

As previously noted the U.S. and Canada have already begun implementation of many measures called for in the convention, such as a requirement to install best available technologies on new coal-fired power plants, waste incineration plants and cement factories. Both governments have issued public statements praising the convention (U.S. Department of State 2013, Environment Canada 2013c). Once the treaty comes into force, an effectiveness evaluation mechanism must be in place that includes national monitoring programs to assess the levels of mercury reductions from implemented controls.

The IJC is convinced of benefits to the Great Lakes ecosystem resulting from implementation of the Minamata Convention due to future reductions of long-range atmospheric transport of mercury. U.S. and Canadian support for the Convention demonstrates a commitment to

continue reducing mercury contamination and could spur similar commitments in nations whose emissions contribute to Great Lakes atmospheric deposition and speed global implementation.

The Commission recommends that Canada and the U.S. support and advocate international actions, including additional multilateral global agreements to reduce loadings of persistent bioaccumulative toxic substances (PBTs) in addition to mercury that reach the Great Lakes Basin through the atmosphere from other continents. Such actions should supplement current domestic programs that are reducing local and regional atmospheric transport of these substances.

As the case of mercury demonstrates, future regulatory actions to control other PBTs such as pesticides, flame retardants and other chemicals of concern, including some banned in the U.S. and Canada but still in use elsewhere, must address the entire airshed of influence on the Great Lakes, including global sources.

Global actions will likely be required to reduce levels of PBTs such as pesticides and flame retardants in Great Lakes Basin sportfish to the point where fish consumption advisories can be removed and related beneficial uses restored. Governments should thus step up efforts to assist in elimination or reduction of use of these chemicals worldwide. In their 2016 Progress Report of the Parties under the Agreement, the Canadian and U.S. governments should report on global contributions to the Great Lakes via atmospheric deposition of chemicals of mutual concern. Achieving the objectives of the Great Lakes Water Quality Agreement will require global effort to reduce emissions of PBTs.

Conclusion

As Canada and the U.S. continue to realize significant emission reductions of mercury, the importance of monitoring and modeling atmospheric deposition of mercury will increase. Understanding the source regions of mercury will enable the two nations to devise appropriate, cost-effective control strategies. It will also underscore the importance of controlling emissions globally and support international cooperation to attain that objective.

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