Great Lakes Mercury Connections

The Extent and Effects of **Mercury Pollution** in the Great Lakes Region

A publication of the Biodiversity Research Institute
in partnership with the Great Lakes Commission
and the University of Wisconsin-La Crosse

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About this report:

The Great Lakes Commission sponsored a binational scientific synthesis effort through its U.S. Environmental Protection Agency-funded Great Lakes Air Deposition program. The purpose of the synthesis project was to foster binational collaboration among mercury researchers and resource managers from government, academic, and non-profit institutions to compile a wide variety of mercury data for the Great Lakes region, and to address key questions concerning mercury contamination, the bioaccumulation of methylmercury in food webs, and the resulting exposures and risks.

The synthesis effort began in November of 2008 and has involved more than 170 scientists and managers working to compile and evaluate more than 300,000 mercury measurements and to conduct new modeling and analyses. This synthesis provides a comprehensive overview of the sources, cycling, and impacts of mercury in the Great Lakes region. The primary results of this initiative have been published in a series of more than 35 scientific papers in the journals *Ecotoxicology* and *Environmental Pollution* and are distilled here for use by decision makers and the public.

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Executive Summary

Mercury Pollution in the Great Lakes Region

Mercury pollution is a local, regional, and global environmental problem that adversely affects ecosystems worldwide. As the world's largest freshwater system, the Great Lakes are a unique and extraordinary natural resource providing drinking water, food, recreation, employment, and transportation to more than 35 million people. Mercury has been released into the air and waterways of the Great Lakes region since the early to mid-1800s from human activities such as fossil fuel combustion, waste incineration, metal smelting, chlorine production, mining, and discharges of mercury in wastewater. The widespread loading of mercury into the Great Lakes environment is responsible for mercury-related fish consumption advisories in the eight Great Lakes states and the province of Ontario. Past and present inputs of mercury pollution have created a substantial recovery challenge for the Great Lakes region.

The Great Lakes region, as defined in this summary report, encompasses the five Great Lakes (Superior, Huron, Michigan, Erie, and Ontario), the eight U.S. states bordering the Great Lakes, and the Canadian province of Ontario. Mercury has long been recognized as an important problem in the Great Lakes region and numerous efforts are underway to curb mercury pollution. Under the Great Lakes Water Quality Agreement, Environment Canada and the U.S. Environmental Protection Agency (U.S. EPA) signed the Great Lakes Binational Toxics Strategy in 1997 calling for virtual elimination of mercury emissions originating from human activities in the Great Lakes region (U.S. EPA 1997). The Great Lakes Regional Collaboration built on this effort and in 2010 produced the Great Lakes Mercury Emission Reduction Strategy with recommendations for decreasing emissions from the largest remaining sources in the basin. Many additional national and global efforts are underway to decrease mercury pollution.

To inform policy efforts and to advance public understanding, the Great Lakes Commission, in 2008, sponsored a scientific synthesis of information on mercury in air, water, fish, and wildlife through its U.S. EPA-funded Great Lakes Air Deposition (GLAD) program. The results of this scientific collaboration have been published in a series of 35 papers in the journals *Ecotoxicology* (Evers et al. 2011a) and *Environmental Pollution* (Wiener et al. 2011a) and are distilled here for use by decision makers and the public.

Major Findings

Five major findings emerge from the results of the binational scientific synthesis of mercury in the Great Lakes region.

I **The Great Lakes region is an internationally significant freshwater resource that is widely contaminated with mercury largely due to atmospheric emissions and deposition.**

II **The scope and intensity of the impact of mercury on fish and wildlife in the Great Lakes region is much greater than previously recognized. Mercury concentrations exceed human and ecological risk thresholds in many areas, particularly in inland waters.**

III **The northern Great Lakes region is particularly sensitive to mercury pollution. The impact of mercury emissions and deposition is exacerbated by watershed and lake characteristics in areas with abundant forests and wetlands that result in higher mercury inputs, transport, methylation, and uptake to elevated concentrations in aquatic food webs.**

IV **Mercury levels in the environment of the Great Lakes region have declined over the last four decades, concurrent with decreased air emissions from regional and U.S. sources. After initial declines, however, concentrations of mercury in some fishes and birds from certain locations have increased in recent years—revealing how trajectories of mercury recovery can be complex.**

V **While the timing and magnitude of the response will vary, further controls on mercury emission sources are expected to lower mercury concentrations in the food web yielding multiple benefits to fish, wildlife, and people in the Great Lakes region. It is anticipated that improvements will be greatest for inland lakes and will be roughly proportional to declines in mercury deposition, which most closely track trends in regional and U.S. air emissions.**

Efforts to advance recovery from mercury pollution in the Great Lakes region in recent years have yielded significant progress, but have yet to address the full scope of the problem. The findings from this binational scientific synthesis indicate that mercury remains a pollutant of major concern in the Great Lakes region; that the scope and intensity of the problem is greater than previously recognized; and that after decades of declining mercury in fish and wildlife, trends are now increasing in some species in particular areas. While the reasons behind these shifting trends require further study, they also underscore the need to expand existing monitoring efforts to better track progress. This is particularly important as new pollution prevention measures are implemented, as global sources increase, and as the region faces changing environmental conditions.

Bald Eagle

1 Why is Mercury Pollution a Problem in the Great Lakes Region?

The Great Lakes region is an internationally significant freshwater resource that is widely contaminated with mercury largely due to atmospheric emissions and deposition.

At a glance

- Controls on large industrial point-source discharges of mercury to surface waters from chlor-alkali plants and pulp and paper mills have led to a partial recovery from mercury pollution in the lower Great Lakes, demonstrating the benefits of mercury controls.
- Emissions of mercury to the air (and subsequent deposition) are now the primary source of mercury pollution to the Great Lakes region. Coal-fired utility boilers (power plants) are the largest source of mercury emissions in the region (Figure 4), followed by metals production and fossil fuel combustion by nonutility sources.
- The amount of mercury that is deposited annually to the landscape varies due to changes in climatic conditions and appears to be highest in areas near large air emission sources in the region. The highest wet deposition levels were measured in Indiana, Ohio, Illinois, eastern and northwestern Pennsylvania, southern Michigan, and southeastern Wisconsin (Figure 5).
- Five states in the region have issued statewide consumption advisories for mercury in fish from all fresh waters, two have issued statewide advisories for mercury in fish from all lakes, and one has issued advisories for specific water bodies. The province of Ontario has also issued advisories for specific water bodies (Figure 6).
- Among 15 fish species consumed by people and wildlife, six species have average mercury concentrations in fillet above 0.30 parts per million (ppm)(the U.S. EPA human health criterion) and above risk threshold for fish-eating wildlife of 0.27 ppm in the inland waters of the Great Lakes region (Figure 7).

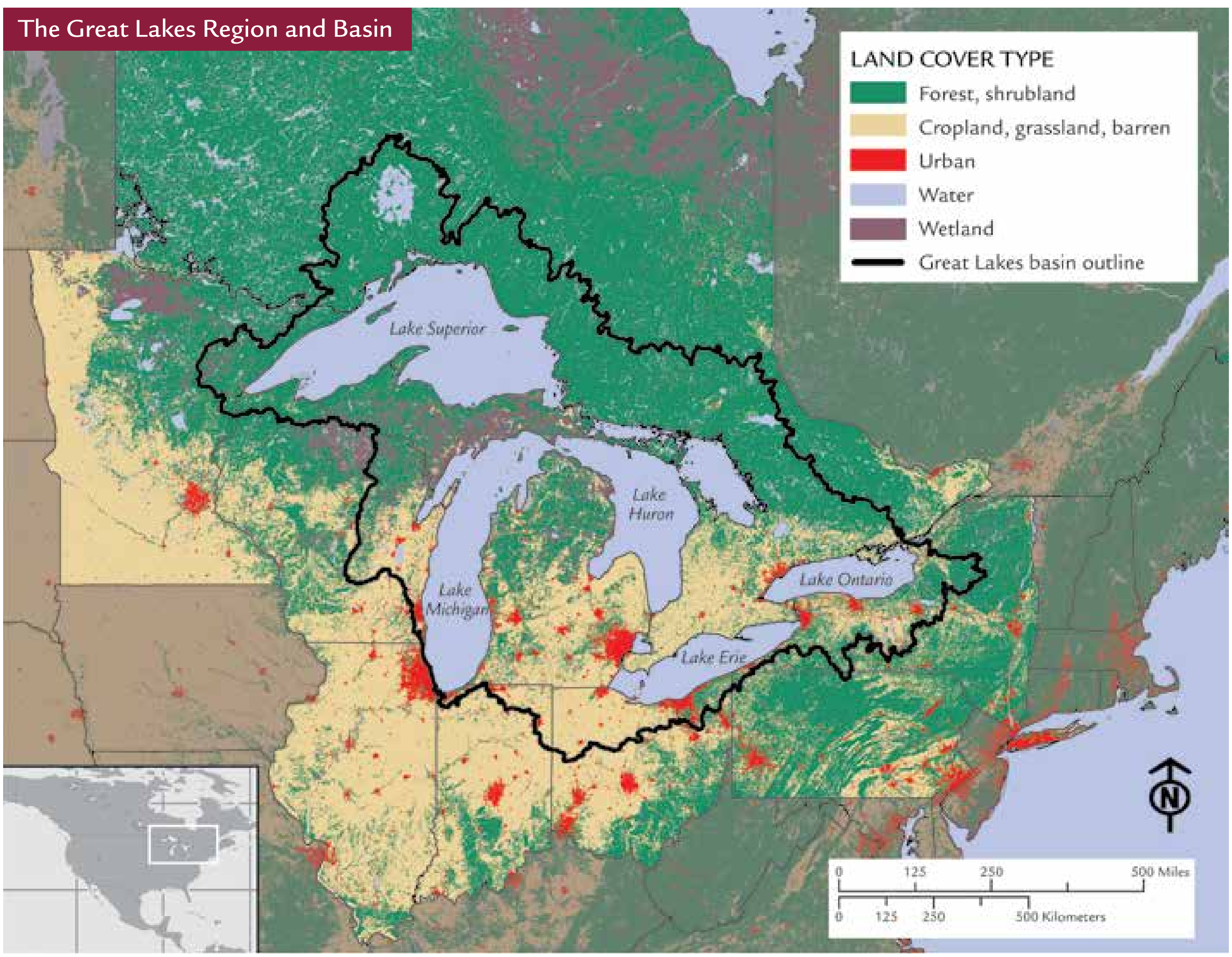
The Great Lakes Region and the Mercury Problem

The Great Lakes are a unique and extraordinary natural resource providing drinking water, food, recreation, employment, and transportation to more than 35 million people, including one-third of the Canadian population and one-tenth of the U.S. population (GLRC 2010). As the largest freshwater system in the world—containing 84 percent of North America's and 21 percent of the Earth's fresh surface waters—the impact of pollution in the *Great Lakes region* (Figure 1) has significant consequences for recreational anglers, commercial fishermen, and subsistence fishers, as well as for the economic status of the region's valuable fisheries and tourism and the health of wildlife that depend on this ecosystem.

The harmful effects of mercury pollution result from the exposure of organisms—including humans—to methylmercury, a highly toxic compound that biomagnifies in aquatic food webs to concentrations that can reach levels several million times higher than those in water (Wiener et al. 2003, Chasar et al. 2009, Rolfhus et al. 2011)(Figure 2). The primary pathway of human exposure to mercury in North America is through the consumption of fish. For more background on the nature of the mercury problem see Box 1.

Figure 1

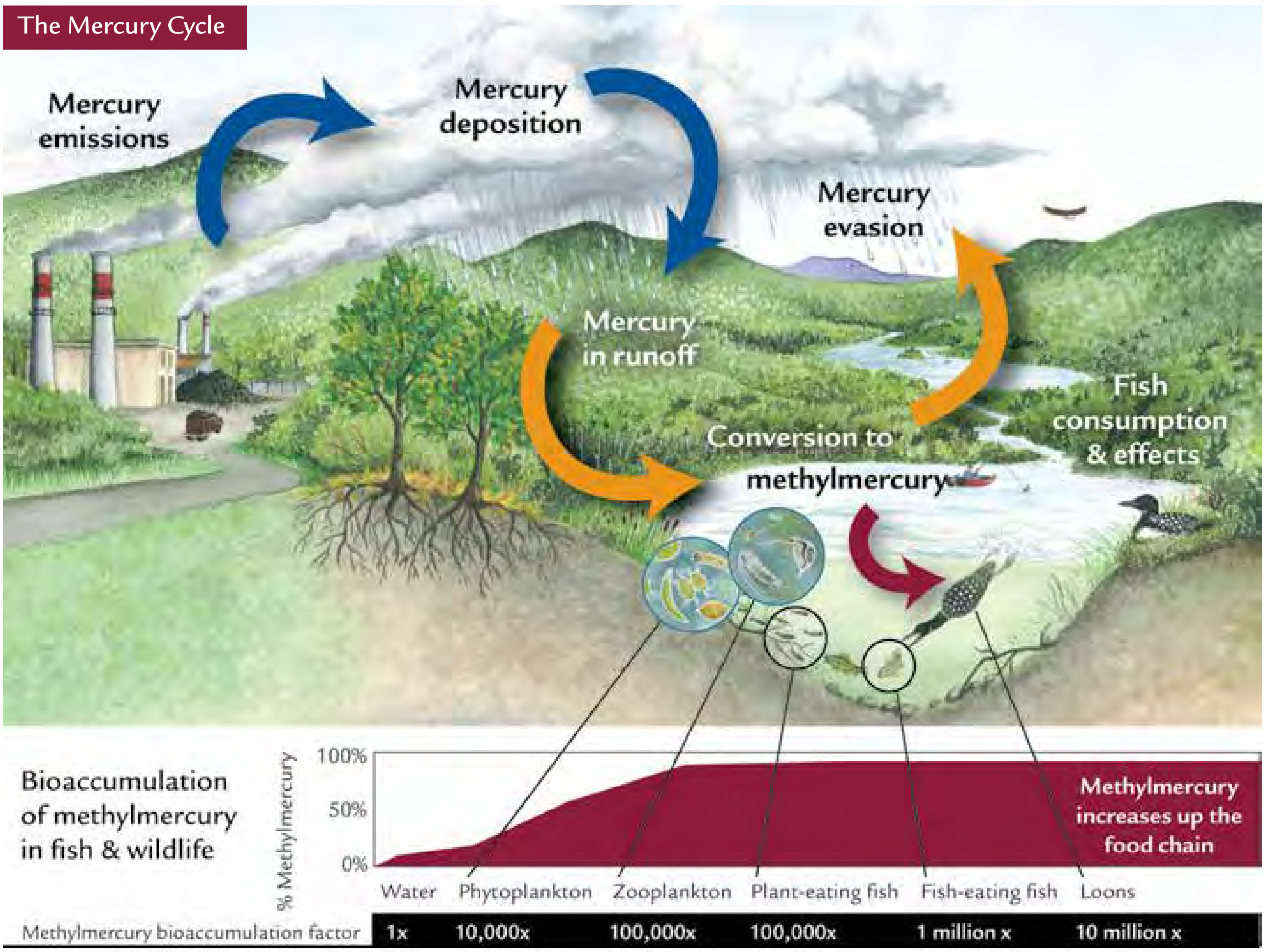
The *Great Lakes region*, as defined in this report, encompasses the five Great Lakes (Superior, Huron, Michigan, Erie, and Ontario), the eight U.S. states bordering the Great Lakes, and the Canadian province of Ontario. The Great Lakes drainage basin is outlined in black. Source: NALCMS 2005.



The Midwest has a higher estimated number of consumers of freshwater fish than any other region in the United States. Sport fishing in the eight Great Lakes states supports more than 190,000 jobs and annually has a total economic impact of more than $20 billion (U.S. dollars; Allen and Southwick 2008). For the millions of people residing in the Great Lakes region who consume sport fish from inland waters (Imm et al. 2005, Allen and Southwick 2008), fish provide an important source of protein that is low in saturated fat and high in omega-3 fatty acids. The contamination of this commercially and nutritionally valuable resource has important socio-economic implications, particularly for subpopulations for whom fish and fishing has cultural significance (Swain et al. 2007)(Box 2).

Figure 2

A simplified mercury cycle showing how mercury enters and cycles through ecosystems, biomagnifies up the food web, and bioaccumulates in fish and wildlife.



food web

Trophic (feeding) relationships between prey and consumers or predators in an ecosystem.

methylation

The conversion of inorganic mercury to the organic form (*methylmercury*). This step greatly increases the bioavailability of mercury, its exposure to wildlife and humans, and ultimately its toxicity. Methylation occurs predominantly under oxygen-poor conditions. Sulfate-reducing bacteria are the primary agents of this process.

bioaccumulation

Accumulation of substances, such as methylmercury, in an organism from various sources (e.g., water, food, air, etc.). Bioaccumulation occurs when an organism absorbs a substance at a rate greater than that at which the substance is excreted.

biomagnification

Increase in concentration of a substance, such as methylmercury, in a food chain. Organisms lower on the food chain contain lower concentrations of methylmercury than the organisms that feed on them (e.g., phytoplankton < zooplankton < plant-eating fish < fish-eating fish < loons/humans).

BOX 1 | Why is Mercury Pollution Harmful?

Mercury is a highly toxic metal that can pose health risks to people and wildlife that consume sufficient quantities of fish. Classified as a persistent bioaccumulative toxin (PBT), mercury is naturally occurring and does not break down over time. Due to the processes of bioaccumulation, even very small quantities of methylmercury in water can result in levels 1 million to 10 million times higher in fish and fish-eating animals such as loons (Driscoll et al. 2007)(Figure 2).

While mercury is a naturally occurring element, it is also released into the environment as a result of human activities. Mercury can be emitted from natural sources such as volcanoes, and released by natural processes, such as wildfires. However, a large amount of the mercury released by natural processes actually originated from human activities. Several studies have determined that, globally, approximately two-thirds of the mercury released to today's environment originates from human activities (Mason et al. 2005, UNEP Chemicals Branch 2008).

The primary form of mercury that poses health risks is methylmercury. The most common pathway of human exposure to methylmercury is through the consumption of contaminated fish. Mercury is typically emitted into the environment as inorganic mercury and through a series of complex processes it can be converted to an organic form known as methylmercury. Methylmercury accumulates more readily in the muscle tissue of fish and their prey (National Research Council 2002). Approximately 95 percent of the mercury in fish is methylmercury.

The greatest concern related to human methylmercury exposure is with respect to sensitive (e.g., women of childbearing age and children) and highly exposed populations (e.g., recreational anglers and their families, subsistence fish consumers, tribal group members that rely on fish, and other consumers of fish with high mercury concentrations). It is estimated that approximately six percent of U.S. women of childbearing age had levels at or above the U.S. EPA reference dose (CDC 2004). Consequently, an estimated 300,000 to 400,000 children born each year are exposed to methylmercury *in utero* at levels high enough to cause neurological health impairment (Mahaffey et al. 2004, Trasande et al. 2005). It is further estimated that women in the United States who rely on fish for subsistence tend to have mercury levels that are on average 3.5 times higher than the U.S. EPA reference dose (U.S. EPA 2011). While most mercury health effect studies focus on changes in intelligence quotient (IQ)(Swain et al. 2007), emerging research suggests that a spectrum of health effects can occur as a result of methylmercury exposure, including cardiovascular disease in men (Salonen et al. 1995, Guallar et al. 2002, Roman et al. 2011).

In response to concerns about methylmercury exposure via fish consumption, in 2004, the U.S. EPA and U.S. Food and Drug Administration issued a joint federal advisory for mercury in fish for the following populations: women who might become pregnant; women who are pregnant; nursing mothers; and young children. U.S. EPA also established a human health criterion for methylmercury in fish (0.30 ppm) to protect the health of frequent consumers of recreationally caught fish. A number of jurisdictions have adopted stricter criteria and use much lower thresholds to issue fish consumption advisories for sensitive populations (Box 2; Table 1).

Mercury's ecological effects were once thought to be limited to aquatic habitats containing fish populations. However, recent studies have shown that wildlife inhabiting terrestrial environments, including mountain and wetland-dwelling songbirds such as the Bicknell's thrush and the rusty blackbird, are also exposed to high concentrations of methylmercury. Scientists have determined that mercury levels in many fish and fish species are high enough not only to pose a threat to human health but also to degrade the health, growth, and reproductive success of the fish themselves (see Section II). While some have suggested that selenium (an essential element) can provide protection against the effects of methylmercury, research in this area is still quite limited and the results are decidedly mixed (Box 3).

Lake Michigan, Sleeping Bear Dunes

Mercury Sources for the Great Lakes Region

Initial regulatory attention in the 1970s focused on large industrial sources of mercury such as chlor-alkali plants and pulp and paper mills. These point sources discharged mercury directly to the Great Lakes and to the rivers and streams that drain into them. Many of these sources have been controlled, leading to the partial recovery from point-source mercury pollution as reflected in lower mercury concentrations in lake sediments in the lower Great Lakes (e.g., Lake Ontario) and in declines in fish mercury since the years of peak pollution.

Atmospheric emissions and deposition are now the largest source of mercury to the Great Lakes region (GLRC 2010). Large stationary sources emit mercury to air as gases and particles. After it is emitted, mercury may travel less than one kilometer to tens of thousands of kilometers before it is deposited back to the Earth's surface, depending on its form. As a result, mercury deposition to the Great Lakes region can originate from sources that are local, regional, national, or global.

U.S. and Global Mercury Emissions

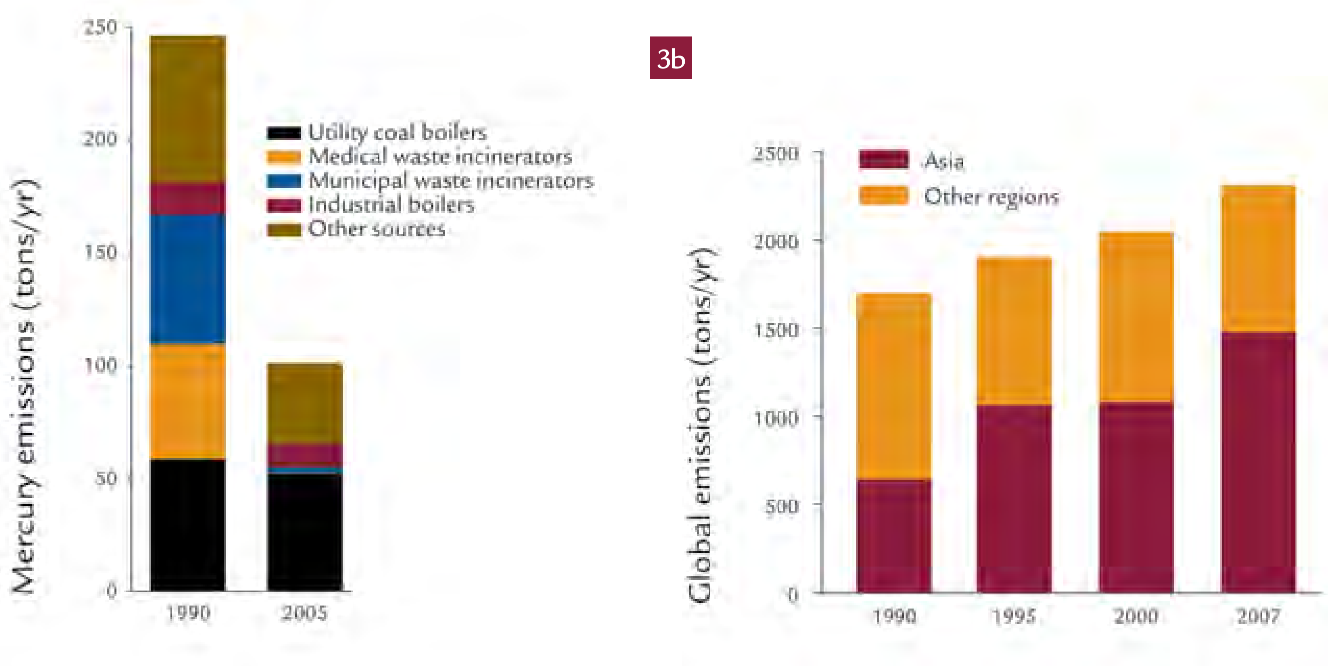
In the United States, approximately 100 tons of mercury are emitted from anthropogenic sources (i.e., from human activities) each year (Schmeltz et al. 2011). Between 1990 and 2005, total U.S. anthropogenic emissions declined by approximately 59 percent, with the largest decreases occurring from hospital and municipal incinerators (95-99 percent decrease) and chlor-alkali facilities (97 percent reduction)(Figure 3a)(Schmeltz et al. 2011). Global emissions inventories suggest that during the same time period, global anthropogenic emissions increased 17 percent (Pirrone et al. 2010). Asia posted the largest increases in mercury emissions due largely to expanding energy production from coal-fired power plants (Figure 3b).

Figure 3

Major sources of U.S. emissions from U.S. EPA inventories (1990 and 2005) (3a). Global emissions and the proportion of global emissions from Asian sources (3b). Note the different scale used for the two figures.

Total U.S. and Global Mercury Emissions from Human Activities

3a



Mercury Emissions in the Great Lakes Region

The Great Lakes basin has served as the industrial engine for North America since the Industrial Revolution. The Great Lakes region accounts for an estimated 56 percent of all raw steel production and 40 percent of electric arc furnace production capacity in the United States (GLRC 2010). Therefore it is not surprising that a large fraction of the total U.S. and Canadian atmospheric mercury emissions originate from the Great Lakes basin (Denkenberger et al. 2011).

In 2005, coal-fired power plants were by far the largest source of anthropogenic mercury emissions to the atmosphere in the Great Lakes states and Ontario, accounting for an estimated 57 percent of total anthropogenic emissions (Figure 4a). They are also the single largest sources in Ontario and most of the Great Lakes states except Minnesota and New York (GLRC 2010). Among the Great Lakes states, Pennsylvania has the highest annual emissions of mercury followed by Illinois, Ohio, and Indiana. However, the mapping of major and minor sources indicates that there is a high density of anthropogenic mercury emission sources across the region (Figure 4b).

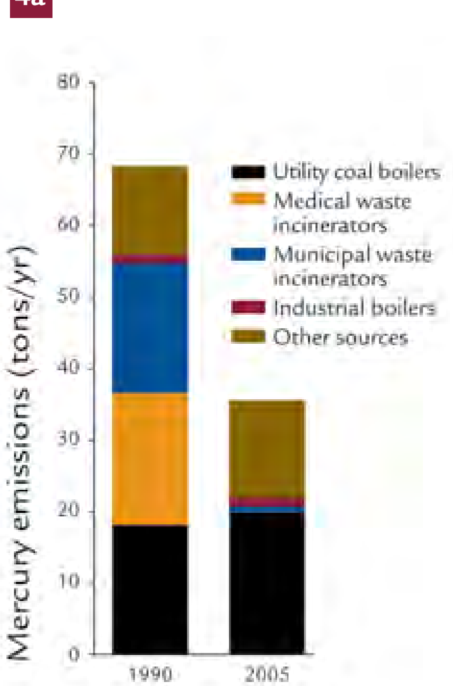
Total mercury emissions to the atmosphere from inventoried anthropogenic sources in the Great Lakes states declined by approximately 50 percent between 1990 and 2005 (NEI 1990, NATA 2005). This decline reflects the leadership the region has demonstrated in controlling mercury emissions through state, regional, binational, and voluntary actions (Cain et al. 2011). Approximately 43 percent of anthropogenic emissions from sources in the Great Lakes basin are reactive gaseous mercury (RGM) or particulate mercury (PHg), the forms that are most likely to be deposited within the region (Denkenberger et al. 2011). This emissions profile suggests that regional and local scale mercury emissions are undoubtedly important to mercury deposition and effects in the Great Lakes basin (Denkenberger et al. 2011).

Figure 4

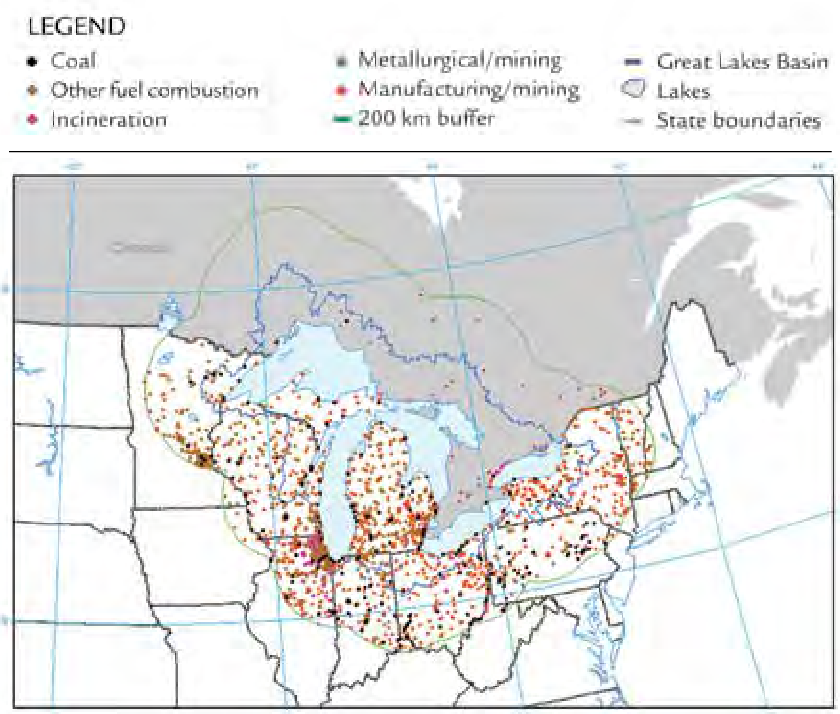
Mercury emissions by source category in the Great Lakes states (4a). Locations of inventoried emission sources (2005) in the Great Lakes basin and an adjacent 200 km buffer area (4b). Note that the U.S. mercury emissions inventory includes more source categories than the Canadian inventory used here.

Mercury Emission Sources in the Great Lakes Region from Human Activities

4a



4b



Mercury Deposition in the Great Lakes Region

After mercury is emitted to the atmosphere, it eventually returns to the Earth's surface in a process termed atmospheric deposition. Deposition is categorized as wet deposition (i.e., mercury deposited in precipitation), dry deposition (i.e., mercury deposited as gas and particles), and litterfall (i.e., mercury that is incorporated into the needles and leaves of plants, which eventually fall to the ground as the foliage is shed). Litterfall is considered a component of dry deposition.

Measurements from monitoring networks in Canada and the United States show that the average annual input of mercury in wet deposition between 2002 and 2008 did not change appreciably over that period. In general, wet deposition of mercury was highest in Indiana, Ohio, Illinois, eastern and northwestern Pennsylvania, southern Michigan, and southeastern Wisconsin (Risch et al. 2011a)(Figure 5), areas with relatively high emissions of mercury from anthropogenic sources (Wiener at al. 2011a). Studies of mercury in litterfall indicate that the dry deposition of mercury to forest landscapes can be similar in magnitude or somewhat greater than mercury in wet deposition, ranging from 25 to 69 percent of total deposition (Risch et al. 2011b).

Mercury Wet Deposition in the Great Lakes Region (2002-2008)

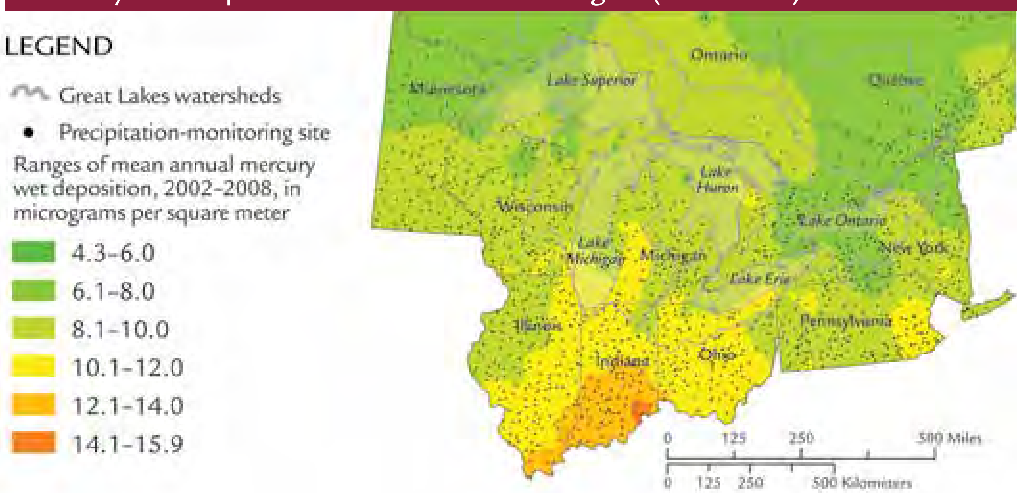
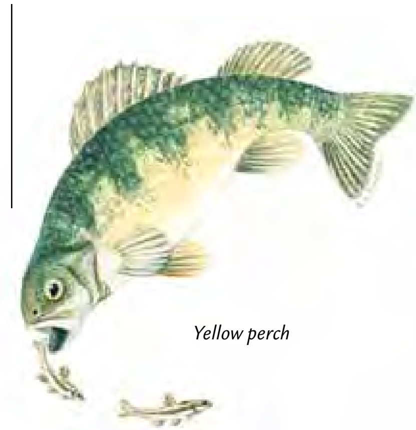


Figure 5

Seven-year mean annual mercury wet deposition in the region based on monitoring data from the National Atmospheric Deposition Program's Mercury Deposition Network.

Fish Advisories and Mercury in the Great Lakes Region

As a result of historic and ongoing mercury pollution, and despite notable improvements, mercury remains an environmental and economic problem across the Great Lakes region. Five states in the region have issued statewide fish consumption advisories due to elevated mercury concentrations in fish from all fresh waters (Illinois, Indiana, Ohio, Pennsylvania, Wisconsin), two states (Michigan and Minnesota) have issued statewide advisories for all lakes, and one state (New York) has issued advisories for specific water bodies (Figure 6).



Yellow perch

Advisories are issued by the individual states, provinces, and tribal agencies and are based on risk assessment policies and guidelines adopted by those jurisdictions. In an effort to create a common approach for the regional fish consumption advisories, the Great Lakes Fish Advisory Workgroup has put together a protocol with recommended mercury-based fish consumption advisory categories

Fish Consumption Advisories for Mercury in the Great Lakes Region

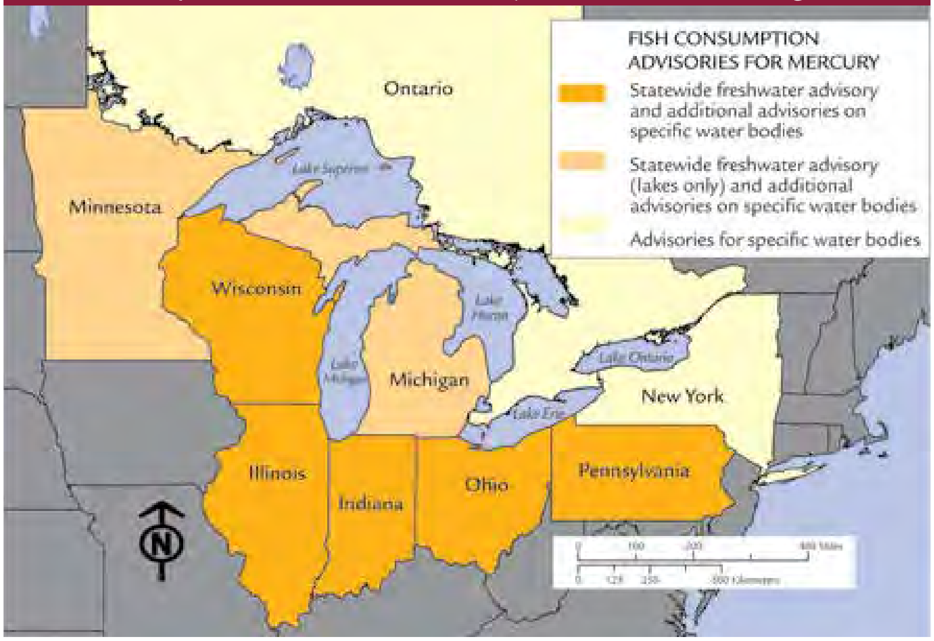


Figure 6

All states and provinces in the Great Lakes region have fish consumption advisories related to mercury contamination. For more detailed information on water body classifications and the basis for each state's advisory, see the U.S. EPA website "Advisories Where You Live," available at: http://water.epa.gov/scitech/swguidance/fishshellfish/fishadvisories/states.cfm, and Ontario province's "Guide to Eating Ontario Sport Fish," available at: http://www.ene.gov.on.ca/environment/en/resources/STD01_078455.html.

and the associated mercury concentrations in fish (Box 2). A summary of average fish mercury concentrations in frequently consumed fish species show that six out of 15 studied species from the inland waters of the Great Lakes region have average mercury concentrations above 0.30 ppm (the U.S. EPA human health criterion). Highest average concentrations are reported for large predatory fish in the inland waters (Figure 7).

Mercury in Selected Fish Species in the Great Lakes Region

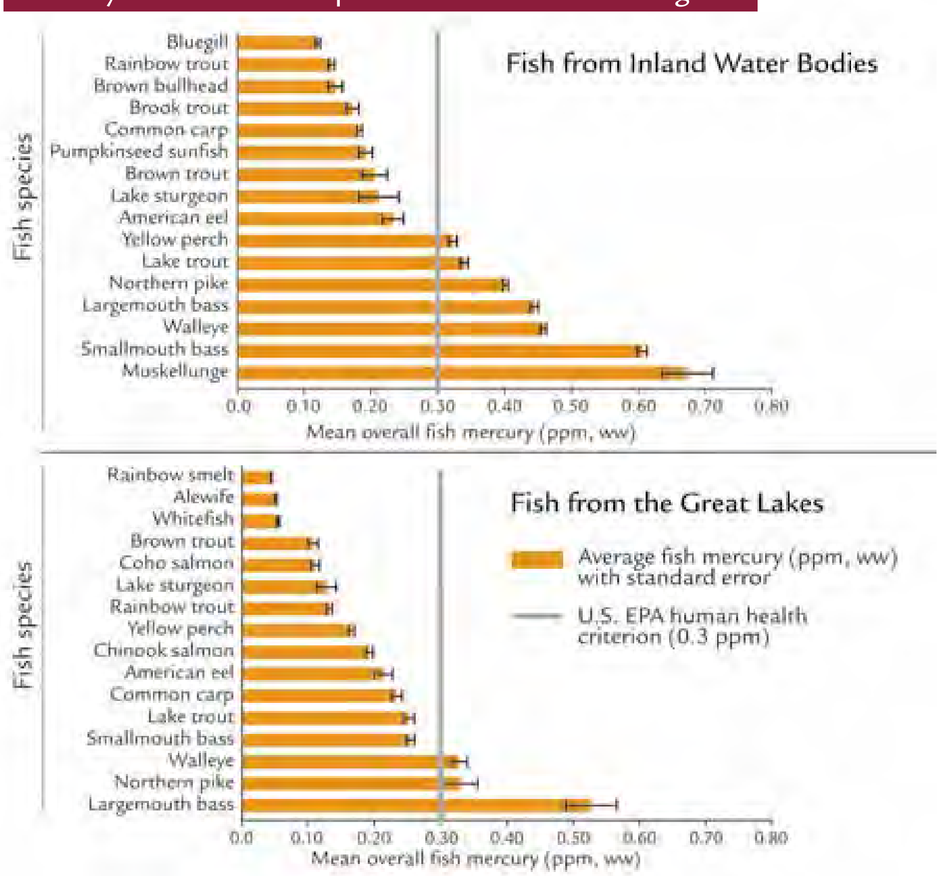


Figure 7

Each histogram displays the mean fillet mercury concentration on a wet weight (ww) basis of fish collected from 2000 to 2008 from inland water bodies and the Great Lakes. Samples were collected by a variety of state and other fish monitoring programs (39,110 fish samples from inland water bodies and 6,572 from the Great Lakes). Species shown are commonly consumed by humans in the region. Standard error bars are presented for each species. The solid line represents 0.30 ppm, the U.S. EPA human health criterion. The estimated effects threshold for fish-eating wildlife is 0.27 ppm in fillet (or 0.16 ppm in whole body fish samples).

BOX 2 | Mercury Degrades Economically Valuable Natural Resources

Brown Trout

The contamination of fish with methylmercury has diminished the economic, health, and cultural benefits of the region's fishery resources. Concentrations of methylmercury in sport fish from surface waters in the region commonly exceed state, provincial, and federal criteria established for the protection of human health. As a consequence, state, provincial, and tribal agencies in the Great Lakes region have issued fish consumption advisories and provided information on the risks and benefits of eating wild-caught fish (Great Lakes Indian Fish and Wildlife Commission 2000, Anderson et al. 2004, Ontario Ministry of the Environment 2011).

The Great Lakes Fish Advisory Workgroup includes members from the eight Great Lakes states who have substantial knowledge and expertise concerning contaminants in fishery resources of the Great Lakes region and the health risks to humans who consume wild fish. In a consensus report, the Great Lakes Fish Advisory Workgroup (2007) recommended a protocol for issuing mercury-based fish consumption advisories for sport-fish consumers. Persons who also eat commercial fish are advised to reduce their consumption of wild-caught fish even further. Based on information obtained from fish consumption surveys, case studies, and exposure assessments (Imm et al. 2005, Knobeloch et al. 2006, 2007) and on the Workgroup's recommended fish consumption guidelines (Table 1), people who frequently consume sport fish from some of the region's inland lakes risk exposure to harmful levels of methylmercury.

Table 1 Fish Mercury Concentrations and Meal Frequency Guidelines

Recommended guidelines and criteria for protection of *sensitive populations* (children and women of childbearing age) who eat wild-caught (noncommercial) fish, in relation to mercury concentrations in fish fillets.

Guideline or criterion	Mercury in fish (ppm wet weight)	Fish consumption guidance
Great Lakes Fish Advisory Workgroup [1]	≤0.05	unrestricted
	>0.05-0.11	2 meals per week
	>0.11-0.22	1 meal per week
	>0.22-0.95	1 meal per month
	>0.95	no consumption
U.S. EPA [2]	0.30	2-3 meals per month [3]
Ontario [4]	<0.26	8 meals per month
	0.26-0.52	4 meals per month
	>0.52	no consumption

1 Recommended guidelines by the Great Lakes Fish Advisory Workgroup (2007).
2 A water quality criterion for methylmercury in fish established to protect the health of persons who consume noncommercial fish (Borum et al. 2001).
3 Based on a consumption rate of 17.5 grams of fish per day (equivalent to 0.53 kilogram or 1.2 pounds of fish per month).
4 Guidelines by Ontario Ministry of the Environment (Bhavsar et al. 2011).

Wood Thrush

II What Risks Does Mercury Pollution Pose in the Great Lakes Region?

The scope and intensity of the impact of mercury on fish and wildlife in the Great Lakes region is much greater than previously recognized. Mercury concentrations exceed human and ecological risk thresholds in many areas, particularly in inland waters.

At a glance

- Mercury pollution is ubiquitous across the Great Lakes region. Elevated mercury levels have been detected in many animal groups (e.g., birds, fish, mammals), at all levels of the food web in lakes (e.g., plankton, fish, loons), and across many different habitat types (e.g., lakes, wetlands, streams, forests) throughout the region.
- With expanding research, the number of documented wildlife species with mercury levels of concern has increased substantially. For example, over the past two decades the number of bird species cited in the scientific literature as adversely affected by mercury has increased by a factor of six (Figure 8).
- During recent decades, research on the toxicological impacts of mercury pollution has demonstrated that effects on fish (Table 2) and wildlife occur at lower mercury concentrations than previously reported.
- A screening analysis for mercury in the Great Lakes region illustrates that risks to fish, wildlife, and people who consume fish in the region can be substantial, particularly in inland waters. Specifically:
  - ✓ Average mercury concentrations in four top predator fish exceeded the adverse effects threshold for fish of 0.20 ppm in 8 percent (largemouth bass) to 53 percent (walleye; see Figure 10) of the study grid cells.
  - ✓ Twenty-four percent of the study grid cells had average estimated blood mercury levels in common loons equal to or exceeding 2.0 ppm, a threshold associated with at least a 22 percent decrease in the number of fledged young (Figure 11).
  - ✓ Average mercury concentrations in six commonly eaten fish species were above the U.S. EPA human health criterion (0.30 ppm) in 61 percent of the study grid cells. All study grid cells exceeded the Great Lakes Fish Advisory Workgroup recommended threshold for unrestricted consumption of fish by sensitive populations (0.05 ppm)(Figure 12).

Mercury Effects Expand With Research

Decades of study compiled for the Great Lakes region reveal that mercury pollution has caused widespread contamination of sediment, water, fish, and wildlife across the region and in many different habitat types (see list of papers, page 38). A summary of the literature shows that with increasing studies, elevated mercury has been documented in a growing number of bird species across the Great Lakes region and northeastern United States (Figure 8). Moreover, the habitats in which high mercury levels in birds have been documented have expanded and now include floodplain forests, bogs, and marshes. The results indicate that mercury can be transferred through food webs not only in animals that eat fish (termed piscivores) but also in animals that eat insects and other invertebrates (known as invertivores).

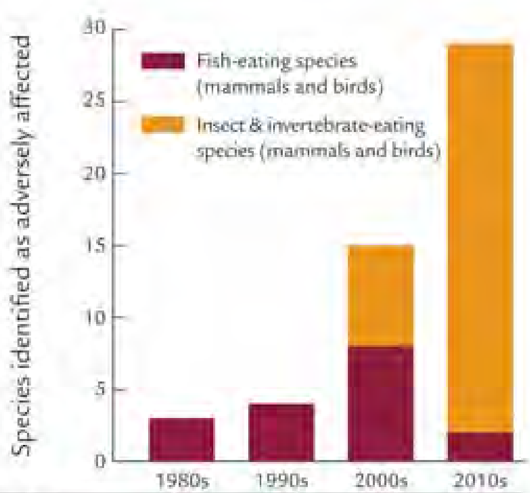
Increased research has also led to discoveries of progressively lower effect levels related to methylmercury exposure. For example, in 1985, the human health *reference dose* (RfD) for methylmercury was calculated by U.S. EPA at 0.30 micrograms per kilogram per day (µg/kg-day), but based on results of additional studies, the U.S. EPA lowered the RfD to 0.1 µg/kg-day (U.S. EPA 2001). This action was affirmed by the National Academies of Sciences. The lower RfD reflects the intent to protect prenatally exposed children against neurological effects (i.e., diminishing the ability of a child to learn and process information) associated with exposure to elevated methylmercury levels while in the womb.

Mercury effects in fish and wildlife have also been documented at progressively lower concentrations since the first reports in the mid-1970s. For example, the research literature shows that studies focused on mercury levels that caused death in fish reported effect levels of 5 to 10 ppm in whole fish (Wiener and Spry 1996). Recent research shows that much lower levels are toxicologically significant. For example, sublethal effects on reproduction, changes in biochemical processes, and damage to cells and tissues can occur at whole-fish concentrations as low as 0.20 to 0.30 ppm (Beckvar et al. 2005, Dillon et al. 2010, Sandheinrich and Wiener 2011)(Table 2).

reference dose (RfD)

The estimated daily oral exposure of a substance to an individual that is likely to be without an appreciable risk of deleterious effects during a lifetime. In the United States, the U.S. EPA has set the reference dose for methylmercury at 0.1 µg/kg-day (5.8 micrograms per liter (µg/L) in blood). The European Union and Health Canada use slightly higher values of 0.20 µg/kg-day and the World Health Organization uses 0.23 µg/kg-day.

Species Adversely Affected by Mercury



	1980s	1990s	2000s	2010s
Mercury effects studies	3	9	26	34
Habitat types with at-risk species	2	3	8	9

Figure 8

With increased research, the number and types of bird and mammal species identified as experiencing adverse effects from ambient concentrations of mercury in the environment has grown. Fish-eating birds such as loons as well as insect-eating species, such as tree swallows and saltmarsh sparrows, can have high mercury levels (Lane et al. 2011, Cristol et al. 2011).

Common Loon

BOX 3 | Selenium and Mercury—What's the Connection?

Mallard Ducklings

The argument has recently been made that selenium protects fish, wildlife, and people against mercury's harmful effects. The scientific evidence suggests that many important questions remain before such statements could be supported. Early research indicated that the survival of Japanese quail and hatching of their eggs was reduced by dietary intake of selenium (in the form of sodium selenite) and methylmercury by themselves. However, when the two compounds were fed in combination to the birds, the effects were not as severe (El-Begearmi et al. 1977). Additional studies provided more evidence that sodium selenite can protect birds against methylmercury poisoning (Stoewsand et al. 1974, Sell and Horani 1976). The traditional explanation for this interaction has been that mercury and selenium bind to each other, nullifying each other's toxicity. A study by Ralston and Raymond (2010) proposed a different mechanism. They concluded that methylmercury binds to selenium compounds, resulting in a harmful shortage of selenium. It follows from this hypothesis, that if an animal's (or human's) diet is supplemented with selenium, then harm normally attributed to the methylmercury is absent. However, a study of breeding mallards exposed to methylmercury and selenium in combination showed that the combination of the two actually caused more deformities than either compound by itself (Heinz and Hoffman 1998). What seems to have happened is that the methylmercury treatment improved the vigor of the embryos that were exposed to very high selenium (in the form of selenomethionine), allowing them to survive longer and in some cases even hatch, but the co-exposure to methylmercury apparently did not protect the embryos from selenium's adverse effects. In fact, more deformities and a greater variety of deformities were observed when both methylmercury and selenomethionine were injected into eggs (Heinz et al. 2011).

Laboratory and field studies are needed to better understand the complex interactions of mercury and selenium.

Given the confounded findings in lab studies and the common co-occurrence of mercury and selenium in bird tissues and eggs in the wild, both laboratory and field studies are needed to better understand the complex interactions of mercury and selenium.

Adapted from a summary contributed by Gary Heinz, U.S. Geological Survey, Patuxent Wildlife Research Center.

It is important to note that many of the effect levels for mercury have been established with less sensitive species and life stages, which may actually underestimate mercury's ecological impact. For example, common loons are widely used to study mercury, yet they have relatively low sensitivity compared to many of the other bird species that have been tested (Figure 9). In addition, studies are typically conducted on adult animals and a recent study reports that embryos are sensitive to low levels of mercury (Box 4).

Bird Mercury Sensitivity by Species

Figure 9

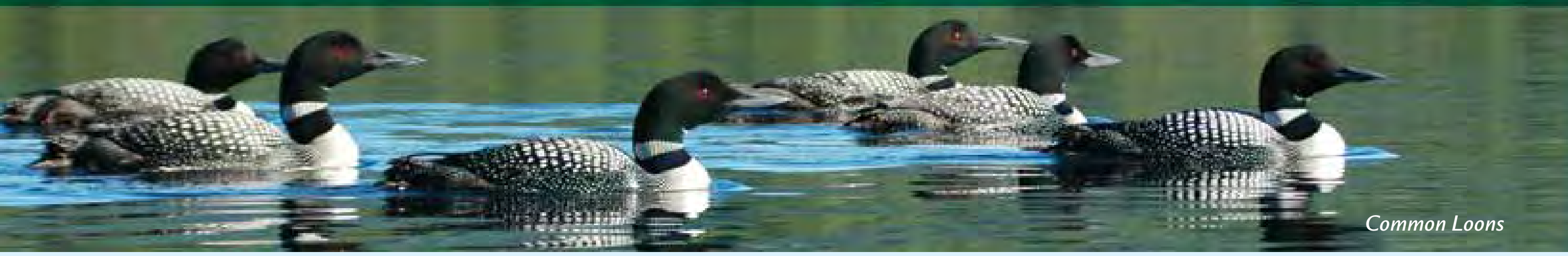
Relative differences in mercury sensitivity in 20 bird species based on the approximate 50 percent lethal concentration (LC50) of MeHg injected into eggs (Heinz et al. 2009, Kenow et al. 2011). Common loons are often used as bioindicators of mercury exposure and effects but have relatively low sensitivity compared to other bird species.

Table 2 Mercury Effects Levels in Fish

Summary of progress in estimating threshold concentrations of mercury (present as methylmercury) in whole body or muscle tissue of fish that cause harmful effects, based on published critical reviews and analyses of scientific literature. Increased understanding has led to the lowering of fish effect levels over time.

Year	Estimated adverse effects threshold (ppm, wet weight)		Documented effect(s)	Reference
	Whole body	Muscle tissue		
1979	—	10.0	Mortality (inferred)	Armstrong 1979
1996	5.0–10.0	5.0–10.0	Sublethal effects and mortality	Wiener and Spry 1996
2005-2010	0.20–0.30	—	Sublethal effects on reproduction, growth, development, behavior	Beckvar et al. 2005, Dillon et al. 2010
2011	0.30	0.50	Reduced reproduction; changes in biochemical processes; damage to cells and tissues	Sandheinrich and Wiener 2011

BOX 4 | Mercury Affects Sensitive Life Stages in Common Loons



Common Loons

While it is well known that high concentrations of mercury in fish carry risks to people who eat large amounts of fish and to sensitive populations, the significance of the impacts on fish and wildlife are often underappreciated. A synthesis of data from recent toxicological studies with freshwater fish and wildlife suggest that adverse effects occur at levels much lower than previously reported, particularly in more vulnerable species and life stages (Scheuhammer et al. 2007, Heinz et al. 2009, Sandheinrich and Wiener 2011, Kenow et al. 2011). This knowledge demonstrates that even as mercury contamination of the physical environment has declined, our estimates of the associated ecological risk caused by exposure to methylmercury have increased.

Common loons offer a useful case study of methylmercury's ecological effects. Studies on wild loons have established clear relationships between methylmercury exposure and behavioral and reproductive effects in adult loons (Nocera and Taylor 1998, Burgess and Meyer 2008, Evers et al. 2008). Developing embryos are more sensitive to mercury, especially at lower exposures, than adult loons. A study on loon eggs found reduced embryo survival for eggs at 1.3 ppm, and a 50 percent reduction in embryo survival for eggs with mercury above 1.78 ppm (wet weight). In birds, the developing embryo in the egg is the most mercury-sensitive life stage, and effects at this stage are most likely to cause population-level impacts.

Recent studies suggest that adverse effects of mercury exposure on freshwater fish and wildlife occur at levels much lower than previously reported.

Mercury in Fish in the Great Lakes Region

A risk screening for the Great Lakes region demonstrated that fish in many waters of the region contain methylmercury at levels associated with adverse effects on the health and reproduction of fish, as well as on the wildlife and people who consume them. The risk is particularly high for top-predator fish from inland waters (Figure 7).

Mercury levels are generally higher in fish from inland waters than in the Great Lakes. For example, the median concentration of mercury in yellow perch fillets from the Great Lakes was 0.090 ppm wet weight (range 0.010-0.90 ppm) compared to 0.14 ppm (range 0.010-2.60 ppm) in perch of similar length from inland lakes and reservoirs (Wiener et al. 2011b). For walleye, mercury concentrations were about 55 percent lower in Great Lakes than in fish from inland lakes. For largemouth bass, levels were 25 percent lower in the Great Lakes compared to inland lakes (Monson et al. 2011). Higher concentrations of mercury are also found in large, long-lived fish-eating species (such as walleye, largemouth bass, and lake trout) compared to lower *trophic level* species (such as yellow perch).

trophic (feeding) level

A position organisms sharing similar prey and predators occupy in a food web.

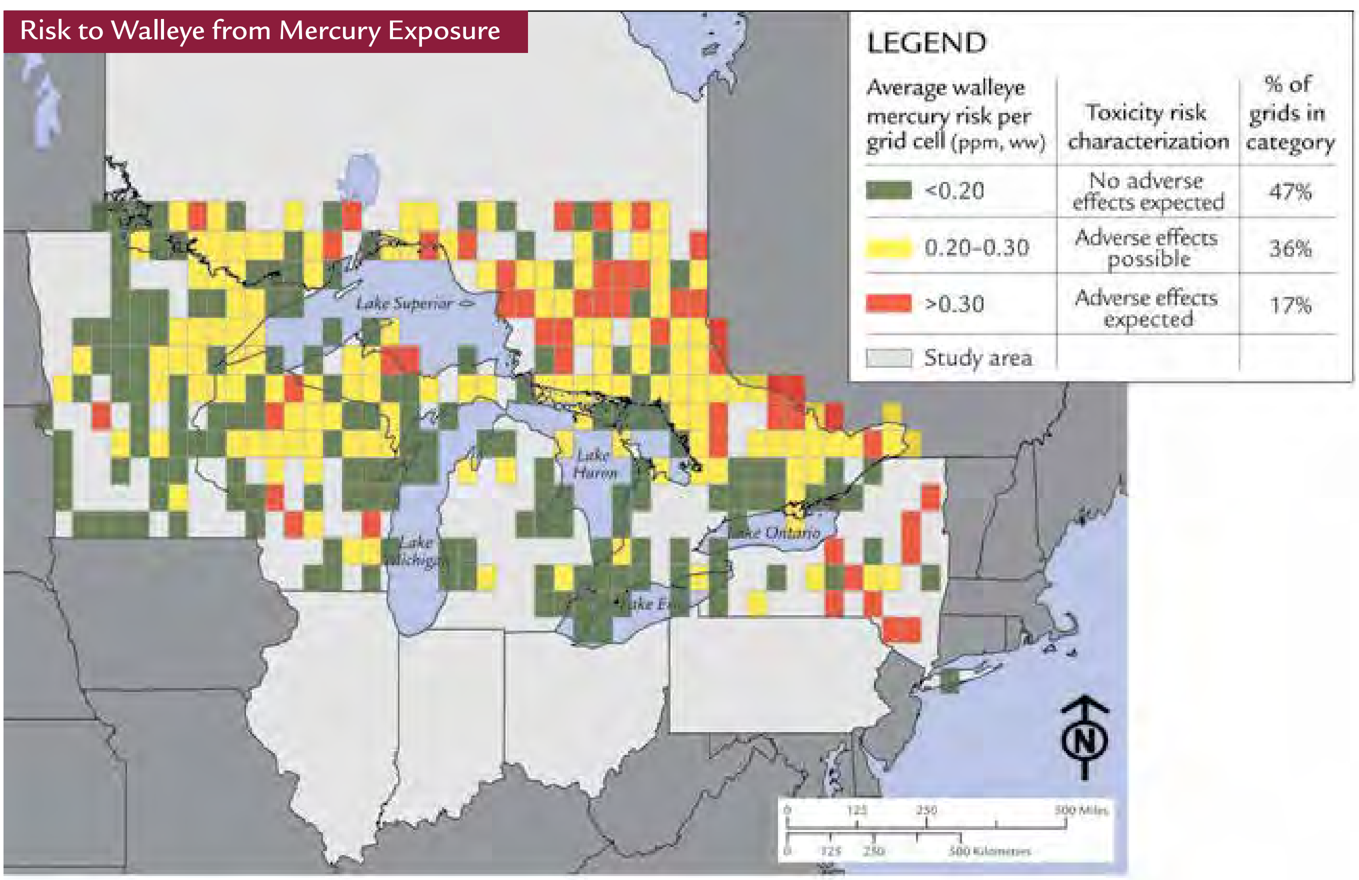
Risk to Fish from Mercury Exposure

Elevated methylmercury in fish can have detrimental impacts on their health and reproduction. The potential risk to fish from elevated methylmercury concentrations has been assessed for the Great Lakes region by comparing average mercury concentration in four top-predator fishes (walleye, northern pike, smallmouth bass, and largemouth bass) for the period 1990 to 2008 with several fish effect thresholds (Figure 10). The screening used average mercury concentrations in the whole bodies of female fish of reproductive size and included approximately 43,000 measurements in fish from 2,000 locations (Sandheinrich et al. 2011). The results of the risk screening show that, depending on the species, the average fish mercury levels in 8 percent (largemouth bass) to 54 percent (walleye) of the study grid cells were above the lowest effects threshold of 0.20 ppm (Figure 10)(adapted from Sandheinrich et al. 2011). Fish at two percent to 17 percent of the study grid cells had average fish mercury concentrations exceeding 0.30 ppm, a concentration high enough to cause injury and reduce reproduction of fish (Sandheinrich et al. 2011). Walleye, a commercially and recreationally important game fish that is distributed throughout the region, provides a good example of the risk that mercury poses to top-predator fish (Figure 10).

The results from the analysis of mercury in predatory fish described above are corroborated by findings from a binational synthesis of data for 6,400 individual yellow perch caught in inland lakes, reservoirs, and the Great Lakes (Wiener et al. 2011b). While lower trophic level fish such as yellow perch tend to have lower mercury concentrations than predatory fish (Figure 7), whole yellow perch from 6.5 percent of waters examined had average levels of total mercury associated with adverse effects on fish.

Figure 10

The risk screening for mercury shows the average risk from mercury exposure in walleye in the Great Lakes region (adapted from Sandheinrich et al. 2011). Reproductive-age walleye mercury concentrations were averaged per 30x30 minute grid cell to estimate possible population-level risk across the region. 53 percent of the study grid cells have average reproductive-age walleye concentrations of at least 0.20 ppm.



Risk to Wildlife from Mercury Exposure

Common loons have been widely used as an indicator species for assessing mercury exposure and risk in birds (Evers et al. 2008, 2011b). The common loon is listed as a threatened species by the state of Michigan and as a species of special concern in Wisconsin and New York. They are long-lived predators that feed almost exclusively on fish and crayfish. As such, these high trophic-level birds typically have relatively elevated mercury concentrations in their bodies but are considered less sensitive to mercury than many other species (Figure 9).

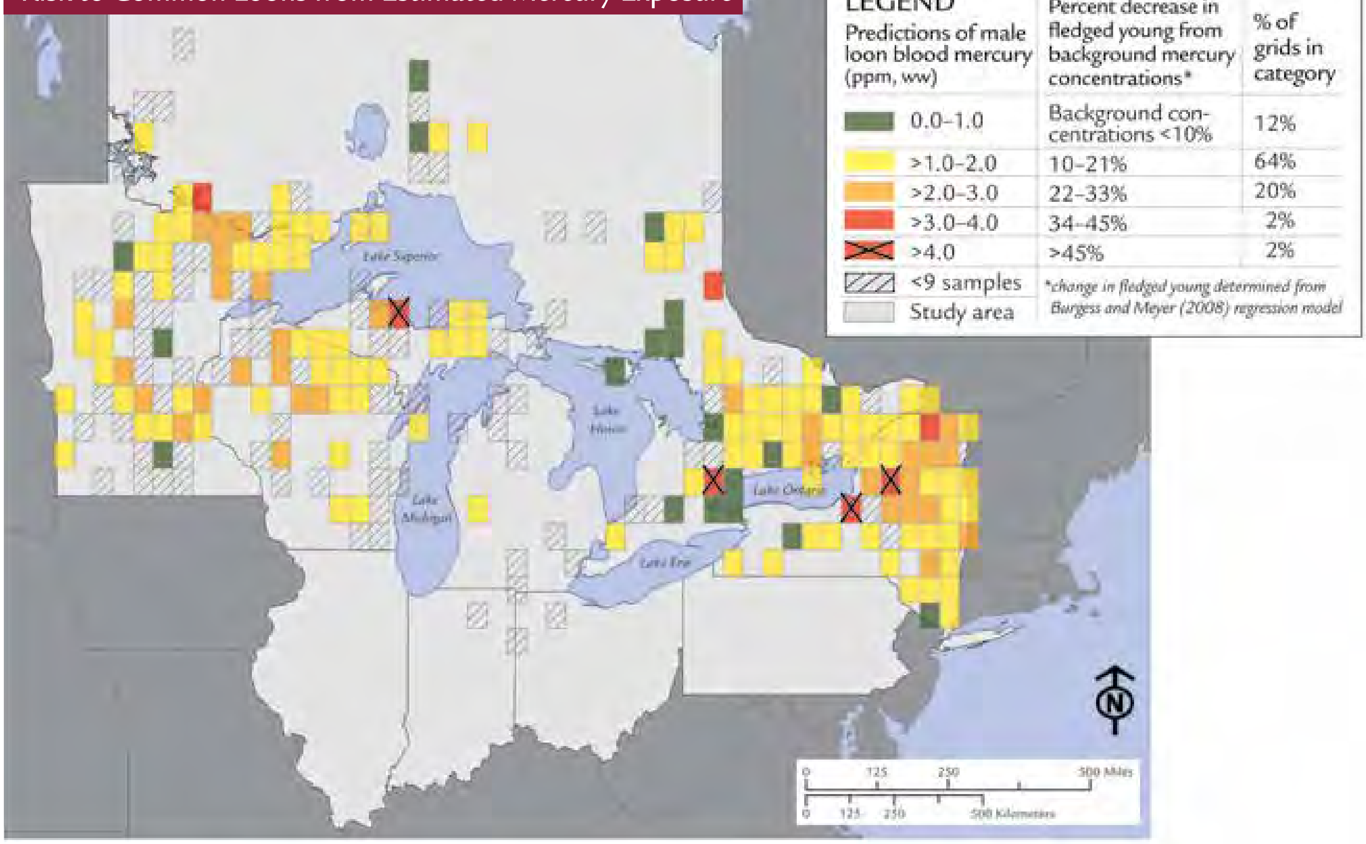
An analysis of measured and estimated mercury concentrations in the blood of male common loons in the Great Lakes region indicates that blood mercury levels surpass thresholds for important ecological effects and that in some locations concentrations are high enough to impair productivity (i.e., the number of fledgling chicks produced per loon pair) (Evers et al. 2011b)(Figure 11). Twenty-four percent of the study grid cells had average estimated total mercury levels in common loons that equaled or exceeded 2.0 ppm, a level associated with at least a 22 percent decrease in the number of fledged young compared to reference conditions (Figure 11; Burgess and Meyer 2008, Evers et al. 2011b).

There are seven distinct locations in the Great Lakes region where the average of estimated blood mercury levels for common loons exceeded 3.0 ppm (Figure 11). Efforts are underway to determine the driving factors for these elevated mercury levels; such factors may include widespread atmospheric deposition of mercury combined with the effects of water level fluctuations in reservoirs, large point sources of mercury, and landscape sensitivity (Evers et al. 2011b).

Figure 11

Values shown are average total mercury concentrations in male loon blood based on measured or predicted values from 8,101 loon tissues and yellow perch tissues (reported as mercury in male loon units (MLUs) in µg/g, wet weight; data are from state and other fish monitoring programs). Yellow perch are a common prey item for loons, and loon blood mercury levels are related to perch tissue mercury (Evers et al. 2011b). The estimated percent decrease in fledgling loons produced per pair for each blood mercury level was determined from Burgess and Meyer's (2008) regression model of reproductive success in relation to adult loon mercury exposure. Grid cells with fewer than 9 samples were excluded from analysis. Data from Evers et al. 2011b (1990-2009).

Risk to Common Loons from Estimated Mercury Exposure



Wildlife effects in the Great Lakes region have also been assessed based on studies of mink and bald eagles. A study of mink showed that total mercury levels in the liver of mink from these locations varied from low to moderate. Most of the mink sampled (79 percent) had liver mercury levels below 5 ppm (dry weight; approximately 1 ppm wet weight)(Hamilton et al. 2011). None of the mink analyzed in this study had mercury levels approaching lethal concentrations (25 ppm, wet weight; Wren et al. 1987) but many had levels associated with subclinical changes (Basu et al. 2007). The highest total mercury concentrations in the liver of mink in this particular study were found in large marshes along rivers in impounded areas that have fluctuating water levels and are downstream of large historical point sources (Hamilton et al. 2011).

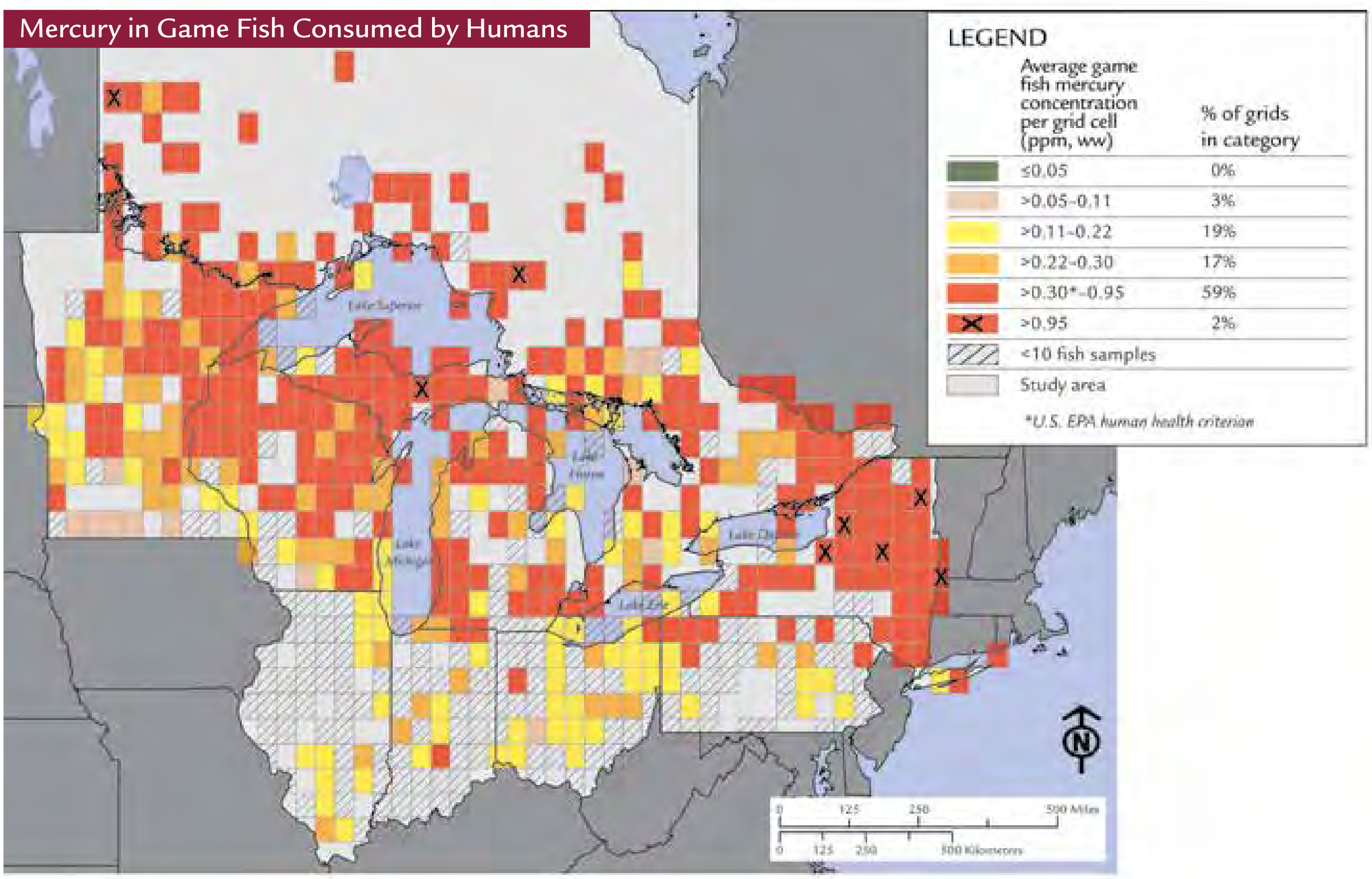
The bald eagle population in the Great Lakes region began to decline in the early 1900s. Since 1977, when a number of regulatory measures went into effect and toxic pollutants such as DDT began to decline, the bald eagle population in the Great Lakes have made a comeback. However, more work is needed to ensure that this population remains healthy and viable because these birds still carry elevated levels of mercury and other contaminants. An analysis of mercury in bald eagles in the Great Lakes region suggests that they have accumulated mercury at levels that can cause subclinical neurological damage, and that 14 to 27 percent of the eagles studied had tissue burdens at or above proposed risk thresholds for birds (based on a toxic effects threshold for mercury in the liver of 16.7 μg/g; Zillioux et al. 1993) (Rutkiewicz et al. 2011).

Mink

Mercury in Game Fish and Potential Human Exposure

A screening of the potential risk to human health posed by mercury in fish in the Great Lakes region was conducted by comparing methylmercury concentrations in six commonly consumed game fish (small- and largemouth bass, lake trout, walleye, northern pike and muskellunge) to the mercury concentrations corresponding to the recommended meal frequency categories developed by the Great Lakes Fish Advisory Workgroup (2007)(Table 1) and to the U.S. EPA human health criterion of 0.30 ppm. The results show that all study grid cells had average mercury concentrations in game fish that equaled or exceeded 0.05 ppm, the level at which the Workgroup recommends that sensitive populations should limit fish intake (Figure 12). Sixty-one percent of the study grid cells had average fish mercury concentrations above 0.30 ppm. Two percent of the study grid cells had average fish mercury concentrations above 0.95 ppm, the level at which the Workgroup recommends that no fish be eaten by sensitive populations. The results of the binational yellow perch analysis show that mean concentration in fillets of perch larger than the minimum size retained by anglers (15 cm) exceeded the U.S. EPA criterion (0.30 ppm) in 6.4 percent of U.S. waters in the Great Lakes region and exceeded the Ontario criterion (0.26 ppm) in 20 percent of Ontario waters (Wiener et al. 2011b). It is important to note that this information is presented for risk screening purposes and is not intended to represent the existing fish consumption recommendations throughout the region.

Figure 12 The mean mercury concentration in 30x30 minute grid cells for six common game fish species (lake trout, largemouth bass, muskellunge, northern pike, smallmouth bass, and walleye). Each study grid cell's color represents the mean mercury concentration of the game fish fillet samples taken from within the grid cell. A total of 25,177 fish samples were included across the region (1990-2008; data are from state and other fish monitoring programs). Grid cells with fewer than ten samples were excluded from analysis. Sixty-one percent of the study grid cells had an average mercury value in game fish fillets of more than 0.30 ppm.



Kawartha Highlands Provincial Park north of Peterborough, Ontario

III Where are Mercury Levels Highest in the Great Lakes Region?

The northern Great Lakes region is particularly sensitive to mercury pollution. The impact of mercury emissions and deposition is exacerbated by watershed and lake characteristics in areas with abundant forests and wetlands that result in higher mercury inputs, transport, methylation, and uptake to elevated concentrations in aquatic food webs.

At a glance

- Walleye and largemouth bass sampled from water bodies in the Great Lakes region show higher mercury concentrations from south to north and from west to east.
- Consistent with these broad geographic patterns of fish mercury concentrations in the Great Lakes region, areas of high mercury concentrations in fish are positively correlated with areas of high forest cover and wetland area (Figure 14).
- The forested areas in the northern reaches of the Great Lakes region receive higher dry deposition of mercury and have other watershed features that produce mercury-sensitive conditions and exacerbate the impacts of mercury emissions and deposition.
- Mercury concentrations in walleye and largemouth bass are 55 and 25 percent lower, respectively, in the Great Lakes than in nearby inland lakes, which may reflect differences in the food web structure, land-water linkages, and methylating potential between the large Great Lakes and smaller inland waters.

Fish Reveal Mercury Patterns

In the Great Lakes region, spatial patterns in mercury concentrations are most evident in the very extensive datasets for fish. In general, fish mercury concentrations tend to be higher in the northern and eastern parts of the region and in the inland lakes. While many factors have likely contributed to these spatial differences, landscape and lake sensitivity, food chain effects, and proximity to current and legacy point sources of mercury have been identified as important factors in this binational synthesis for the Great Lakes region (Monson et al. 2011, Evers et al. 2011b, Zananski et al. 2011, Hamilton et al. 2011).

Mercury-Sensitive Watersheds

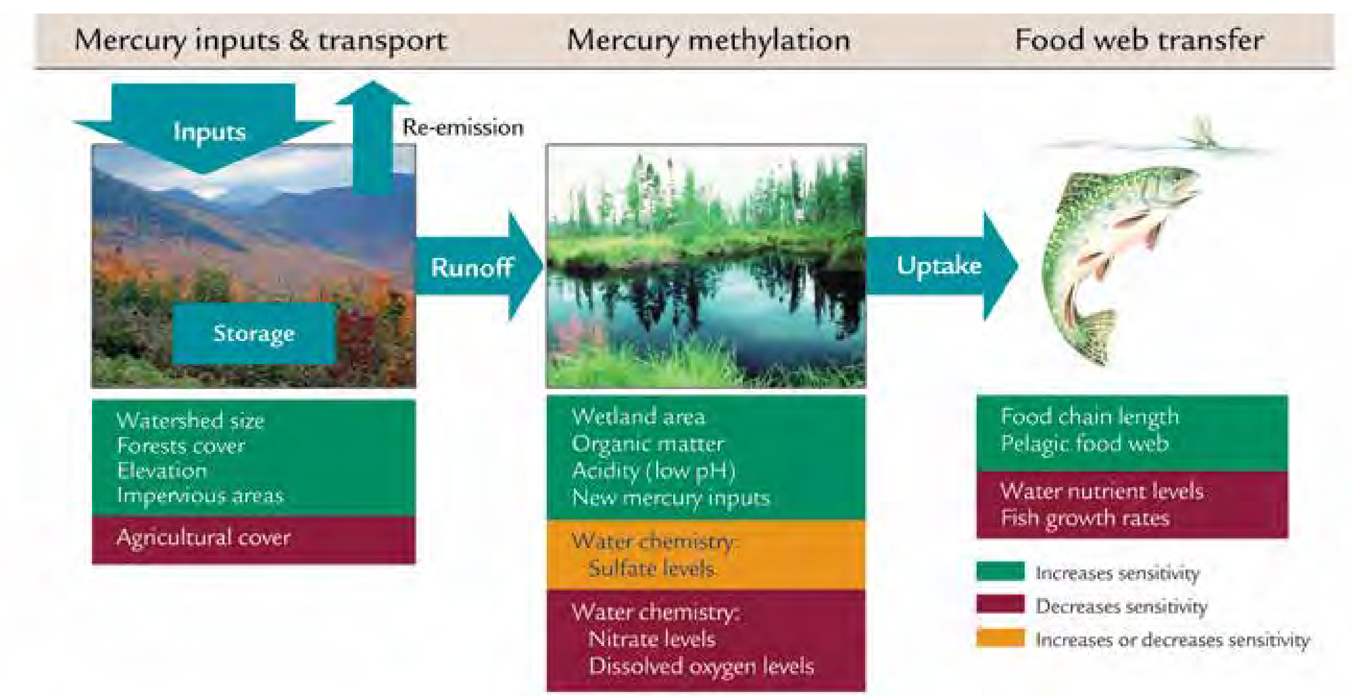
An analysis of mercury concentrations in walleye and largemouth bass from inland lakes, the Great Lakes, impoundments, and rivers show that mean concentrations tend to increase from south to north (Monson et al. 2011)(see also, for example, Figure 10). Mean fish mercury concentrations also tend to be higher in the eastern part of the region compared to the west (Monson et al. 2011).

The northern Great Lakes region and areas to the east are relatively sensitive landscapes where mercury inputs from atmospheric emissions and deposition are more readily converted to methylmercury and bioaccumulated in food webs. The mercury sensitivity of an area is determined by characteristics that influence the inputs, transport, and bioavailability (i.e., methylation and trophic transfer) of mercury in aquatic food webs (Figure 13).

Figure 13

The amount of mercury emitted into the atmosphere affects the levels of mercury in wildlife, but landscape and surface water characteristics also influence how much mercury accumulates in fish and other animals. Landscape characteristics, including the land cover (such as forest or agricultural land), the size of the watershed, the rate of mercury evasion back to the atmosphere, and how water moves through the landscape all affect mercury accumulation, transport and loss. Water characteristics, including the water's acidity (influenced by acid rain and sulfur deposition) play a role in determining how much inorganic mercury becomes methylated and available for uptake by biota. Once methylmercury is formed, it is taken up in the lower food chain and increases with food web transfer. The nutrient levels in the water, the length of the food chain, and other factors affect how much mercury accumulates in wildlife like game fish and common loons.

Watershed Mercury Sensitivity



The northern reaches of the Great Lakes region contain forests which enhance mercury deposition through litterfall. They also have abundant wetlands, which are sites of methylmercury production and sources of methylmercury for surface waters located down gradient (Branfireun et al. 2005, Brigham et al. 2009). Concentrations of methylmercury are also typically elevated in fish and wildlife inhabiting lower-pH waters (Wiener et al. 2003, Burgess and Meyer 2008), which are abundant and widespread in northern parts of the region (Eilers et al. 1988, Clair et al. 1995).

Mercury Cycling in Sensitive Watersheds

In the Great Lakes region, mercury-sensitive areas with abundant forests receive elevated mercury inputs in litterfall originating from atmospheric emissions and dry deposition to the forest canopy. The Great Lakes basin is a net sink for mercury inputs (Denkenberger et al. 2011), with more mercury entering the basin through emissions and deposition than leaving through re-emission to the atmosphere or export via the St. Lawrence River. As a result, mercury deposited to the Great Lakes region has been accumulating in soils, some of which will gradually leach out to surface waters. It is important to note, however, that mercury recently deposited to the landscape tends to be more bioavailable than mercury long buried in soils and sediments. Yet, mercury in soils can be mobilized rapidly by disturbances, such as floods and forest fires, for centuries to come.

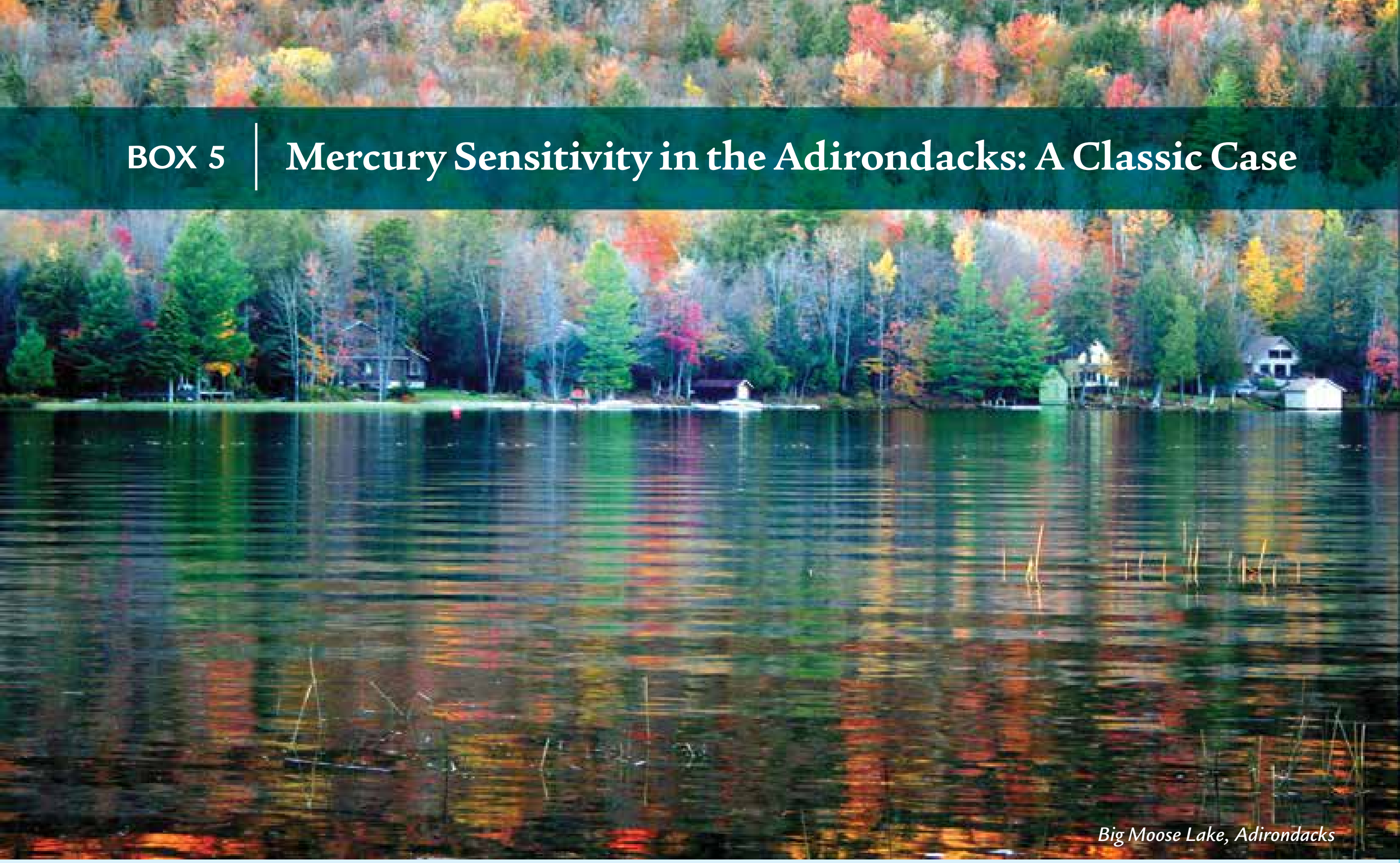
A fraction of the mercury deposited to sensitive landscapes is converted to the bioaccumulative form, methylmercury, in wetlands, sediments, and other favorable environments. This conversion process is amplified under conditions of low pH, high sulfate concentrations, and in dark water systems (i.e., high dissolved organic carbon) that are common in these northern forest landscapes. A classic example of landscape sensitivity and its effects is the Adirondack region of New York (Box 5).

In agricultural lands in the southern part of the Great Lakes region, nutrient inputs from fertilizer and animal waste have increased the amount of algae in surface waters. The increased algal biomass at the base of the food web tends to biodilute the methylmercury, even in individual basins within large lakes (Chen et al. 2011), resulting in lower concentrations in fish and other animals at the top of the food web as compared to levels in more mercury-sensitive watersheds.

An analysis of land cover types (e.g., forest, wetland, and agriculture) and the average total mercury in several top trophic level fishes in inland waters demonstrates these relationships. Fish mercury levels increase with increasing forest cover and wetland area, and decrease with increasing agricultural land (Figure 14). The spatial patterns of fish track the distribution of forest cover in the region (increasing south to north and west to east)(Monson et al. 2011).

In addition to differences in watershed sensitivity throughout the region, there are climatic differences within the region that could affect spatial patterns of fish mercury levels. Many studies report that slower growth rates allow

BOX 5 | Mercury Sensitivity in the Adirondacks: A Classic Case



Big Moose Lake, Adirondacks

The Adirondack region of New York state has been identified as a biological mercury hotspot (Evers et al. 2007). The water chemistry and land cover characteristics of the Adirondack region make it a classic example of a mercury sensitive system. Landscape characteristics and water chemistry can heighten sensitivity to mercury deposition by influencing mercury inputs, transport, methylmercury production, and subsequent transfer up the food web.

Recent studies of mercury in a range of ecosystem compartments (water, sediments, zooplankton, crayfish, fish, and loons) from 44 Adirondack lakes within the biological hotspot confirm that mercury levels in fish and wildlife are related to water chemistry and landscape characteristics, in addition to atmospheric mercury deposition rates (Yu et al. 2011). A substantial number of the lakes studied had average mercury concentrations in fish that exceeded established criteria for human (0.30 ppm, fillet analyses) and wildlife health (0.16 ppm, whole body analyses). Approximately 20 percent of the common loon population exceeded blood mercury levels of 3.0 ppm, a level associated with adverse impacts on reproduction.

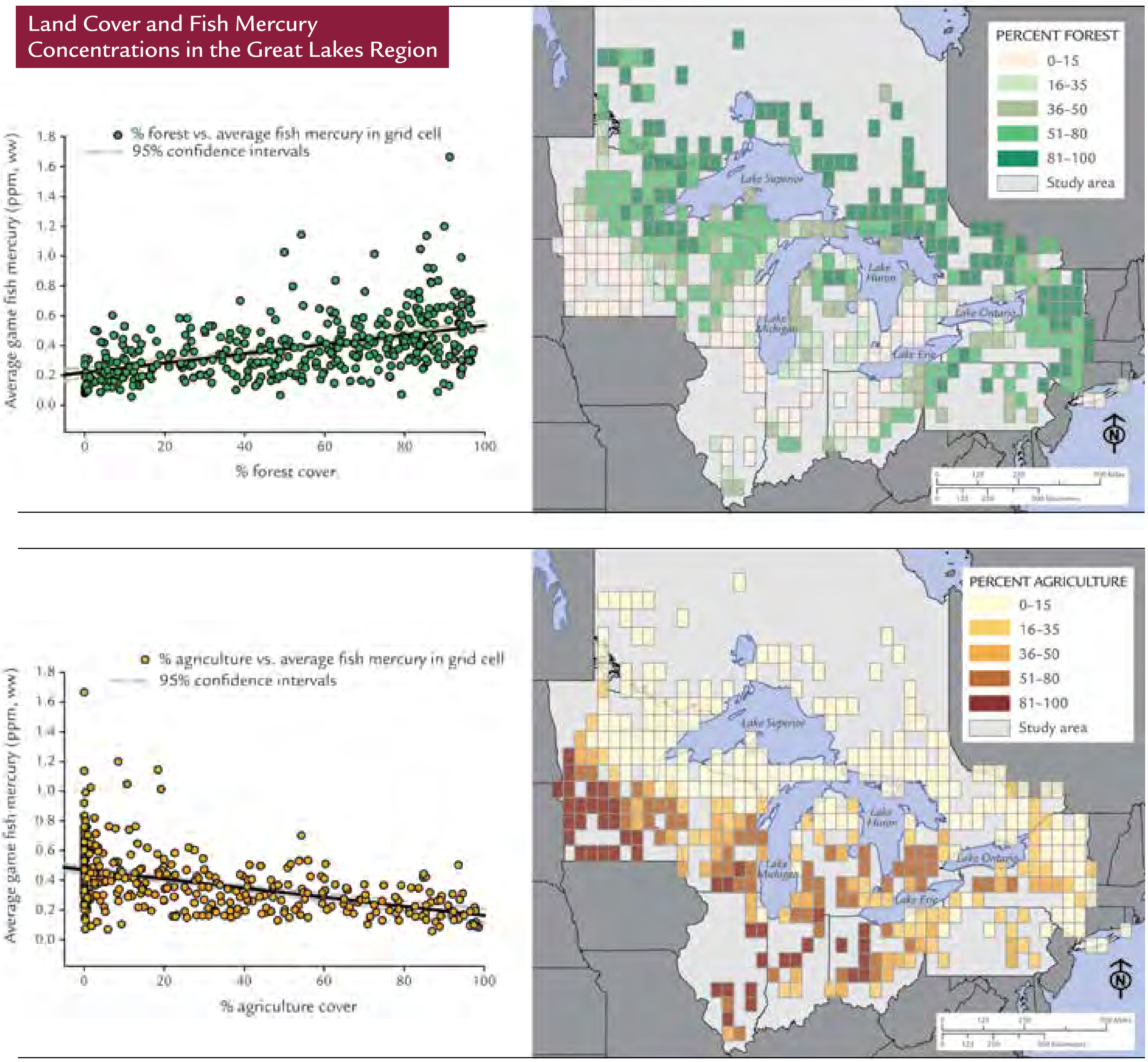
Mercury and methylmercury concentrations in snapping turtles shed further light on the factors that contribute to a biological mercury hotspot in the Adirondack region. Forty-eight snapping turtles from New York State were sampled for mercury to determine what factors account for differences in total mercury levels (Turnquist et al. 2011). Total mercury concentrations ranged from 0.04 to 1.50 ppm and 0.47 to 7.43 ppm in muscle tissue (wet weight) and shell, respectively (Turnquist et al. 2011). Snapping turtles are occasionally consumed locally, or exported to foreign food markets. Sixty-one percent of muscle samples and the mean muscle total mercury concentrations from seven of the 10 sampled lakes exceeded the U.S. EPA's human health criterion of 0.30 ppm (Turnquist et al. 2011). Mercury concentrations in all the study organisms varied spatially. Water chemistry and landscape features explained a significant proportion of this variation. In turtles, mercury in both muscle and shell were positively correlated with maximum watershed elevation. High total mercury concentrations in snapping turtles were also correlated with low acid-neutralizing capacity and high sulfate concentrations in surface water, lake elevation, mercury deposition, and other watershed characteristics. Many of these same correlations were reported for fish and common loons. It is clear that the characteristics of Adirondack lakes and watersheds—including high acidic deposition, substantial forest and wetland cover, and low nutrient inputs—contribute to elevated mercury concentrations in aquatic life.

accumulation of more mercury in fish tissue (e.g., Harris and Bodaly 1998; Simoneau et al. 2005). Fish growth rates tend to be lower in cooler waters and water temperatures tend to decrease with latitude (Jobling 1981). Growth rates are negatively correlated to latitude in largemouth bass and walleye (Helser and Lai 2004, Simoneau et al. 2005). The lower fish mercury concentrations in the south could also reflect the higher fish growth rates in these warmer waters compared to lower growth rates in cooler waters to the north. However, this is an area in need of additional study since rapid growth rates are also sometimes associated with accumulation of higher mercury concentrations in fish tissue over time.

Figure 14

As the percentage of wetlands and forests in the landscape increase, so do the average fish mercury concentrations. Agricultural lands that release large amounts of nutrients into local water bodies can have the effect of decreasing mercury in fish as the nutrients result in a biodilution effect.

Land Cover and Fish Mercury Concentrations in the Great Lakes Region



Herring Gull

IV How is Mercury Contamination Changing Over Time in the Great Lakes Region?

Mercury levels in the environment of the Great Lakes region have declined over the last four decades, concurrent with decreased air emissions from regional and U.S. sources. After initial declines, however, concentrations of mercury in some fishes and birds from certain locations have increased in recent years—revealing how trajectories of mercury recovery can be complex.

At a glance

- Sediment cores from inland lakes within the Great Lakes region indicate that declines in local and regional mercury emissions have decreased mercury delivery to inland lakes across the Great Lakes region by about 20 percent since the mid-1980s (Figure 15).
- Mercury concentrations in walleye, largemouth bass, lake trout and herring gull eggs in the Great Lakes region show downward trends in recent decades, consistent with declines in regional emissions and sediment accumulation in inland lakes (Figure 16).
- In certain areas within the region, mercury concentrations in some fish and wildlife species have been trending upward in the last 10 to 15 years (Figure 17).
- The challenge of interpreting patterns and change in mercury contamination and methylmercury in fish and wildlife underscores the need for comprehensive mercury monitoring at multiregional or national scales and over decadal time scales.

Changing Mercury Inputs: Records From Lake Sediments

Ecosystem recovery from mercury pollution is expected to be a long-term process given that the region is a net sink for mercury and that inputs from industrial point source discharges and regional atmospheric emissions and deposition have been accumulating in the environment for more than a century. Some of this legacy mercury will gradually become bioavailable. Nevertheless, a retrospective look at changes in mercury levels in the environment over time shows that controls on mercury sources have led to important improvements in the Great Lakes region.

The mercury that accumulates in sediments at the bottom of lakes provides an excellent record of changing mercury inputs over time. Data from 91 sediment cores from inland lakes in the region were compiled to assess historical and recent changes in mercury deposition (Drevnick et al. 2011). The inland lakes included in the analysis were limited to lakes in relatively undisturbed watersheds, to allow examination of trends related to atmospheric mercury deposition without the confounding influence of land use. The analysis indicates that during years of peak mercury deposition, the Great Lakes region as a whole was receiving seven times more mercury than during pre-industrial times (~1850)(Figure 15)(Drevnick et al. 2011). Rates of mercury accumulation in cores from Lake Superior and inland lakes reflect a pattern that mirrors changes in atmospheric emissions and deposition. Rates of mercury accumulation in these cores gradually increased to a peak in the mid-1980s and then declined approximately 20 percent from the peak to recent years (Drevnick et al. 2011). The decrease in region-wide lake sediment mercury levels over the past two decades is generally consistent with trends in mercury emissions in the U.S. and the Great Lakes region (Drevnick et al. 2011). However, the decrease in sediment mercury deposition is somewhat less than recent declines in mercury emissions for the U.S. and the Great Lakes basin.

Substantial progress has been made in controlling point-source discharges of mercury to water bodies in industrially polluted zones of the region's Great Lakes. The pattern of mercury accumulation in sediment cores taken from the lower Great Lakes (e.g., Lake Ontario) suggest they were strongly affected by point-source pollution from industrial and wastewater discharges and have experienced partial recovery from such legacy pollution (Drevnik et al. 2011). Mercury from these cores exhibited sharp increases in mercury loading between 1850 and 1950 followed by marked decreases during the past half century in response to effluent controls and decreases in the industrial use of mercury (Drevnick et al. 2011).

Mercury Accumulation in Inland Lake Sediments in the Great Lakes Region

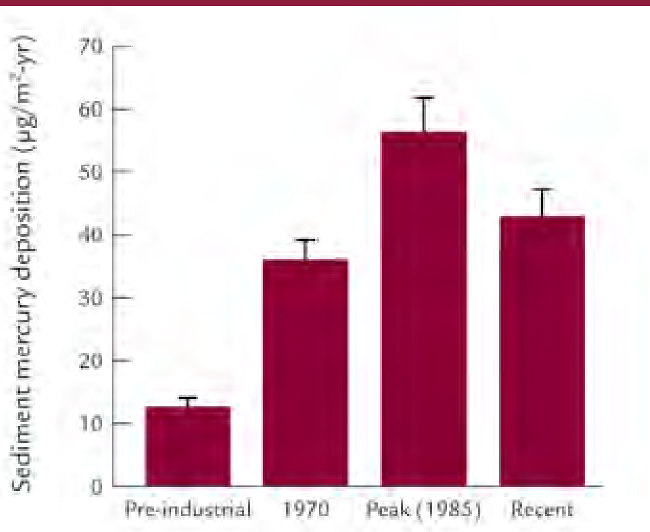


Figure 15

Atmospheric mercury that is deposited to inland lakes and accumulates in bottom sediments forms a record of historical mercury through time. Sediment cores taken from 91 inland lakes around the region indicate that the highest atmospheric mercury deposition occurred around 1985. Recent deposition of mercury is lower, but still three to four times greater than pre-industrial (~1850) levels.

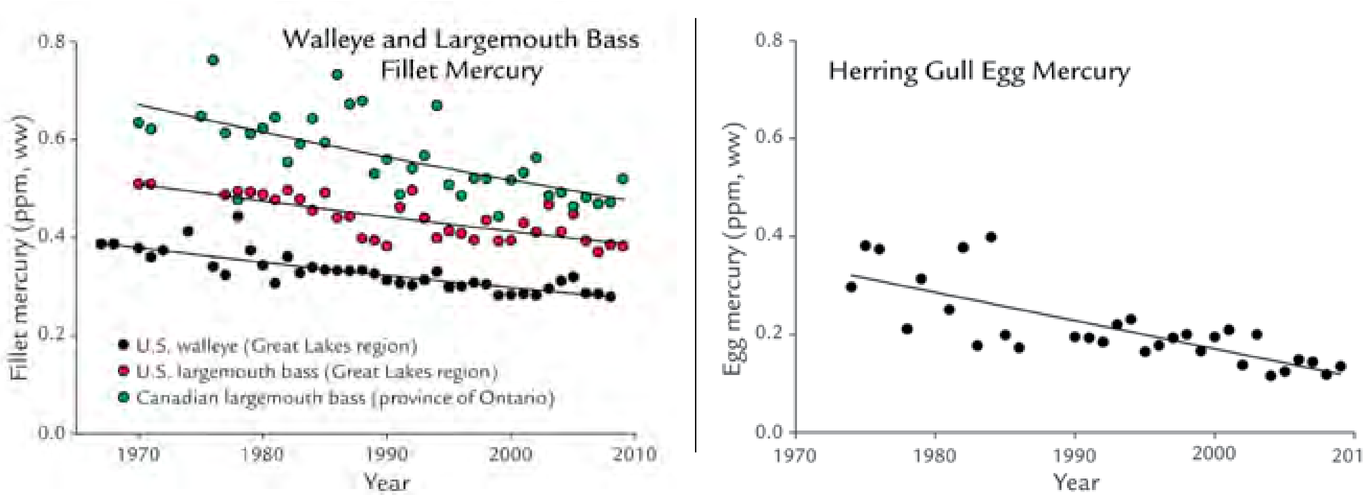
The findings from this comprehensive review of mercury flux from sediments across the region have important policy implications. First, they suggest that local and regional sources of atmospheric mercury emissions are important sources of mercury loading to the Great Lakes region compared to global sources. Atmospheric mercury emissions within the Great Lakes region have decreased in recent decades, whereas global sources have increased since 1985 (Figure 3)(Pirrone et al. 2010). Second, they suggest that recent regional and local controls on atmospheric emissions have been effective in decreasing the amount of mercury delivered to lakes across the region, regardless of watershed size (Drevnick et al. 2011). Given that recent declines in sediment mercury levels have been observed in a large number of lakes sampled by several investigators, these observations suggest a cause and effect relationship between controls on local and regional emissions of mercury to the atmosphere and partial ecosystem recovery from mercury contamination (Drevnick et al. 2011).

Long-Term Declines in Mercury in Fish and Wildlife

In addition to declines in mercury in lake sediments, mercury concentrations in fish and birds of the Great Lakes region have shown an overall decline from 1967 to 2009. Specifically, mercury concentrations in walleye, largemouth bass, and herring gull eggs from different areas within the Great Lakes region decreased during this period (Figure 16). These data are characteristic of the regional trend of decreasing mercury concentrations in fish and wildlife in recent decades. Much of this decrease has been attributed to reductions in regional mercury emissions, though other factors such as shifts in diet may be contributing as well (Weseloh et al. 2011).

Figure 16 Temporal trends in herring gull egg mercury concentrations (averaged by year across multiple sites in the Great Lakes region; Weseloh et al. 2011) and fish fillet mercury concentrations (walleye and largemouth bass, averaged by year across multiple sites in the Great Lakes and inland water bodies in the U.S. Great Lakes states and the province of Ontario; Monson et al. 2011). These data are characteristic of the regional trend of decreasing mercury concentrations in fish and wildlife in recent decades. Much of this decrease has been attributed to reductions in regional mercury emissions, although there may be other contributing factors as well (Weseloh et al. 2011).

Long-Term Mercury Trends in Fish and Wildlife (1967-2009)



Recent Increases in Mercury in Fish and Wildlife

Several studies report that in some areas and in certain species in the Great Lakes region, mercury concentrations may again be on the rise (Figure 17). Since the 1990s, increases in previously declining mercury concentrations have been found in walleye from the province of Ontario and in walleye and northern pike from Minnesota (Monson et al. 2011, Monson 2009), though levels are still lower than peak mercury concentrations in the 1970s. A more recent increase, beginning in 2005, was identified in walleye from Lake Erie (Bhavsar et al. 2010, Zananski et al. 2011). Mercury has also increased in adult loon blood mercury from northern Wisconsin (Meyer et al. 2011) and bald eagles from Voyageur National Park (Pittman et al. 2011). Beyond North America, a similar biphasic pattern has been detected in northern pike in Swedish lakes (Akerblom et al. 2011).

The reasons for these recent apparent increases—and whether they are consistent trends or short-term oscillations within a long-term decline—are not fully understood. Several hypotheses have been presented to explain this shift including factors influenced by changing climate (Monson 2009), lower water levels, and greater exposed shoreline associated with drought (Meyer et al. 2011), changes in food webs associated with introduced exotic species (Monson et al. 2011), and reversal of the biodilution effect through decreases in nutrient loading (Zananski et al. 2011).

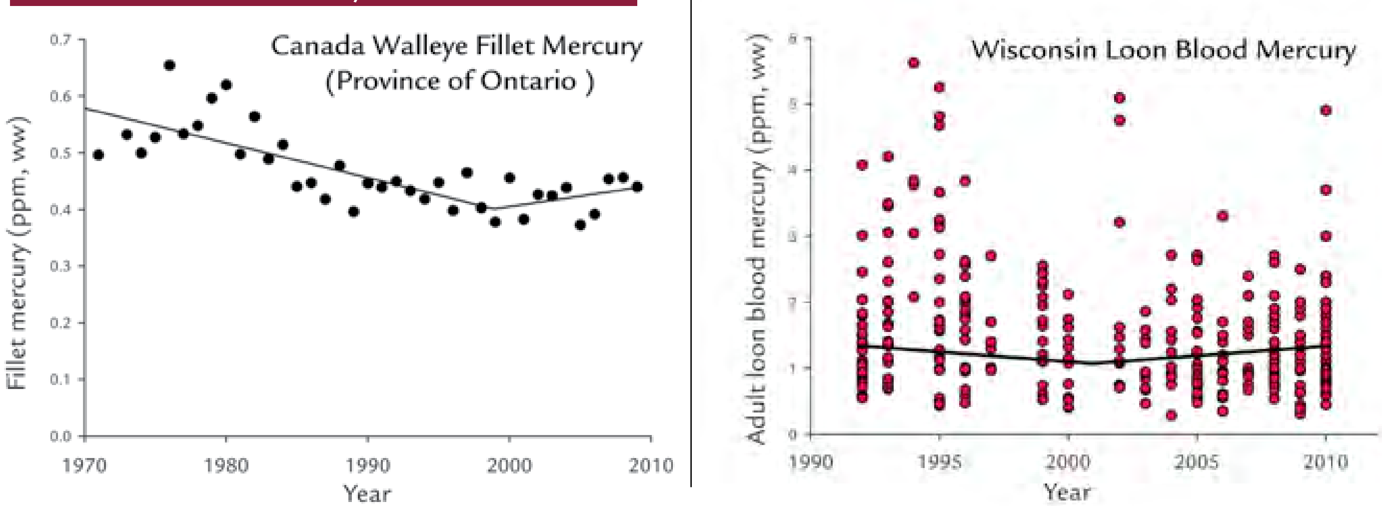
Focused research and monitoring are needed to confirm and interpret these trends, yet it seems plausible that changes in disturbance regimes can increase mercury sensitivity and therefore change the trajectory of methylmercury concentrations in fish and wildlife (Munthe et al. 2007). These interactive effects complicate predictions of how mercury may change over time in the future and further highlight the need for enhanced mercury monitoring.

The complexity of interpreting the spatial patterns and temporal trends in environmental contamination and methylmercury in biota highlight the need for mercury monitoring that is comprehensive (e.g., linking measurements in air, water, sediment, and biota) and large-scale (i.e., regional to national), and that utilizes a probability-based design with repeat sampling to improve prediction and assessment capabilities.

Figure 17

Several studies have found evidence that in some areas and in certain biological species in the Great Lakes region, mercury concentrations may again be on the rise. Monson et al. (2011) found a recent increase in previously declining mercury concentrations in walleye fillets from the province of Ontario. Meyer et al. (2011) saw a 1.8 percent per year increase in common loon adult and chick blood mercury concentrations from northern Wisconsin.

Recent Increases in Mercury in Fish and Wildlife



Lake Erie

V What are the Key Mercury Policy Connections in the Great Lakes Region and Beyond?

While the timing and magnitude of the response will vary, further controls on mercury emission sources are expected to lower mercury concentrations in the food web yielding multiple benefits to fish, wildlife, and people in the Great Lakes region. It is anticipated that improvements will be greatest for inland lakes and will be roughly proportional to declines in mercury deposition, which most closely track trends in regional and U.S. air emissions.

Mercury Policy: An Overview

The binational scientific synthesis of mercury in air, water, sediments, fish, and wildlife has shed important new light on the status and effects of mercury pollution across the Great Lakes basin. The information presented here can inform many of the regional, national, and global policy initiatives currently underway. Such efforts include: (1) recommendations by the Great Lakes Regional Collaboration to decrease mercury loading to the environment; (2) U.S. rules to limit atmospheric emissions of mercury from major sources (e.g., coal-fired power plants and cement plants); (3) international negotiations to establish a global legally binding mercury treaty through the United Nations; and (4) advances in mercury monitoring and research initiatives.

The Great Lakes Mercury Emission Reduction Strategy

Under the Great Lakes Water Quality Agreement, Environment Canada and the U.S. EPA signed the Great Lakes Binational Toxics Strategy in 1997 calling for virtual elimination of mercury emissions originating from human activities in the Great Lakes region (U.S. EPA 1997). The Great Lakes Regional Collaboration (GLRC), established in 2004 by executive order to restore ecosystem health in the Great Lakes, built on this effort and in 2010 produced the Great Lakes Mercury Emission Reduction Strategy (GLRC 2010). The strategy includes more than 34 recommended regulatory and voluntary actions to further control mercury pollution. The following three policy recommendations are particularly pertinent:

- **Lower Regulatory Thresholds for Major Mercury Emission Sources**
  The GLRC strategy recommends that the U.S. EPA lower the current major source category threshold for mercury emission sources. The current threshold for a major source category for hazardous air pollutants (HAPs) is 10 tons for a single HAP and 25 tons for a combination of HAPs. A lower threshold for major sources that emit mercury would extend requirements to implement maximum achievable control technology to smaller sources (GLRC 2010), which are widely present throughout the region (Figure 4b).
- **Require Best Available Control Technology for New and Modified Sources**
  The GLRC strategy also recommends that all states require Best Available Control Technology for new and modified sources if they annually emit 10 pounds of mercury (or fewer, at the state's discretion).
- **Mandate Mercury Emissions Reporting**
  The GLRC strategy further recommends that states implement mandatory reporting requirements of new and existing mercury air emissions sources (with a recommended threshold of five pounds or fewer per year).

U.S. Mercury Regulations

Mercury pollution in the United States is regulated by an array of state and federal regulations (see: http://www.epa.gov/hg/). There have recently been substantial advances in regulatory efforts to decrease mercury emissions from major source categories. Specifically, the U.S. EPA has proposed or finalized Maximum Achievable Control Technology (MACT) standards for mercury from coal-fired power plants, national emissions standards for hazardous air pollutants for gold ore processing and production facilities, final rules to control mercury emissions from Portland cement manufacturing facilities, and proposed new source performance standards and emissions guidelines for new and existing sewage sludge incinerators.

United Nations Global Mercury Treaty

In addition to these national efforts, the United States and Canada are both participating in international negotiations to establish a global legally binding mercury treaty. The need for a global approach to mercury stems from the transboundary nature of mercury pollution. As some regions and nations decrease mercury emissions to the air, a proportionally higher share of mercury deposition originates from sources beyond their boundaries. Moreover, as mercury is phased out of products in the United States, some manufacturing is moved to sites overseas where regulations are less stringent, ultimately resulting in continued releases to the atmosphere and in growing global emissions. In 2009, the Governing Council of the United Nations Environment Programme agreed to work with participating countries to negotiate an international agreement to decrease mercury supply, demand, international trade, atmospheric emissions, and waste handling issues while at the same time increasing awareness, capacity, and technical and financial assistance as appropriate. The intent is to complete negotiations in 2013, at which point the ratification process begins. For more information see: http://www.unep.org/hazardoussubstances/Mercury/tabid/434/Default.aspx.

Mercury Monitoring and Research

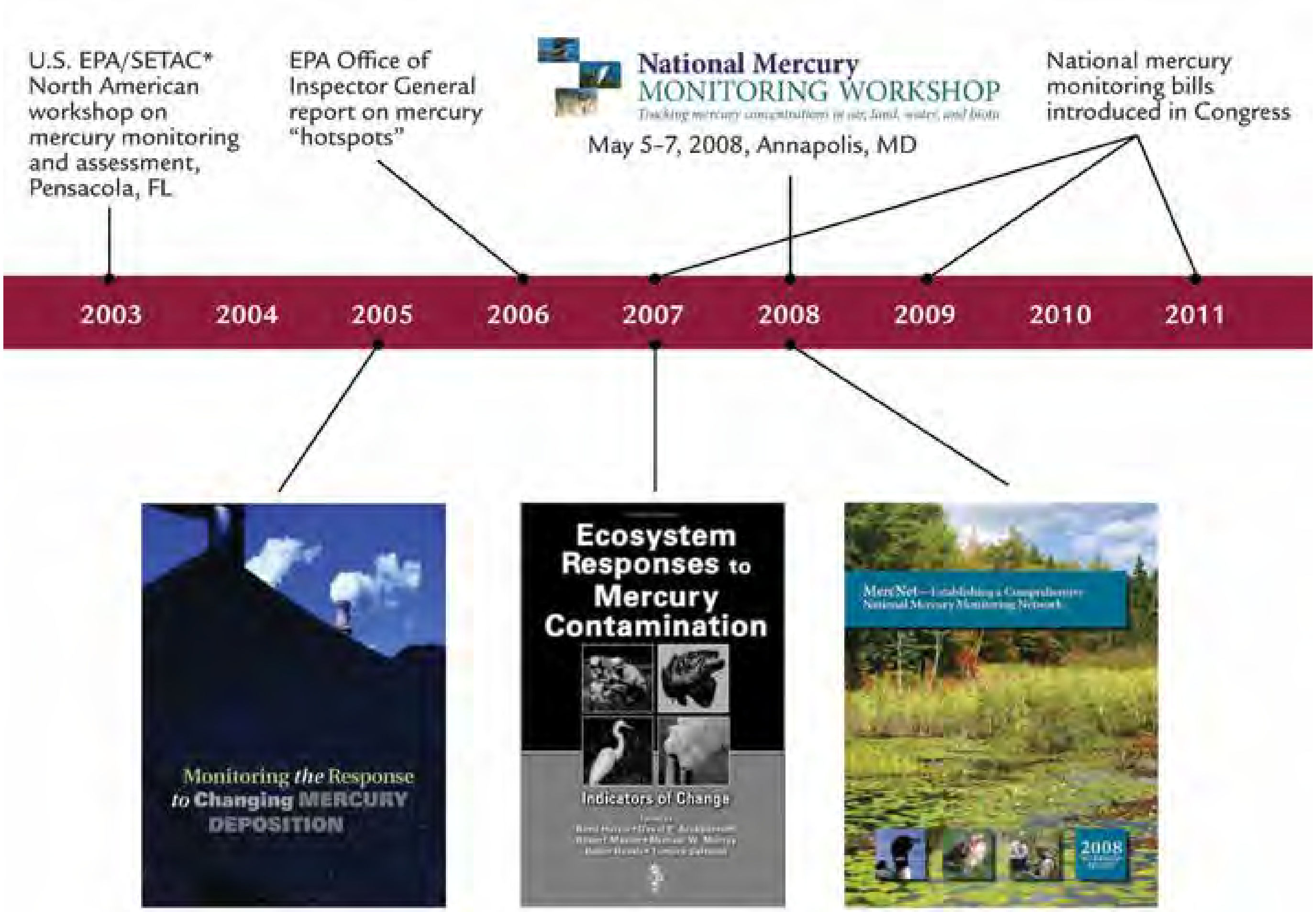
The results of this binational scientific synthesis highlight the need for expanded mercury monitoring and research along with associated status and trends assessments, synthesis work, and modeling efforts that can inform policy decisions and guide future management efforts on a regional, national, and international scale. Canada is implementing a scientific support program to monitor the effectiveness of regulations and provide accountability. This science program is intended to provide the knowledge base necessary to craft effective regulations, and to assess the effectiveness of mercury regulations in achieving environmental and health benefits. The Canadian Mercury Science Program plays an important role in understanding current mercury pollution conditions, monitoring changes as regulations are phased in, and tracking the effectiveness of those regulations (Morrison 2011)(see: http://mercury2011.org/program-ps2).

In the United States, a comprehensive National Mercury Monitoring Network (MercNet) has been proposed to address key questions concerning changes in anthropogenic mercury emissions and deposition, associated linkages to ecosystem effects, and recovery from mercury contamination. More than 50 academic scientists, government scientists, natural resource managers, and representatives from tribal groups have worked together over the past five years to develop a framework for the National Mercury Monitoring Network in the United States (Mason et al. 2005, Schmeltz et al. 2011)(Figure 18). For details of the monitoring network see: http://nadp.isws.illinois.edu/mercnet/MercNetFinalReport.pdf.

MercNet has garnered substantial support at the state and federal levels. The Environmental Council of the States (ECOS) and the Great Lakes Commission have issued resolutions supporting the establishment of the National Mercury Monitoring Network. Federal legislation with bipartisan support has been introduced in multiple sessions of Congress to establish and authorize appropriations to the U.S. EPA to develop a national mercury monitoring program. Expanded mercury monitoring in the U.S. and beyond is crucial for evaluating the effectiveness of public policy and for untangling interactive effects with other large-scale drivers of environmental change.

Figure 18 Beginning in 2003, academics and resource managers, the U.S. Congress, and the U.S. EPA have worked to develop a framework for comprehensive, national-scale monitoring of mercury in the environment in the United States. The National Mercury Monitoring Bill was introduced to Congress for the third time in 2011.

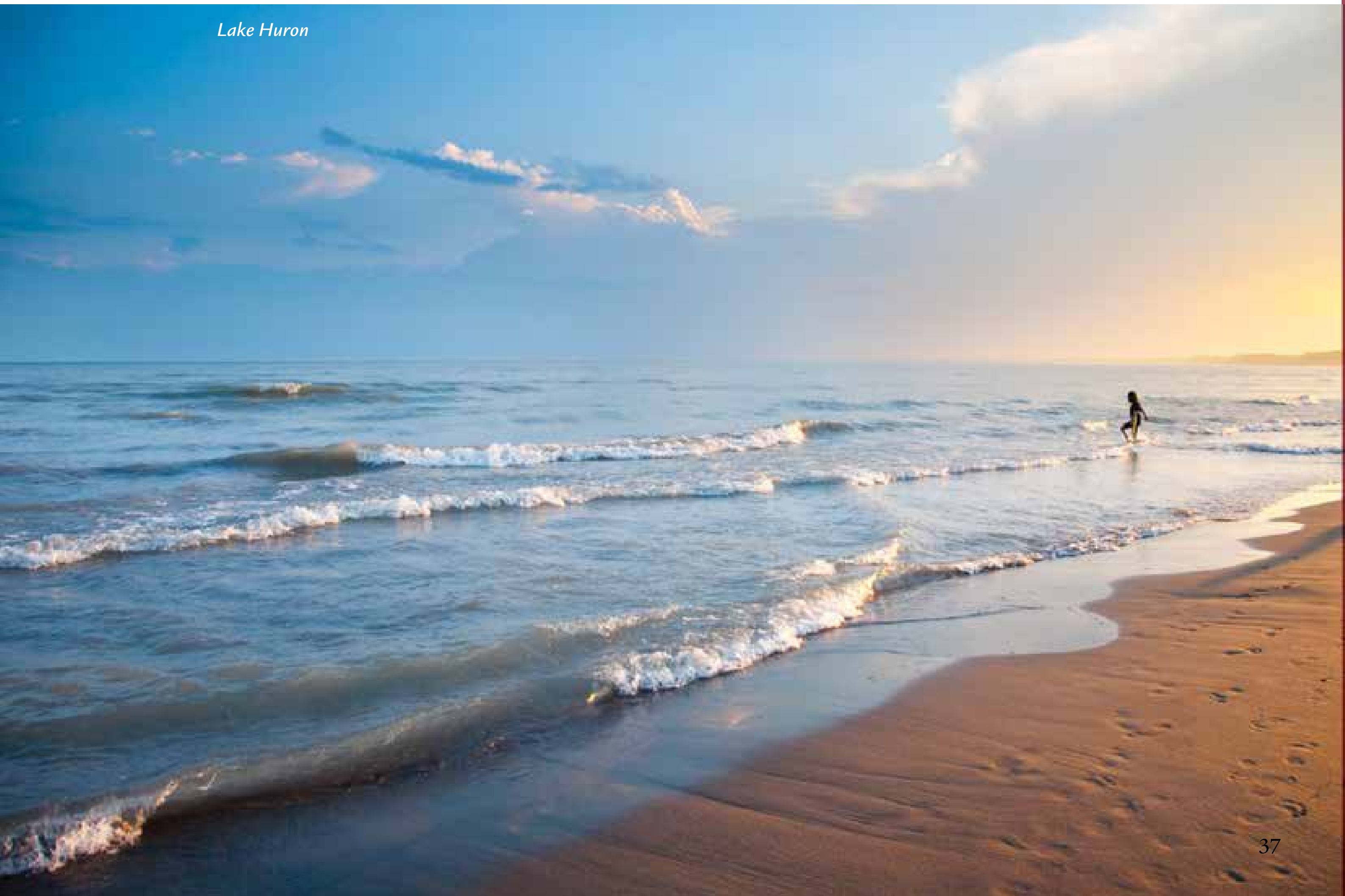
Comprehensive National Mercury Monitoring Network



*Society of Environmental Toxicology and Chemistry

In Summary

Efforts to control mercury pollution in the Great Lakes region have resulted in substantial progress but have not yet addressed the full scope of the mercury problem. The findings from this binational scientific synthesis indicate that mercury pollution remains a major concern in the Great Lakes region and that the scope and intensity of the problem is greater than previously recognized. While many measurements show declining mercury concentrations in fish and wildlife for decades, some observations indicate recent increases in mercury concentrations in particular species in certain areas. Mercury research in the Great Lakes region underscores the benefits of policy advances such as decreases in anthropogenic mercury emissions regionally and nationally. The general trends observed in the Great Lakes region indicate that controlling air emission sources should lower mercury concentrations in aquatic food webs yielding multiple benefits to fish, wildlife, and people in the region. It is expected that these improvements will be greatest for inland lakes and will be roughly proportional to declines in mercury deposition, which most closely track trends in regional and U.S. mercury air emissions.



Lake Huron

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The content for this report was distilled, in large part, from 35 peer-reviewed papers published as special issues in the journals *Ecotoxicology* and *Environmental Pollution*. The guest editors for the *Ecotoxicology* special issue are: David C. Evers, Niladri Basu, James G. Wiener, Drew Bodaly, and Heather Morrison. The *Ecotoxicology* papers are indicated by a plus sign (+). The guest editors for the *Environmental Pollution* special issue are James G. Wiener, David C. Evers, David A. Gay, and Heather A. Morrison. The *Environmental Pollution* papers are indicated by an asterisk (*).

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Credits

Figures: Figure 4 was developed by Kim Driscoll; Figures 1, 7, 10, 12, and 14 were developed by Madeline Turnquist. All figures were designed by RavenMark, Montpelier, VT.

Illustration: Shearon Murphy (Figure 2, yellow perch (p. 12), common loon (p.16), mink (p. 22), trout (p.25). The illustrations for Figure 2 were provided courtesy of The Hubbard Brook Research Foundation.

Photographs: cover: trout by flickr/helti; mink by Eric Bégin; bald eagle by Daniel Poleschook and Ginger Gumm; boy with walleye by flickr/OakleyOriginals; common loon by Steve Walls; Menominee River in Iron Mountain, Mich. by flickr/Lukinosity. *Page 3*: common loon by Daniel Poleschook and Ginger Gumm. *Page 5*: bald eagle by Daniel Poleschook and Ginger Gumm. *Page 9*: Sleeping Bear Dunes by Anne Oeldorf-Hirsch. *Page 15*: wood thrush by Jeff Nadler Photography. *Page 17*: mallard ducklings by Bruce Irschick. *Page 19*: common loons by Fran West. *Page 23*: girl with northern pike by P. Verdonk, woman with walleye by Heather Zibbel. *Page 24*: Kawartha Highlands Provincial Park by Martin Cathrae. *Page 27*: Big Moose Lake by Lida Perfetto. *Page 29*: herring gull by Tom Green. *Page 33*: Lake Erie by flickr/Nicholas_T. *Page 37*: Lake Huron by iStockphoto/ssuni. *Note: photographs appearing more than once are identified by first appearance.*

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