Chapter Three

CHEMICAL INTEGRITY: THE EXAMPLE OF MERCURY

Introduction

The chemical integrity of the Great Lakes is dynamic. The waters of the Great Lakes are continuously changing through the addition, interaction, and loss of both natural and man-made substances. Natural geophysical processes change these substances' spatial and temporal distribution within the Great Lakes system. While much is known, considerable uncertainty remains concerning the chemical integrity of the Great Lakes and the impacts of various chemicals, and combinations of chemicals, on the basin's human and other inhabitants.

Mercury, a persistent bioaccumulative toxic metal, provides an excellent example of the challenges inherent in understanding impacts on the chemical integrity of the Great Lakes. It occurs widely in nature, both in concentrated form in cinnabar (ore) and in small amounts in fossil fuels such as coal. Humans have used mercury for over 3,000 years in medicine and industry.[1] The Commission's Great Lakes Water Quality Board in 1985 identified mercury as one of a "dirty dozen" chemical substances for virtual elimination under the Great Lakes Water Quality Agreement. The governments included this list in the 1987 Agreement in Annex 12: Persistent Toxic Substances. In keeping with this Annex the United States and Canada developed a binational strategy for eliminating releases of 12 persistent toxic substances,[2] including mercury, that provides a framework to achieve specific actions from 1997-2006.[3]

Sources and Forms of Mercury

Mercury reaches the waters of the Great Lakes directly, through discharges into the waters, and indirectly, through disturbances of previous mercury deposition and through atmospheric deposition. This report focuses on contributions from atmospheric sources to the Great Lakes.

Mercury can be released into the air by human activities such as metallurgical processing, municipal and medical waste incineration, and electrical power generation such as from coal combustion. It is also released to the atmosphere by various natural phenomena, including volcanic eruptions, forest fires, and the weathering of geological formations.[4]

Mercury occurs principally in three different chemical forms, or species: elemental mercury, reactive gaseous mercury, and mercury associated with particulates. Different forms of mercury have different solubility, reactivity, and toxicity, behave differently in the atmosphere and the environment, and have different impacts on the ecosystem and on human health.[5]

Elemental mercury can persist for over a year in the atmosphere in a vapor state and, thus, can travel globally with the prevailing winds. Most mercury reaching the Great Lakes from distant sources is in this form.[6] Elemental mercury has limited solubility in water and, as a result, is largely unavailable to fish and other living things. It can be transformed to the other forms of mercury, including the reactive form; however, this reaction proceeds very slowly.

Reactive gaseous mercury (or the ionic form of mercury) is both substantially more soluble in water and more reactive than elemental mercury. It remains in the atmosphere from one to ten days, and therefore tends to be deposited locally and regionally – from a few miles to a few hundred miles from its source. Its limited range of travel, solubility, and high reactivity contribute to its ultimate presence in biota on a regional basis.[7]

Mercury particulate is mercury bound to airborne particles. Mercury particulate can remain in the atmosphere for one to ten days – comparable to reactive gaseous mercury – and thus is deposited regionally and locally. However, it is less available to living organisms than the reactive gaseous form.[8]

Mercury and Human Health

Once deposited in or discharged to water bodies, mercury can be converted by bacteria into organic mercury compounds, such as methyl mercury, that accumulate in the food chain. Human exposure to methyl mercury is predominantly through fish consumption.

Methyl mercury compounds can cross biological membranes, are soluble in lipids and adipose tissues, and can bind to various cell receptors and enzyme sites. Methyl mercury has not been found to be a carcinogen and has not been conclusively established as a teratogen (a chemical which causes a birth defect). Without cancer as a complicating factor, scientists have been able to conduct relatively straightforward analyses of the risks posed by human exposure to mercury compounds. At sufficient levels of accumulation of methyl mercury compounds, toxic effects occur. Serious toxic effects include neurotoxicity (brain and nerve tissue damage) and nephrotoxicity (kidney damage). These toxic effects can impact organisms from birds to mammals, including humans.

At very high levels of methyl mercury contamination, such as observed in Minimata Bay, Japan, in the 1950's, serious health effects occur.[9] Recently, scientists have been exploring the effects of chronic low doses of methyl mercury, particularly for higher risk populations including children, fetuses, and women of child-bearing age. Developing fetuses may be at greatest risk because of methyl mercury's ability to pass through the placenta.

Several cohort studies have been conducted on children who were exposed to methyl mercury before and after birth in the Seychelle Islands and in the Faroe Islands. No neurodevelopmental deficits were identified in the Seychelle Islands children, while some neuropsychological effects were identified in the Faroe Islands children. Notable differences exist between the two populations that may explain the differing results, including diet (ocean fish in the Seychelles versus the higher levels of methyl mercury in pilot whale meat in the Faroe Islands).[10] The studies also raise questions concerning the complicating factor of selenium, its interaction with mercury, and subsequent health effects.[11] Selenium, which is found in some ocean fish, provides a substitute for sulfur that permits a weaker bond with mercury, allowing the human body to remove mercury more easily and excrete mercury in greater quantities, reducing both the exposure period and the dose.[12] No comparable studies to these international efforts have been undertaken in the Great Lakes area. However, recent work intended to investigate the effects of PCB levels on the development of children whose mothers consumed large amounts of fish during pregnancy in the Oswego, New York, area, have also raised questions concerning effects of mercury.[13]

Studies reviewed by the U.S. National Academy of Sciences associate chronic low-dose prenatal methyl mercury exposure with poor performance by children on neurobehavioral tests that measure such things as attention, language ability, fine motor skills, and intelligence.[14] Further research is required to investigate methyl mercury exposure and coronary disease. The majority of epidemiological studies performed has been retrospective, in which linkages are inferred from past events; prospective studies are needed that make a hypothesis and then follow events to observe actual linkages.

Several organizations have established a "reference dose" for methyl mercury. A reference dose is an estimate of a daily exposure to the human population that is likely to be without an appreciable risk of deleterious effects during a lifetime. Different agencies and organizations have established different reference doses, some of which are shown in Table 3. Different Great Lakes states also have different threshold levels for the general public and sensitive populations.[15]

Table 3. Organizations Reference Doses for Methyl Mercury

Organization	Reference Dose (micrograms/kilogram/day)	Uncertainty Factor[16]
U.S. Environmental Protection Agency	0.1	10
Health Canada	0.2	5
Agency for Toxic Substances and Disease Registry (U.S.)	0.3	4.5
World Health Organization	0.47	10
U.S. Food and Drug Administration	0.5	10

Mercury and Fish Consumption

Eating fish offers many nutritional benefits, including protein and omega-3 polyunsaturated fatty acids. However, caution must be taken to avoid eating too much fish containing excessive levels of methyl mercury or other persistent toxic substances. The primary human exposure to methyl mercury is through fish consumption.

In the United States in March 2004, the U.S. Environmental Protection Agency and the U.S. Food and Drug Administration issued a joint consumer advisory on methyl mercury in fish and shellfish for reducing the exposure to mercury in women who may become pregnant, pregnant women, nursing mothers, and young children. The advisory unified and superseded the agencies' 2001 advisories. It advised avoiding fish with relatively high levels of mercury (shark, swordfish, king mackerel, and tilefish), eating up to 12 ounces a week of a variety of fish and shellfish that are lower in mercury (including shrimp, canned light tuna, salmon, pollock, and catfish), and checking local advisories about the safety of fish caught by family and friends in local lakes, rivers and coastal areas (and eating up to 6 ounces a week of fish caught from local waters if no advice is available provided no other fish is consumed that week.)[17]

This third, and perhaps most complicated, provision of the EPA's and FDA's joint advice has particular resonance in the Great Lakes. Site-specific advisories continue to limit or ban consumption of certain fish caught in the Great Lakes because of methyl mercury contamination. In fact, due to localized contaminated sediment, methyl-mercury related fish consumption advisories are expected to exist for decades to come in some Great Lakes Areas of Concern. As an example, the Guide to Eating Ontario Sport Fish contains detailed advice on selecting fish for eating from Ontario rivers and lakes, including the Great Lakes; recommends not eating any organs, fat or skin of any fish; and advises eating smaller fish, eating bass, pike, walleye, perch, and pan fish from the Great Lakes instead of fatty species such as salmon and trout; and allowing fat to drip away when cooking fish.[18]

The Commission previously recommended in its 2000 biennial report that the governments improve fish consumption advisories in the Great Lakes, and the Commission's Health Professionals Task Force (HPTF) recently reported in detail on this issue. The HPTF members support a more effective approach to the development of fish consumption advisories, through better protection of those people at risk, without deterring the majority of people from fish consumption. To develop such an approach, environmental monitoring and exposure assessments are urgently needed to track trends in persistent organic pollutants. Efforts are needed to continue to reduce contaminant levels in all Great Lakes fish.[19]

The Commission's concerns remain relevant today. Advisories are often technical, sometimes offer conflicting advice, and typically fail to reach at-risk populations, including children and women of child-bearing age.[20] For example, according to a study by Kearney and Cole,[21] only 85% of licensed Ontario anglers were aware of the Guide to Eating Ontario Sport Fish, only 29% had a copy of the Guide and followed its advice at least sometimes, and 27% had a copy of the Guide but never followed the advice. Only 50 percent of Great Lakes sport fish consumers reported awareness of a health advisory.[22] Ontario's development of outreach programs directed at school children in non-english speaking communities (in two Areas of Concern) is an example of a communication tool that could improve these statistics.

Complications of Chemical Mixtures

Fish advisories often mention concerns with mercury and PCBs for the same species in the same water bodies. PCBs affect the thyroid, which controls brain development.[23] Mercury binds to brain tissue and may cause other problems. Both PCBs and mercury can pass through the placenta.[24] Therefore, their combination may pose a greater risk to a developing fetus than either alone. Current epidemiological studies are exploring this linkage, and further study is warranted.

Reductions in Mercury Emissions

The U.S. Environmental Protection Agency cites rough estimates showing that 20% of global mercury emissions are from natural emissions, 40% from global re-cycling of previous anthropogenic activity, and 40% from current anthropogenic[25] emissions.[26] As shown in Table 4, North America contributed approximately 11% of the total global anthropogenic mercury emissions in 1995.

Table 4. Global Emissions in Tonnes (~Tons) of Total Mercury from Major Anthropogenic Sources in the Year 1995 – (Pacyna & Pacyna)[27]

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

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

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

With their later industrialization, mercury emissions are now increasing in developing countries. Preliminary findings from U.S. Environmental Protection Agency and Environment Canada indicate that increases in global anthropogenic mercury emissions reaching North America, largely from Asia, offset anthropogenic mercury reductions achieved within the United States and Canada. In the 2001-2003 Great Lakes Priorities Report to the Commission, the findings of the International Air Quality Advisory Board on the transportation and deposition of mercury to each of the Great Lakes via the atmospheric pathway enlarge on these issues.[28] With respect to Lake Superior, the lake most remote from regional industrial sources, the majority of specific sources of mercury deposition were located at a distance greater than 700 kilometres away. Although global emissions are largely of the unreactive form, the sheer volume and increasing proportion of the global mercury balance warrants attention.

Mercury emissions arising from human activity in both the United States and Canada dropped substantially between 1990 and 1999. In the United States, significant mercury reductions came principally from emission controls on municipal and medical waste incinerators, as well as improved screening and removal from the waste stream of commercial products such as batteries and paint. In Canada, significant reductions were achieved largely through controls and process alterations in the metal smelting industry, the near-complete closure of the chlor-alkali industry, and further control and restrictions on waste incineration. In 1999, U.S. mercury emissions were estimated as approximately 124 tonnes (137 tons); further detailed verification of these data now indicate total 1999 emissions were 105 tonnes (116 tons). Canadian mercury emissions were approximately 11 tonnes (12.1 tons). Coal-fired utilities account for approximately 35% and 27% of mercury emissions in the U.S. and Canada, respectively. (See Figures 4 and 5).[29]

Governments in both countries are examining ways to reduce mercury emissions from coal-fired electrical generation facilities. The removal of mercury from coal is technologically challenging.

On December 17, 2003, the Environmental Protection Agency proposed significant reductions in sulfur dioxide (SO2) and nitrogen oxides (NOx) emissions from power plants. Although the proposal targets these chief components of acid rain, it is anticipated that actions taken to meet those standards

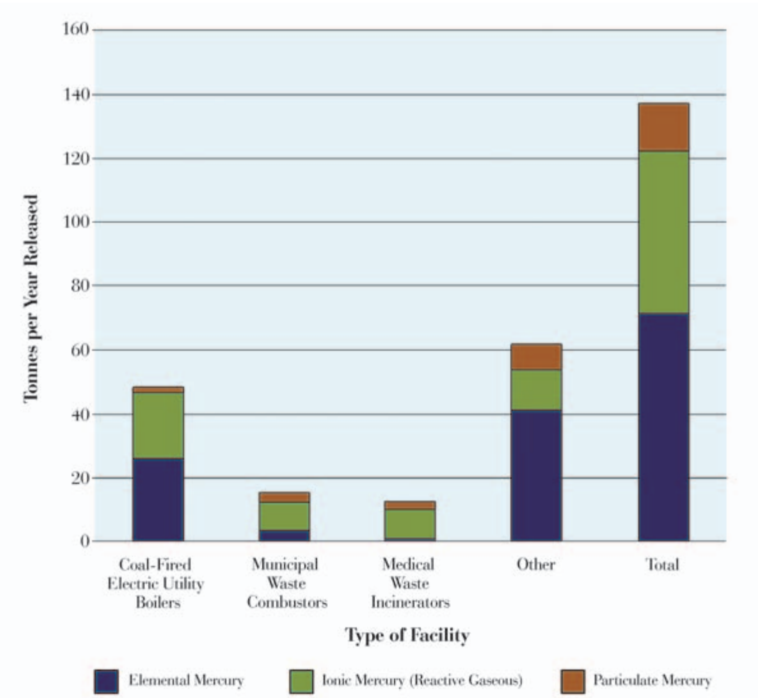

Figure 4. Emission Profile of Mercury Releases — 1999

(Anne Pope, U.S. EPA, 1999 U.S. Natural Emissions Inventory Draft)

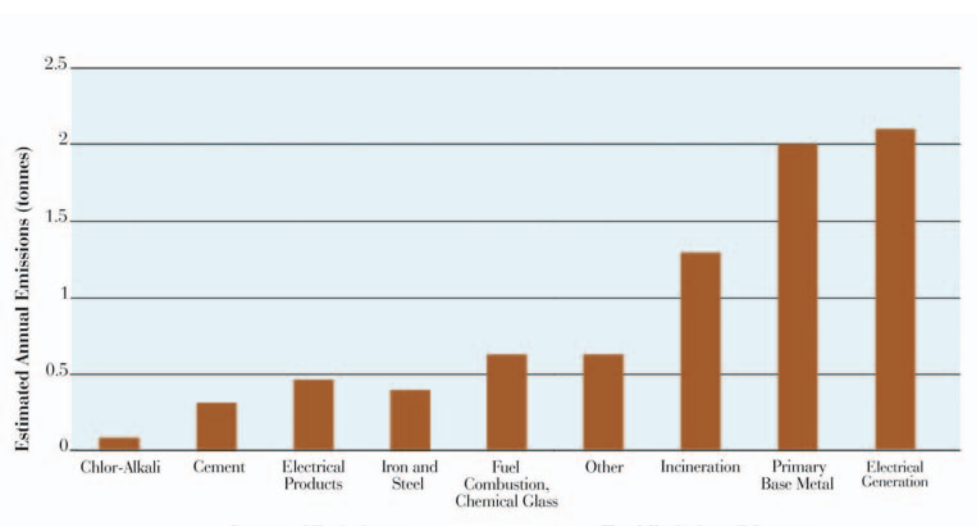

Figure 5. Preliminary Mercury Emissions in Canada — 2000

(Marc Deslauriers, Pollution Data Branch, Environment Canada) will result in "co-benefits" of reductions in mercury emissions and fine particulate matter. The U.S. Environmental Protection Agency has also proposed alternatives for ways to reduce mercury emissions from utilities. These alternatives include creating a market-based "cap and trade" program to reduce mercury emissions in a two-phased approach, and requiring utilities to install controls known as "maximum achievable control technologies" (MACT).

The Canadian Council of Ministers of the Environment has committed to develop a Canada-wide standard to reduce mercury emissions from the coal-fired electric power generation sector by 2010 (with variance in provincial application of the national target or standard), to explore the national capture of mercury from coal burned in the range of 60-90% (including all efforts to reduce mercury releases, from pollution prevention through emissions control), and to align with U.S. standards for mercury. The standard will apply to existing and new plants.[30] As well, Canada-wide standards for mercury-containing fluorescent lamps and dental amalgam waste will assist in meeting the Canada-Ontario Agreement (2002) commitment to reduce mercury releases by 90% by 2010 compared to 1988. Jurisdictions are required to develop an implementation plan which describes what actions will be taken to implement a Canada-wide standard and achieve compliance by the deadline set for the standard, except that as Quebec is not a signatory to the Canada-wide Accord on Environmental Harmonization nor the Canada-wide Standards, it is not required to develop an implementation plan;[31] however, the principal mercury sources from Quebec are included in the National Pollutant Release Inventory.

With respect to the lower Great Lakes, information provided by the Commission's International Air Quality Advisory Board in the 2001-2003 Priories Report shows that there are significant regional and local sources of mercury emissions. The Board's report shows that 40% of the mercury emission from coal-fired generation facilities in that region is in the more biologically-available reactive form. The waters of the Great Lakes also continue to receive mercury from previously contaminated sediments. In addition, contaminated groundwater and wastewater discharges contribute to the local mercury burden, especially in Areas of Concern. Because the U.S. and Canadian governments can control emissions from sources within their jurisdictions more effectively than some global emissions, and because reactive gaseous mercury is more biologically available, governments should substantially reduce the deposition of reactive gaseous mercury in the Great Lakes region.

The combined impacts of mercury contamination in Canada are difficult to quantify,[32] and the exact proportion of the impact which can be ascribed to natural mercury and to past and present anthropogenic releases cannot presently be quantified.[33] The U.S. Environmental Protection Agency cited a plausible link between mercury from industrial combustion sources and methyl mercury in fish, but noted that it was not possible to quantify how much of the methyl mercury in fish consumed by the U.S. population is contributed by U.S. emissions relative to other sources of mercury (such as natural sources and re-emissions from the global pool).[34] A recent study in the Florida Everglades estimated how quickly fish tissue levels respond to decreased regional mercury emissions. Reductions in total mercury emissions of approximately 90% since the late 1980's have been paralleled by a reduction in average fish tissue methyl mercury of about 80%.[35] However, more definitive information, ideally through studies focused on the Great Lakes, would be helpful in exploring linkages between mercury emissions and deposition, and biologic uptake and effects.

Conclusions

Mercury provides a case study for chemical integrity. Much is known about mercury's toxic effects at higher doses, and there is a growing body of knowledge concerning effects at lower doses. At sufficient levels of accumulation of methyl mercury compounds, toxic effects occur, including neurotoxicity (brain and nerve tissue damage) and nephrotoxicity (kidney damage).

The Commission recognizes that both governments are currently considering proposals for further reductions in mercury emissions. The Commission encourages both governments to adopt and implement initiatives that will further reduce the release of mercury to the environment, including atmospheric emissions.

The effects of past mercury emissions, compounded by continuing emissions, will remain an issue for decades to come. Associated risks must be effectively communicated. While both governments have compiled and disseminated fish advisories, difficulties remain in reaching those most at risk and in effecting changes in behavior that would reduce that risk.

Significant gaps in knowledge remain about the processes by which mercury moves from source to water body, to fish and wildlife, to humans, and about the effects of low doses of mercury on human health. Scientists continue to explore plausible connections and build on the knowledge base. In addition to general studies of this nature, specific focused studies on mercury deposition and its effects on the Great Lakes are required.

Recommendations

The Commission recommends that the two federal governments, in conjunction with the states and provinces and institutions:

- Undertake retrospective and prospective epidemiological studies, in Areas of Concern and other pertinent locations of the Great Lakes basin, to better understand potential neuro-developmental effects associated with methyl mercury and PCBs.

- Make fish advisories clear, simple, and consistent, and ensure that they are reaching the intended audiences.

- Select and promptly implement programs in both the United States and Canada that would substantially reduce the deposition of mercury in its reactive gaseous form in the Great Lakes region; and also pursue multi-lateral strategies for further control of this persistent toxic substance on a global basis.