Lake Superior Lakewide Management Plan:
1990-2010
Critical Chemical Reduction Milestones
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"We need to bring all the people of Lake Superior together. We need to talk to each other about what is happening in our villages and our communities, to share our experiences, our concerns, and our hopes for the future. We need to meet our neighbors and learn from them."

- Walter Bressette, Anishanabe elder

Lake Superior watershed sign installed by the Minnesota LaMP program.

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Executive Summary

Background. In 1990, the International Joint Commission's (IJC's) Fifth Biennial Report on Great Lakes Water Quality challenged the governments of Canada and the United States to "designate Lake Superior as a demonstration area where no point source discharge of any persistent toxic substance will be permitted." The following year, the *Lake Superior Binational Program to Restore and Protect Lake Superior* (or the *Lake Superior Binational Program* [LBSP]) was announced, providing for a Zero Discharge Demonstration Program (ZDDP) and a "broader program" focusing on ecosystem restoration. (Appendix A1 includes the complete 1991 agreement.) The Lake Superior Lakewide Management Plan (LaMP), a management strategy developed by Lake Superior partners, was developed to implement the ZDDP and ecosystem restoration programs. The LaMP set reduction schedules for the nine ZDDP chemicals:

- Mercury;
- Polychlorinated biphenyls (PCBs);
- Pesticides (including: aldrin/dieldrin, chlordane, DDT/DDE, and toxaphene);
- Dioxin;
- Hexachlorobenzene (HCB); and
- Octachlorostyene (OCS).

Results. This *2010 Critical Chemical Reduction Milestones* report documents progress in reducing the nine ZDDP chemicals between 1990 (baseline) and 2010 and includes discharge and emissions inventories from sources within the Lake Superior Basin (LSB) in 1990, 2000, 2005 and 2010. The program has many successes resulting from the activities, collaborations and commitments of a wide range of actively-engaged state, provincial, tribal, and federal agencies, industries, non-governmental organizations, and citizens. Challenges, however, still remain. Details of these activities since 2005 can be found in Appendix B.

In 2010, discharge and emission inventories for the ZDDP chemicals were updated for the entire LSB. These included an analysis of emissions from a broad range of sources including: mining, fuel combustion, incineration, waste handling, commercial products, and municipal wastewater and solid waste facilities. Details of the inventories are presented in Appendix C.

Notable achievements in reducing discharges and emissions include:

- 80% reduction in mercury discharges and emissions;
- 86% reduction in dioxin discharges and emissions;
- Ongoing safe collection and disposal of PCB-containing materials; and
- Ongoing safe collection and disposal of ZDDP pesticides, including more than 4,800 kg (10,600 pounds) collected between 1992 and 2007.

While the LaMP program is limited in its ability to differentiate the relative effects of in-basin versus out-of-basin sources of contaminants on the Lake Superior ecosystem, levels of ZDDP chemicals in general have declined and concentrations in Lake Superior are often (but not always) lower in Lake Superior air, water, sediment, fish, and wildlife. Some persistent issues under evaluation include:

- A notable exception is mercury in fish, which has begun to trend upwards. Lake Superior also exceeds the other Great Lakes in mercury levels in fish.
- While toxaphene levels in Lake Superior fish remain higher than other Great Lakes fish, a recent study of the trend in lake trout indicates a steady decline since 2000 and possible leveling off starting in 2007.
- Fish consumption advisories in Lake Superior continue for mercury, PCBs, chlordane, dioxin, and toxaphene.
- PCBs, dieldrin, and toxaphene levels in the open waters of Lake Superior exceeded water quality standards in both the 2005 and 2010 milestone reports.
- 8% of newborns in the Lake Superior watershed exceeded the Reference Dose (RfD) for mercury. A seasonal effect was also found, suggesting locally-caught fish is an important source of pregnant women's mercury exposure.

Remaining challenges include inventory development and quantification, identifying further reduction challenges and opportunities and improving the ability to accurately quantify ZDDP chemical sources, such as:

- In-service (or in-use) PCB-containing articles and equipment;
- Unknown stockpiles of banned pesticides;
- HCB estimate(s) for iron sintering; and
- Smaller sources that are known to emit ZDDP chemicals, but are not easily quantified (e.g., land clearing and mobile sources).

Other reduction challenges include:

- Emissions from existing taconite mining and possible new or expanded mining;
- A lack of dioxin reduction progress between 2005 and 2010; and
- Sources that are proportionately more important as other sources have been reduced (e.g., mercury from human cremation).

Achieving the program's 2015 targets and moving toward the 2020 goal of zero discharge and zero emission will be difficult. The Lake Superior partners, however, remain committed to achieving the goals of the Lake Superior Binational Program.

Summary. Emission reductions of critical chemicals have been documented by – and achieved through – the ZDDP. The ZDDP has shown that Great Lakes stakeholders can indeed be successful in reducing sources of toxic chemicals. In summary, 2.1 tonnes (2.3 tons) of mercury was released from sources in the basin in 1990; this is now reduced to 0.4 tonnes (0.4 tons) in 2010. Dioxin is following a similar trend as mercury but preventable sources still dominate the inventory. 4.6 tonnes (5.0 tons) of ZDDP pesticides was collected from just the Minnesota counties bordering Lake Superior since 1992 and pesticides are also collected in Wisconsin, Michigan, and Ontario. PCB equipment in service or storage is diminishing but disposal rates have not yet leveled off.

Chapter 1. Scope and Background

1.1 LaMP Critical Chemicals and the Zero Discharge Demonstration

As observed in LaMP 2000 Chapter 4, Annex 2 of the 1987 Canada-U.S. Great Lakes Water Quality Agreement (GLWQA) contains a framework for LaMPs to restore beneficial uses and reduce the loadings of critical pollutants (LSBP, 2000). In their 1990 biennial report on the GLWQA, the IJC called for the Parties to establish a Zero Discharge Demonstration Area for Lake Superior.

In response, government agencies in 1991 established *A Binational Program to Restore and Protect the Lake Superior Basin*, also known as the LSBP (see Appendix A). Included in this program are the ZDDP, with a goal of zero discharge and zero emission of nine persistent bioaccumulative and toxic substances (PBTs) and a "Broader Ecosystem Program" that focuses on the non-chemical elements of the Lake Superior ecosystem. The LSBP identifies nine chemicals that are targeted for zero discharge and zero emissions because of their presence in Lake Superior water, fish, or wildlife.

The LaMP prioritizes actions and projects that will help achieve the goals of the ZDDP. The ZDDP chemicals and the other chemicals already designated as critical under the LaMP process are listed in Table 1-1a. They fall into three management categories: zero discharge, lakewide remediation and local remediation. Further information about the LaMP process can be found at www.epa.gov/glnpo/lakesuperior.

Prevention chemicals are either in the "Monitor" category (present but not exceeding "yardsticks") or "Investigate" category (data from Lake Superior are needed to evaluate this chemical); both categories are listed in Table 1-1b. Prevention chemicals are essentially on a "watch list" that requires additional information and follow-up. Only the nine ZDDP chemicals are targeted for zero discharge in the LSBP; the other critical chemicals are subject to virtual elimination per the GLWQA.

Stages 1 and 2 of the chemical portion of the LaMP, which describe the status of pollutants in the Lake Superior ecosystem and set load reduction targets for critical pollutants, respectively, have been completed. Chapter 4 of the LaMP 2000 then proposed remedial measures for these Lake Superior critical pollutants. The 2005 Chemical Milestones report (LSBP, 2006a) identified actions taken toward those remedial measures, estimated the load reductions since 1990, and identified further reduction strategies. This 2010 Chemical Milestones report updates the load reduction estimates from 1990 to 2010, identifies remedial measures taken since 2005, and identifies additional reduction strategies still needed to achieve future milestones.

The load reduction schedule from Stage 2 (Table 1-2) describes four timelines for reductions of mercury, PCBs, dioxin/HCB/OCS and the targeted pesticides. Note that although 2010 is a milestone year for mercury and PCBs only, the report documents progress on all four chemical groups.

Table 1-1a. Existing Critical Chemicals for Lake Superior

Management Category	Chemical	
Zero Discharge	Chlordane DDT and metabolites Dieldrin/aldrin Hexachlorobenzene PCBs	2,3,7,8– Tetrachlorodibenzo-p-dioxin (TCDD) Toxaphene Mercury OCS
Lakewide Remediation	Polycyclic aromatic hydrocarbons (PAHs) (anthracene, benz(a)anthracene, benzo(b)fluoranthene, dinitropyrene, benzo(a)pyrene, pyrene, benzo(g,h,i)perylene, phenanthrene)	Alpha-hexachlorocyclohexane (BHC) Cadmium Heptachlor/heptachlor epoxide TCDD(TEQ)[1] dioxins and furans
Local Remediation	Aluminum Arsenic Chromium Copper Iron	Lead Manganese Nickel Zinc

[1] The Binational Program lists 2,3,7,8-TCDD (dioxin) for the ZDDP. By convention, dioxin is measured and reported as toxic equivalents (TEQ) of TCDD.

Table 1-1b. Existing Prevention Chemicals for Lake Superior

Management Category	Chemical	
Monitor	1,4-dichlorobenzene 1,2,3,4-tetrachlorobenzene Mirex/photo-mirex	Pentachlorobenzene Pentachlorophenol BHC, gamma congener
Investigate	1,2,4,5-tetrachlorobenzene 3,3-dichlorobenzidine 2-chloroaniline Tributyl tin	BHC, beta and delta congeners Hexachlorobutadiene

Table 1-2. Summary of Reduction Targets for Lake Superior ZDDP

Pollutant	Reduction Schedule (1990 base line)
Mercury	60% reduction by 2000 80% reduction by 2010 100% reduction (zero discharge/zero emission) by 2020 (applies to in-basin sources)
PCBs	Destroy PCBs in service or in storage 33% destruction by 2000 60% destruction by 2005 95% destruction by 2010 100% destruction by 2020
Pesticides **Aldrin/Dieldrin,** **Chlordane,** **DDT/DDE, and** **Toxaphene**	Retrieve and destroy all cancelled pesticides in the basin by the year 2000
Dioxin [1] **HCB** **OCS**	80% reduction by 2005 90% reduction by 2015 100% reduction by 2020

1 The LSBP lists 2,3,7,8-TCDD (dioxin) for the ZDDP. By convention, dioxin is measured and reported as toxic equivalents (TEQ) of TCDD.

1.2 Progress and Accountability

In the LaMP 2000 Chapter 4, the Chemical Committee identified reduction strategies for different sectors (e.g., mining, forestry, health care, schools, etc.). The 2005 Chemical Milestones report includes additional reduction strategies. Reduction and inventory activities needed to make progress toward the 2010 reduction milestone were identified in Addendum 4C of the 2008 LaMP. Agency reports on progress toward the LaMP activities and strategies identified in these documents between 2005 and 2010 are included in Appendix B. Highlights are summarized below.

Note that the reduction activities in Appendix B are split into three types: 1) LaMP Chemical Reduction Activities, 2) Other Projects Aligned with LaMP Goals, and 3) New Regulations and Policies Aligned with LaMP Goals. While the LaMP program directly implements toxic reduction projects depending on available funding, the LaMP agencies recognize the importance of tracking other programs' projects as well as developments in broader regulations and policies that are aligned with LaMP goals.

1.2.1 LaMP Chemical Reduction Activities

The following highlighted activities are a direct result of the LaMP (i.e., activities that were funded for LaMP implementation and in which workgroup members had an active role):

- Collections were carried out in different parts of the basin, including electronic waste and pharmaceutical collections by the non-profit, faith-based organization, Earth Keepers, in the Upper Peninsula. First time hazardous waste collections also took place in some Ontario communities. Various outreach activities promoted collections and waste diversion on tribal reservations and in Canadian First Nations communities. The Western Lake Superior Sanitary District (WLSSD) continued "Medicine Cabinet Clean-Out Days". Several local governments sponsored mercury product collections.
- Technical data sharing included the first LaMP-hosted Lake Superior conference held since 1990. The 2007 "*Making a Great Lake Superior"* conference included a toxic chemical session facilitated by the LaMP Chemical Committee.
- Other outreach efforts included presentations at tribal events (fisher meetings, tribal open houses and health fairs, powwows), the *Midwest Society of Environmental Toxicology and Chemistry* and the *Eighth International Conference on Mercury as a Global Pollutant* by the Great Lakes Indian Fish and Wildlife Commission (GLIFWC) on reducing health risks from eating fish. GLIFWC also regularly publishes articles on healthy fish consumption in its triannual newspaper, the *Mazina'igan,* and distributes Mercury Maps to tribal members illustrating the mercury concentrations in walleye from various lakes.
- Open burning abatement projects included the conclusion of a project that involved three local Minnesota governments and the Minnesota Pollution Control Agency (MPCA). In Ontario, Neebing Township prohibited trash burning. Outreach was conducted on First Nations communities to discourage open burning.

1.2.2 Other Projects Aligned with LaMP Goals

The following highlighted projects were not a direct result of the LaMP but are in alignment with LaMP goals and took place in the LSB:

- Energy conservation and alternative energy projects were carried out by a variety of entities at several levels. For example, a number of buildings recently built or remodeled in the LSB have been certified by the Leadership in Energy and Environmental Design (LEED), including nine college campus projects, two health care clinics, the Resource Management and Tribal Court building on the Fond du Lac reservation, a business, Coast Guard station and a rural electric cooperative building. An additional 33 projects have registered for LEED certification.
    - Minnesota Power has increased wind power development in its portfolio, including Oliver County and Bison wind farms in North Dakota and Taconite Ridge in Minnesota. Several tribes in the Lake Superior basin (Fond du Lac, Bad River, Keweenaw Bay Indian Community, Red Cliff and Grand Portage) have begun evaluating wind power potential on their reservations. The Fond du Lac Band has also installed solar panels on its Ojibwe school.
    - The Greenwich Wind Farm under construction in the Township of Dorion (Ontario). Once complete, it will provide approximately 100 megawatt (MW) of power under optimal operating conditions. This is enough renewable energy to power around 30,000 typical Canadian homes each year.

- The Prince Wind Farm, located northwest of Sault Ste. Marie, was Canada's largest wind farm when it began operation in November 2006. It has 126 wind turbine generators and a combined installed capacity of 189 MW.

- Wastewater treatment plants throughout the basin are being upgraded. Canada and Ontario have announced funding for upgrading Nipigon and Red Rock sewage treatment facilities from primary to secondary treatment. The Town of Nipigon has completed the upgrade of its primary sewage treatment plant to secondary treatment standards and is operational. The Town of Red Rock is currently undertaking an environmental assessment to determine its preferred option to upgrade to secondary treatment standards. These upgrades will help with the delisting of Nipigon Bay as an Area of Concern (AOC) by reducing the amount of municipal wastewater pollution entering the bay. In Michigan, the City of Ishpeming has documented a decrease in mercury discharge after requiring amalgam separators at dental offices. The City of Marquette is upgrading its wastewater treatment plant with activated sludge and new secondary clarifiers. Tribal wastewater projects include extending sewer lines and bringing failing septic systems up to code.
- Household hazardous waste (HHW) and pesticide collections were carried out, including city, county, tribal/First Nations, and regional HHW and waste pesticide collections. The Bad River and Red Cliff Bands carried out programs to eliminate elemental mercury in thermometers and sphygmomanometers at tribal health clinics.
- Sediment projects, including both studies and implementation, have been carried out on both sides of the border. For example, design and federal environmental assessment are underway for the thin-layer cap for contaminated sediment in Peninsula Harbour. Assessment of sediment management options is underway at Thunder Bay North Harbour. In St. Marys River, studies are being done to determine whether deeper, more-contaminated sediments may be exposed during increased flow, ice scour and changes in water level at Bellevue Marine Park. Assessments are being done at two sites downstream of the park to determine what sediment management may be required. Sediment characterization and assessment projects are also underway in the St. Louis River AOC, by Minnesota and Wisconsin, which will facilitate prioritization of areas for remediation and restoration.
- The Keweenaw Bay Indian Community has completed remediation of a brownfield site on its reservation. The Sand Point brownfield site was capped in 2006 and re-vegetated with native flora in 2011.
- Under the Great Lakes Regional Collaboration (GLRC), the Great Lakes states, tribes, and cities worked with the U.S. Environmental Protection Agency (U.S. EPA) to develop two regional strategies to address mercury in the Great Lakes Basin. One strategy focused on mercury in products and the other on atmospheric mercury emissions. The Mercury in Products Phase-down Strategy was developed in 2008 and a Mercury Emissions Reduction Strategy was developed in 2010.
- The Great Lakes Air Deposition Program funded a study that brought together over 170 scientists and managers from around the Great Lakes Basin to compile and evaluate over 100,000 mercury measurements and conduct new modeling and analyses. The *Great*

Lakes Mercury Connections integration report summarizes the technical published documents.

- The MPCA's statewide mercury Total Maximum Daily Load (TMDL) was approved by U.S. EPA in 2007. In 2008, Minnesota stakeholders made recommendations in the Strategy Framework for Implementing Minnesota's Statewide Mercury TMDL. This includes mercury reduction schedules for various sectors.
- In January 2008, the Michigan Department of Environmental Quality (MDEQ) released its Mercury Staff Report, a state-wide strategy to eliminate anthropogenic mercury use and releases in Michigan. As MDEQ implements the report's recommendations, further reductions in mercury loadings to Michigan's environment should result.
- In addition to these reduction projects, the Minnesota Department of Natural Resources Minerals Department has received federal funding through the Great Lakes Restoration Initiative (GLRI) to examine different mercury emission control technologies at taconite plants. This sector is the largest source of mercury emissions in the 2010 LSB inventory.
- A statewide open burning survey was done by Minnesota in 2010. Results show that statewide, rural Minnesotans are burning 12% less than in 2004. In northeastern Minnesota, which includes the LSB, the drop was 18%. In 2005, the Bad River Band of the Lake Superior Tribe of Chippewa Indians initiated a Burn Barrel Buyback Program to reduce open burning on the reservation.

1.2.3 New Regulations and Policies Aligned with LaMP Goals

Some government regulations and policies have been developed since the 2005 Chemical Milestones report that affect releases of the nine chemicals targeted for zero discharge. Those that are most closely aligned with contaminants in the LSB include the following:

- New PCB regulations were published in September 2008 in *Canada Gazette II*. The purpose of these regulations is to minimize the risks posed by the use, storage and release of PCBs by accelerating the elimination of these substances. An amendment was published in *Canada Gazette II* on March 31, 2010.
- On August 24, 2007, Ontario implemented *Ontario Regulation 496/07* that requires cessation of coal use at the remaining four coal-fired plants, including Thunder Bay, by December 31, 2014.
- In December 2011, the U.S. EPA issued the first national standards for mercury pollution from power plants, entitled the *Mercury and Air Toxics Standards (MATS),* pursuant to a 20-year legal requirement to reduce dangerous air toxics. U.S. EPA estimates that these standards will greatly reduce emissions of mercury, arsenic, acid, nickel and cyanide, preventing up to 11,000 premature deaths per year. In Michigan alone, the U.S. EPA estimates that the MATS rules will prevent up to 410 deaths and will result in \$1.4 to \$3.4 billion of health benefits to Michigan residents in 2016.
- Minnesota's Next Generation Act was passed in 2007. It established a strong renewable energy standard which requires energy companies to provide 25% of power from renewable sources by 2025, appropriated funding for energy projects and research, and

established statewide greenhouse gas reduction goals of 15% by 2015, 30% by 2025, and 80% by 2050.

- In 2008, Wisconsin passed a rule to control mercury emissions from coal-fired power plants. The rule requires a 90% reduction of mercury emissions or acceptance of a 0.0080 pounds mercury per gigawatt (GW)-hr limitation from large coal-fired power plants by January 1, 2015. Large coal-fired power plants also have the option of choosing a multi-pollutant alternative. The multi-pollutant alternative requires the affected power plants to achieve nitrogen oxides (NOx) and sulfur dioxide (SO_2) reductions beyond those currently required by federal and state regulations.
- In 2009, Wisconsin enacted a law that prohibited the sale of a number of products that contain mercury, including thermometers, manometers, thermostats, barometers, hydrometers, toys, jewelry and over-the-counter drugs.
- Since February 2009, the United Nations Environment Programme (UNEP) has been developing a globally legally binding instrument to control mercury pollution (UNEP, 2012; U.S. EPA, 2012).

Chapter 2. Introduction

2.1 Purpose

This Critical Chemical Reduction Milestones report is intended to provide a summary of progress that has been made since 1990 towards reducing the nine chemicals targeted for zero discharge (see Table 1-1). The summary includes inventories of mercury, dioxin and PCBs (to the extent possible), including amounts recovered in collections, amounts estimated to be released and where possible, amounts estimated to be retained in storage, in service and in sediment. The estimated reductions are then compared to the Stage 2 reduction targets (see Table 1-2).

As well as summarizing progress towards the 2010 targets, this report also identifies strategies for making progress toward the reduction targets for 2015. In addition, the report examines the strategies for addressing the other critical and prevention pollutants (see Table 1-1) and emerging contaminants as well as the nine chemicals targeted for zero discharge.

2.2 Methods

The original Lake Superior Binational Agreement (Appendix A) provided guidance on three types of activities that should be pursued as part of the ZDDP. These included pollution prevention, special protection designations and controls and regulations. Over time, the binational partners have refined the original guidance into a set of guiding principles.

2.2.1 Three Actions from the Lake Superior Binational Agreement

Of the three types of actions, the most productive so far for achieving reductions has been pollution prevention (P2). A number of the projects listed in Section 1.2.1 and 1.2.2 are classic examples of P2. Through P2, the "low hanging fruit" has been reduced but the remaining sources are more difficult to reduce. It is the intent of the binational partners to prioritize P2 as the preferred reduction strategy (see Guiding Principles in Appendix A2).

The second type of activity involves the development of special protection designations. Most of the special protection designations mentioned in the agreement have been implemented. The Outstanding International Resource Water (OIRW) designations were adopted by Michigan and Minnesota before LaMP 2000 was released. Wisconsin adopted special protection designations for Lake Superior with administrative rule revisions in 2006. The OIRW designation and Wisconsin's equivalent designation require new or expanded discharges to use best technology in process and treatment. Wisconsin also included greater protections for additional Lake Superior tributaries and certain nearshore areas as part of state Outstanding Resource Waters (ORWs) designations in 2006.

On the Canadian side of the basin, Parks Canada is in the process of establishing the Lake Superior National Marine Conservation Area from Thunder Cape at the tip of Sleeping Giant Provincial Park in the west, to Bottle Point just east of Terrace Bay, and extending south in the lake to the Canada-U.S. border. It will include the waters of Black Bay and Nipigon Bay and cover a total area of 10,850km^2. Once created, it will be the largest freshwater protected area in the world. A Memorandum of Agreement between Canada and Ontario for establishing the conservation area was signed in 2007. In the meantime, Parks Canada continues to work with

First Nations, local communities, various government organizations and local stakeholders to address outstanding issues that will bring the area closer to establishment. The Harmonization Committee was created in October 2010. This committee is made up of various provincial and federal governments with overlapping roles within the Lake Superior National Marine Conservation Area.

The third activity type, controls and regulations, includes a number of regulatory activities that were under development when the Lake Superior Binational Agreement was approved in 1991. Since then, various programs have been implemented and others are under development or consideration. Section 1.2.3 summarizes the most recent regulations and government policies that will have the greatest impact in the basin.

2.2.2 Guiding Principles

In 1997, the Lake Superior Task Force (composed of administrators and senior managers from the various government agencies and partners under the Lake Superior LaMP) crafted a set of guiding principles to clarify the approach used to achieve load reduction targets toward reaching zero discharge. These were subsequently published in the LaMP Stage 2 in 1999. In 2004, these guiding principles were updated and served to guide continuing implementation of the ZDDP (Appendix A2).

Chapter 3. Load Reduction Inventory

Since the 1990 baseline year, releases of the nine designated chemicals have declined in the LSB. Between 1990 and 2000, reductions occurred primarily because of the closures of two mining facilities (White Pine Mine copper smelter in Michigan and Algoma Ore Division iron sintering facility in Ontario). Other reductions occurred because of changes in mercury-bearing products such as paint and batteries, changes in incineration rules, a U.S. EPA-driven Great Lakes-wide phase-out of PCB equipment, and hazardous waste and pesticide collections. Since 2000, additional reductions have occurred, mostly in the industrial, incineration, and product source categories.

3.1 Out-of-Basin Sources

As discussed in the LaMP 2000 (Chapter 4, pages 4-82 to 85), reductions in out-of-basin sources of toxic chemicals are needed to reduce contaminant levels in Lake Superior. While the LaMP program itself cannot drive state, provincial, national and international policy and regulations that affect emissions, it is in the best interests of the LaMP partners to participate in these efforts to reduce toxic chemicals from being imported into the LSB via atmospheric deposition and products. Participation in out-of-basin reduction programs by LaMP partners is reported in Sections 1.2.2 and 1.2.3 and Appendix B

3.2 In-Basin Inventory Methodology

This section describes load reduction estimates for 1990, 2000, 2005 and 2010. Both the Canadian and U.S. inventories have been reviewed and updated for all four time periods. Appendix C shows a more detailed version of each of the two nations' updated mercury and dioxin inventories for the LSB. Whenever possible, actual measurements of discharges and emissions were used for the inventory. Where directly measured data were not available, a variety of estimates were used. These include databases such as the National Pollutant Reduction Inventory in Canada and the National Emissions Inventory in the U.S., estimates derived from emission factors and throughput information from basin facilities (e.g., taconite mercury emissions), and population normalized numbers that are based on other inventories. Readers of this document are encouraged to supply updated inventory estimates for review by the Chemical Committee.

When the U.S. and Canadian inventories were combined, there were some differences in categories used to report, methodologies used, or inventory calculation. These reporting differences may have resulted in categorical and subtotal/total changes between the Chapter 3 and Appendix C tables.

In addition to estimating discharges and emissions of mercury and dioxin, Environment Canada and U.S. EPA attempted to estimate discharges and emissions for HCB. Select sources of HCB were also identified and included in the inventory. These estimates are not considered as complete as the mercury and dioxin inventories, but Section 3.7 presents preliminary estimates. In addition to estimating discharges and emissions, the partners have estimated the amount of mercury, dioxin, and HCB in ash, sludge, contaminated soil, contaminated sediment, disposed materials, recycled materials, and/or mine tailings and waste rock when possible. These tables are summarized in Appendix C.

3.3 Mercury

3.3.1 Mercury Reduction Goals

The reduction goals for mercury discharges and emissions described in LaMP Stage 2 include the following (1990 baseline):

- 60% reduction by 2000
- No formal mercury milestone for 2005
- 80% reduction by 2010
- No formal mercury milestone for 2015
- 100% reduction by 2020

In Section 3.3.2 below, it is estimated that an 80% reduction of mercury emissions and discharges has taken place since 1990, which meets the mercury reduction goal for 2010. In order to meet the 100% reduction goal by 2020, an additional 417 kg/yr of mercury must be reduced from 2010 loads.

3.3.2 Sources of Mercury

The mercury inventory in Table 3-1 includes releases to both air and water for the baseline year, as well as the milestone year of 2000, non-milestone year of 2005, and milestone year of 2010. It should be noted that discharges (i.e., to water) are only a small portion of the releases. In 1990, discharges represented <2% of the total discharges and emissions, but by 2010 discharges dropped to <0.7%. (See Appendix C for detailed estimates of discharges and emissions).

Some changes have been made to the inventory tables since the first version appeared in LaMP 2000. In both the 2005 and 2010 milestone inventory analyses, Environment Canada and U.S. EPA investigated the previous methods and assured consistency with the most recent estimates. Table 3-1 shows the revisions to the mercury inventory among the three reports. In the LaMP 2000, the first estimates of the 1990 baseline discharges and emissions were made, along with estimates for 2000 (but no projections were made for future milestones). In the 2005 Milestones report, the 1990 and 2000 estimates were recalculated and the 2005 estimates were made (again, no projections were made for future milestones). In this 2010 Milestones report, 1990, 2000, and 2005 estimates are recalculated and the 2010 emissions have been calculated. No projections were made for 2015.

Although these three reports show a trend of decreasing estimated mercury emissions, this is attributable to improved database and inventory methods over the years. Hence, this decrease should not be interpreted as a decrease in actual emissions. Unless the background inventory documents for each report are consulted, it is inappropriate to compare estimates between LaMP 2000, the 2005 Milestones report, and this current 2010 Milestones report. For the purposes of trend analysis of sources, the 2010 Milestones report estimates are considered the best estimates available. Previous numbers in the LaMP 2000 and 2005 Milestones report are considered out of date.

Table 3-1. Revisions to Mercury Discharge and Emission Estimates in LaMP 2000, 2005 Milestones Report, and Current 2010 Milestones Report, kg/yr

LaMP Report	1990	2000	2005	2010
LaMP 2000	2444	819	NA	NA
2005 Milestones	2250	700	653	NA
2010 Milestones	2136	617	597	417

NA – Not Applicable

While the inventories have improved, there are still uncertainties and limitations that must be noted. For mercury, the caveats that must be considered include:

- In Canada, a considerable quantity of mercury is estimated to be present in discarded mercury relays and instrumentation and control equipment. A high recycling rate was assigned for mercury relays (60%) and for instruments and control equipment (50%) for the year 2010 based on the work of Cain (2005) and Cain et al. (2007), which is in turn based on U.S.-based practices.
- Other consumer products not in the Canadian inventory include preservatives, reagents, mercury compounds, and other mercury-added products. However, the combined mercury present in these products is small compared to the amount in switches and relays, instrumentation and control, dental amalgam, and the other consumer products examined in this report.
- A systematic process is needed in Canada for identifying and managing mercury-containing equipment in industrial, commercial, and institutional facilities. Information gathered from such a project would assist in providing a more accurate estimate of the fate of mercury in these products once they are discarded.
- The inventory of mercury-containing consumer products being disposed in the LSB (Ontario) needs improvement.
- 2010 National Pollutant Release Inventory (NPRI) data were not available during the preparation of the Canadian inventory; values provided in the inventory were for the year 2008 because 2009 and 2010 data were not available. This should be updated once the 2010 NPRI data are available. The quality of the NPRI data is rated as unknown because the methodology used to estimate the amounts released by the individual reporting facilities is not known.
- There was no information available for mercury emission from soil for the Canadian inventory.

- Mercury emission factors for fireplaces, woodstoves, and wood-burning furnaces/boilers are based on limited studies. In addition, it is important to note that while the quantity of mercury present in discarded products is based on data, with the exception of fluorescent lamps, most of the assumptions regarding the fate of mercury are based on professional judgment, resulting in uncertainty about the actual quantities released to the atmosphere, water, and land. See Benazon Environmental Inc. (2011) for additional details.
- It is also difficult to estimate the impact of local reduction efforts because equipment that contains mercury is not inventoried. For example, a hazardous waste collection of 30 kg of mercury cannot necessarily be subtracted from the total amount of mercury known to be in the basin since that total amount is not known. When possible, the amounts captured by local reduction efforts are captured in Appendix B.
- Data from the 2008 National Emissions Inventory (NEI) were used for the U.S. 2010 estimates because 2010 data were not available.
- When NEI data were not available and the in-basin population was used for U.S. 2010 emission estimates, 2008 population estimates were used since the throughput, including cremation rates and vehicle miles traveled, were also 2008 data.
- The population estimate by the U.S. EPA in this report is more accurate than that of the previous Milestones report because a geographic information systems (GIS)-based analysis limited the analysis this year to only those portions of counties that were within the Lake Superior watershed.
- Very small sources of mercury have been removed from the inventory for simplicity. For example, the NEI estimated 0.0008 kg of mercury from a grocery store in Minnesota and 0.0001 kg from a sand and gravel pit in Michigan. In total, 154 minor sources represented <0.4 kg/yr of mercury.

With these caveats, Table 3-2 shows the mercury emissions and discharges from sources in the LSB while Table 3-3 shows the percent reduction.

Table 3-2. Mercury Releases to Air and Water from Sources in the Lake Superior Basin, kg/yr

Source	1990 (kg/yr)			2000 (kg/yr)			2005 (kg/yr)			2010 (kg/yr)		
	U.S. 1990	Canada 1990	Total 1990	U.S. 2000	Canada 2000	Total 2000	U.S. 2005	Canada 2005	Total 2005	U.S. 2010	Canada 2010	Total 2010
Mining/ Metals Production[1]	852.3	604.4	1456.7	338.3	4.5	342.8	303.2	26.2	329.4	257.1	3.7	260.8
Industrial	26.3	23.4	49.7	13.1	14.7	27.7	16.3	7.8	24.1	4.3	0.1	4.4
Products	213.8	31.4	245.2	6.7	1.8	8.5	4.8	1.6	6.4	3.5	1.0	4.5
Fuel Combustion	134.2	61.0	195.2	131.6	60.0	191.6	167.6	40.0	207.6	108.1	10.7	118.8
Incineration[2]	81.1	12.0	93.1	15.8	4.4	20.2	9.0	3.1	12.1	9.5	3.1	12.6
Waste Handling/ Landfills	38.8	27.5	66.3	10.0	5.0	15.0	8.1	5.0	13.1	6.7	5.0	11.7
Municipal/ Institutional	20.8	9.2	30.0	2.1	9.2	11.3	1.8	3.0	4.8	1.7	3.0	4.7
Total	1367.3	768.9	2136.2	517.6	99.5	617.1	510.7	86.7	597.4	390.9	26.6	417.4

[1] Includes iron sintering at Algoma Steel in Wawa, ON in 1990 and Mesabi Nugget in Hoyt Lakes, MN in 2010.
[2] Includes cremation.

Table 3-3. Percent Reduction of Mercury Releases from 1990 to 2010

Source	Reduction (%) 1990-2000	Reduction (%) 1990-2005	Reduction (%) 1990-2010
Mining/ Metals Production	76%	77%	82%
Industrial	44%	52%	91%
Products	97%	97%	98%
Fuel Combustion	2%	-6%1	39%
Incineration	78%	87%	86%
Waste Handling/ Landfills	77%	80%	82%
Municipal/ Institutional	62%	84%	84%
Total	71%	72%	80%
Stage 2 Reduction Goal	60%	70%	80%

[1] Emissions from the U.S. coal-fired utility sector increased in 2005.
[2] Although the LaMP Stage 2 did not have a mercury reduction goal for 2005, 70% is halfway between the 2000 and 2010 goals.

There have been reductions in discharge and emissions of mercury across all major sources from 1990 to 2010 (Figure 3-1). Large reductions in the mining sector (82%) are due to the closure of the White Pine copper smelter in White Pine, Michigan and the Algoma iron sintering plant in Wawa, Ontario. Industrial releases (primarily Canadian pulp and paper) have decreased by 91% since 1990. Product-related releases (i.e., incineration, products and waste handling/landfills) have clearly undergone significant reductions between 1990 and 2010. Municipal/institutional releases have been reduced by 84%. While the percentage reduction from fuel combustion seems low at 39%, it is in part a result of reductions due to installation of mercury emissions control technology rather than simply a reflection of decreased demand. This emissions control technology represents a significant improvement for the individual facilities that have invested in it.

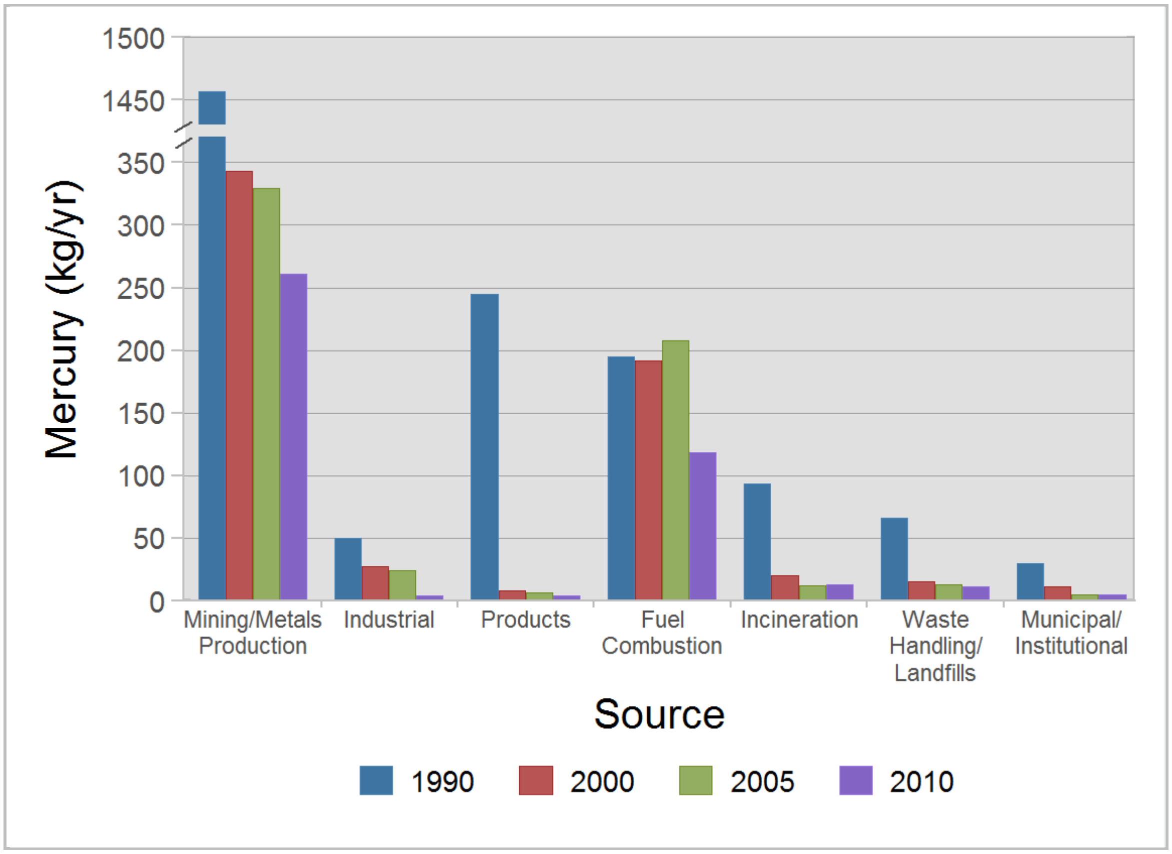
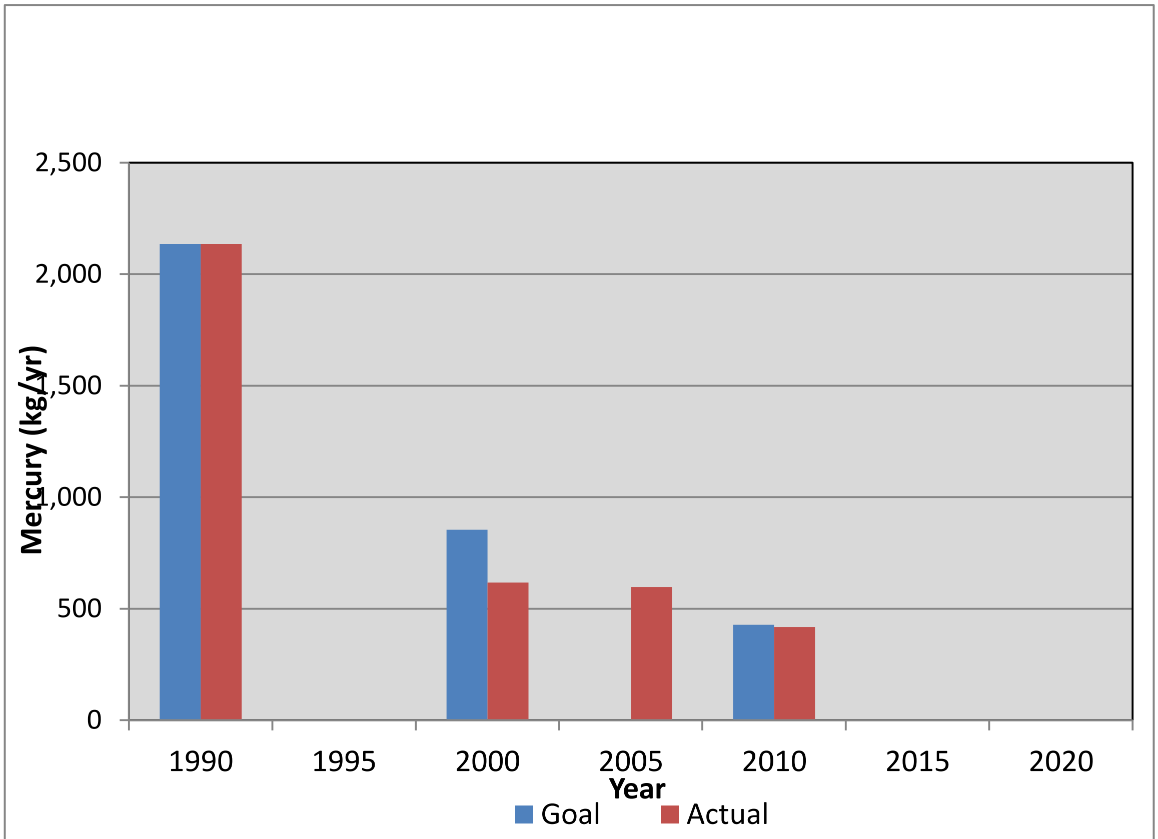


Figure 3-1. Reductions of Mercury Discharges and Emissions from Lake Superior Sectors between 1990 and 2010, kg/yr.

Figure 3-2 shows mercury releases since 1990 compared to the reduction schedule identified in the Stage 2 LaMP. (Note that there were no official Stage 2 LaMP goals for 1995 and 2005.) The trend shows a decrease in releases since 1990. Releases are at or very close to the milestone reduction targets. In 2010, the goal was 427 kg compared to the amount of discharges and emissions estimated for 2010 of 417 kg. An additional 204 kg/yr of mercury must be reduced from 2010 loads to meet the 2015 goal and 417 kg/yr must be reduced to reach the 2020 target of 100% reduction.



[1] No mercury reduction goals were set in LaMP Stage 2 for 1995, 2005, or 2015. No inventory was done for 1995. The goal for 2020 is zero discharge and zero emission.

Figure 3-2. Estimated Mercury Discharges and Emissions from Lake Superior Sources Between 1990 and 2010 Compared to the Stage 2 Load Reduction Goals[1], kg/yr.

The remaining mercury emission sources in the basin in 2010 are shown in Figure 3-3. Mining/metals production represents 63% of the mercury emissions for 2010, and of the 261 kg/yr from mining/metals production, a total of 257 kg/yr is attributed to taconite mining. The next largest source is fuel combustion (28%), which totaled 119 kg/yr, of which 86 kg/yr is from coal-fired utilities. Incineration (including cremation) and waste handling/landfills account for 3% each. Together, institutional/municipal products and industry account for 3% of emissions.

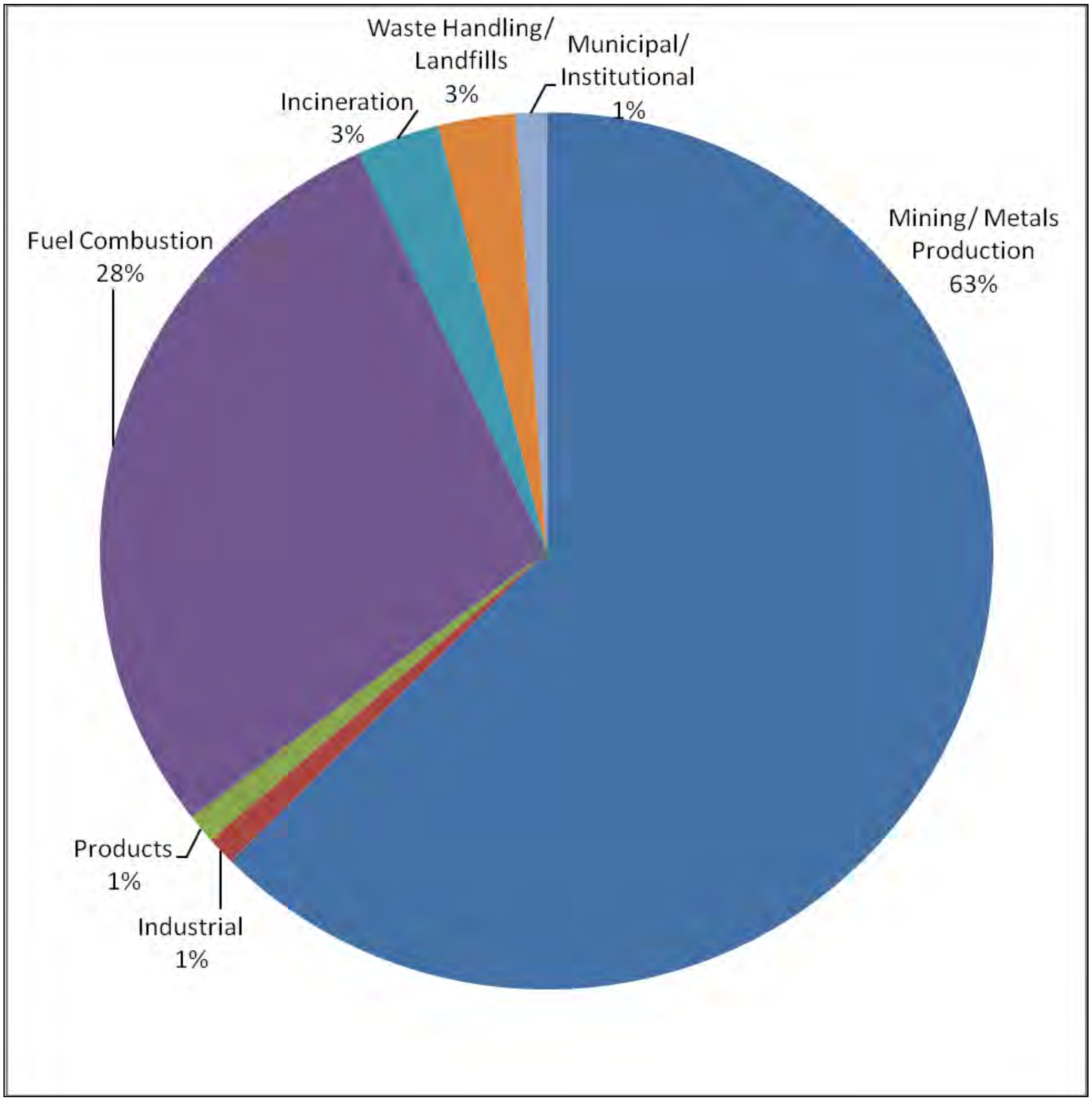


Figure 3-3. Percentage of Mercury Releases from Different Sectors in the Lake Superior Basin, 2010.

3.3.3 2020 Milestone Conclusions

It is possible to anticipate certain mercury reductions and increased releases/emissions from sources in the Lake Superior basin before reaching the 2015 and 2020 milestone years. New facilities or facility expansions may occur before 2015 and may increase the mercury loading.

Fuel Combustion

Figure 3-1 shows a gradual increase in mercury emissions from fuel combustion between 1990 and 2005, but a decline in 2010. This is due to a combination of reduced demand and the installation of mercury emission control equipment at the Presque Isle and Taconite Harbor coal-fired utilities. The utilities that operate these plants are considering whether they will continue to operate in the future. Specifically, the Minnesota Public Utilities Commission (MPUC) required Minnesota Power to conduct a baseload diversification study that looked at closing boilers at the Taconite Harbor and Syl Laskin coal-fired power plants in the Lake Superior watershed (MPUC, 2011). In addition, media reports indicate Wisconsin Energy (which operates the Presque Isle plant) is considering whether the plant might be shut down or converted to natural gas (WE Energies Blog, 2011). In Canada, the Long Term Energy Plan announced by the Government of Ontario in November 2010 stated that Thunder Bay Generating Station will convert from burning coal to burning natural gas, which will virtually eliminate mercury emissions from that site. It is expected that mercury emissions will remain at less than 10 kg/yr until coal combustion ceases in 2014.

According to Table 3-4, the potential reduction for these three largest coal-fired power plants in the basin ranges from <7 kg/yr with just Thunder Bay converting to natural gas, to about 40 kg/yr if all three facilities were either shut down or converted to natural gas. Some of the smaller utility coal-fired power plants are included in Table 3-4 for comparison purposes. Note that the 76 MW Bay Front plant was estimated to emit 13.8 kg of mercury in 2010, while the much larger 625 MW Presque Isle plant was estimated to emit 13.7 kg in 2010, which is much less than in 2005 before the Toxecon® mercury control technology was in place. These smaller plants are becoming more significant as the mercury inventory shrinks and the larger plants improve their pollution control equipment.

Table 3-4. Mercury Emissions from Six[1] Coal-fired Power Plants in the Lake Superior Watershed, 1990-2010

Name	1990 (kg Hg)	2000 (kg Hg)	2005 (kg Hg)	2010 (kg Hg)
Taconite Harbor, MN	0.5	22.6	25.8	28.9
Bay Front, WI	1.5	1.5	13.9	13.8
Presque Isle, MI	68.0	41.7	55.5	13.7
Laskin, MN	2.6	8.1	9.5	7.5
Thunder Bay, ON	57	56	37	7
Hibbard, MN	2.1	0.7	2.7	7.2
Total Mercury, all sectors	2136.6	620.9	635.3	417.4

[1] These six have the highest mercury emissions from coal. There are other coal-fired boilers in the watershed that release less mercury and others that burn a mixture of coal and other fuels.

Although further reductions from coal-fired power plants can be expected, energy agencies also project that energy consumption overall will decrease. For example, trends tracked by the U.S. Energy Information Administration (EIA) suggest that per capita energy use will drop through

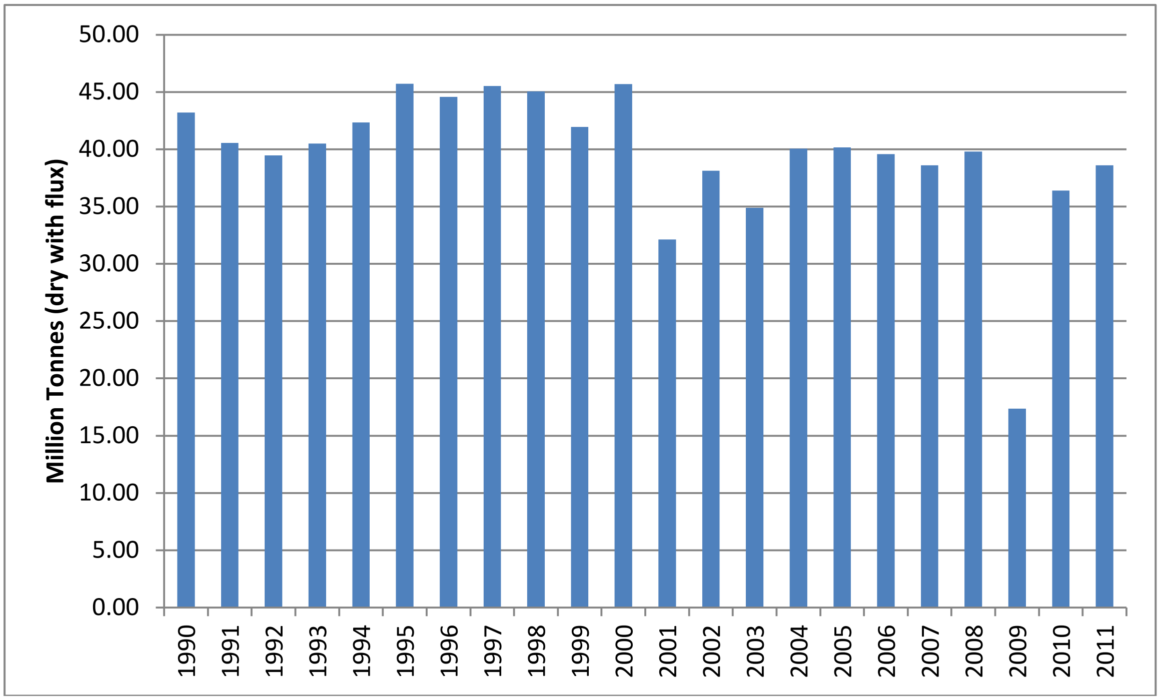
2035 (EIA, 2011). This is coupled with a decline in basin population (i.e., the estimated Canadian population dropped from 247,926 [1990] to 227,108 [2010] while the estimated U.S. population increased from 423,204 to 433,860 in the same period). However, the overall decrease in population may not cause a decrease in emissions since the power plants can sell their excess energy on the grid.

Mining and Metals Production

A variety of projects, including taconite mining, nonferrous mining, scram mining, and a possible refinery expansion have been considered within or near the Lake Superior watershed, although discharge or emission estimates are not possible at this time and some of these new sources may have low mercury emissions.

Sources identified in every milestone inventory have the potential to fluctuate. Because taconite production is such a large source of mercury, and because emissions generally track production, it is important to acknowledge variability in this source (Figure 3-4). For example, the milestone year of 2000 was over 13 M tonnes higher than in 2001. If the 2000 production had been closer to the 2001 production, then the 2005 and 2010 milestones would have shown an increase in mercury emissions from taconite rather than a decrease. Variability in large sources therefore plays an important role in describing progress towards the goal of zero discharge and zero emission.

The status of mercury reductions in the taconite industry is difficult to project at this point. Studies on mercury cycling in taconite plants are ongoing. Taconite plants are currently running at or near capacity and analysts believe demand for taconite pellets, nuggets, and concentrate will remain strong. However, in 2008-2010, the taconite industry was hit by the overall economic recession and plants were idled temporarily in 2009. Production picked up and by 2010 and 2011, levels were only slightly lower than pre-recession production.



Sources: 1990-2009 data from Minnesota Department of Revenue, 2010; 2010 data from Wagstrom, 2011; rough estimate from Myers, 2011

Figure 3-4. Taconite Production in Minnesota, 1990-2011.

For this reason, the overall amount of mercury emitted per tonne of pellets produced may better describe the overall progress made by this industry (Table 3-5). These data suggest some improvement in the rate of mercury emitted from taconite production since 1990. This is probably due to installation of pollution control equipment at the Keewatin Taconite plant in 2005, which dropped mercury emissions at that facility by 28%. The increased ratio in 2005 is due to a higher proportion of pellets from plants with higher emission factors.

Table 3-5. Ratio of Kilograms of Mercury Emitted to Million Tonnes of Taconite Pellets Produced in Minnesota during LaMP Milestone Years

Year	Mercury Emissions (kg)	Pellets (M tonnes)	Ratio
1990	323.9	43.20	7.5
2000	341.8	45.68	7.5
2005	338.1	40.17	8.4
2010	250.4	36.39	6.9

Other Sources and Pathways

Contaminated Sediments. In Canada's portion of the LSB, significant quantities of mercury (~880 kg) are present in contaminated sediments. These are located within two AOCs (Jellicoe Cove, and Thunder Bay Harbour), which are scheduled to be remediated prior to 2020. Also, Santiago (2010) has indicated that limited sediment sampling at Black Bird Creek and Lake "C" has shown possible low level mercury contamination. Additional sediment sampling is underway to further characterize the extent of contamination.

Recycling and Disposal. Large quantities of mercury (2,900 to 5,800 kg) are also present in sludge disposed in drums contained within reinforced concrete vaults at a waste disposal site in Marathon, Ontario. The site is being monitored; however, groundwater sampling results indicate that no mercury is leaching from the waste site.

In addition, many industries in Canadian North Shore towns recycle fluorescent lights. Concern has been raised that preparation of lamps for recycling may result in significant emissions of mercury. For example, the use of a drum top crusher to crush used fluorescent lamps before they are sent for recycling may be a preventable source of mercury emissions. Further investigation is required to identify potential sources of mercury emissions once used fluorescent lamps are collected, stored and transported for recycling.

Cremation. Mercury emissions from cremation have increased due to increased cremation activity and quantity of amalgam in the teeth of deceased. Increases of mercury emissions from cremation are expected to continue over the next 15 years (MPCA, 2008) followed by a gradual decline as less amalgam will be present in the future generations.

Mercury-containing Products:

- Thermometers: There are bans in sale and use of mercury-containing fever thermometers, and mercury-free alternative thermometers are replacing them (Interstate Mercury Education and Reduction Clearinghouse [IMERC], 2008a, Cain, 2005), resulting in corresponding decline in mercury emissions (to ~2015).
- Thermostats: Various states also have restrictions on mercury-containing thermostats, resulting in many companies ceasing manufacture or sale, resulting in an expected gradual decline in emissions over the next decade (to ~2020).
- Switches and Relays: Legislative restrictions of certain mercury-added switches and availability of new non-mercury technologies have contributed to the decline in mercury in switches and relays in 2007 compared to 2004 (IMERC, 2008b), resulting in many companies ceasing manufacture or sale of these products across the U.S. However, because of the long life-spans of switches and relays, a large quantity of mercury is expected to remain in the public realm. North American automakers voluntarily phased out the use of mercury in switches in new motor vehicles at the end of 2002 (Michigan Mercury Switch Study, 2002) and associated mercury emissions are expected to decrease over the next 10 years (to 2020). Voluntary discontinuation of mercury switches by Canadian appliance manufacturers occurred in 1999/2000, but because these products

have long service life, mercury will continue to be found in discarded appliances for the next 20 to 30 years (Association of Municipal Recyclers [AMRC], 2004).

- Measurement and Control Devices: Sales of new measurement and control devices containing mercury have declined considerably in recent years in the U.S. (Wienert, 2009), likely due to product restrictions.
- Button-Cell Batteries: After 2011, mercury emissions from button-cell batteries are expected to decline considerably when battery manufacturers voluntarily produce mercury-free alternatives (from 2010 to 2015). However, embedded mercury button-cell batteries in products manufactured off-shore will likely continue to be consumed in the U.S. and Canada for some time to come.

Special Materials Program. The Canadian Municipal Hazardous and Special Waste (MHSW) Program is designed to collect consumer hazardous and special materials so they can be recycled or disposed of safely. The first phase began in July 2008 and includes nine materials. The second (consolidated) phase began July 1, 2010 and includes 22 materials (including the original nine).The program is expected to substantially increase the quantity of mercury-containing products recycled in the LSB.

As older mercury-containing products are discarded and replaced with non-mercury devices, it is expected that emissions from this source will continue to decline accordingly. Despite the restriction, bans, voluntary phase-out and recycling/waste management activities, some mercury-containing products will still be found in use, storage, or disposal past the 2020 ZDDP target.

3.4 Dioxin

In this inventory, the term dioxins and furans refers to two groups of chemical compounds: the polychlorinated dibenzo-*p*-dioxins and the polychlorinated dibenzofurans. These chemicals are not created intentionally, but can be generated by sub-optimal combustion conditions and incomplete combustion processes. Because of their hydrophobic nature and resistance toward metabolism, dioxins and furans persist and bioaccumulate in the fatty tissues of animals and humans. There are numerous individual compounds, or congeners, associated with each of these chemical groups that exhibit varying degrees of similar "dioxin-like" toxicity. The most widely studied and highly toxic compound is 2,3,7,8-TCDD. Dioxins and furans are usually quantified in terms of total toxicity relative to TCDD expressed as a TEQ, in which a series of toxic equivalency factors (TEFs) are assigned to each of the dioxin-like compounds in a mixture to obtain the relative toxicity with respect to TCDD. Therefore, the TEQ is the amount of TCDD needed to equal the combined toxic effect of all dioxins and furans found in the mixture.

Different TEF schemes have been used to calculate TEQ, including:

- I-TEQ, a scheme adopted by the U.S. EPA in 1989;
- $TEQ\text{-}WHO_{94}$, adopted by the World Health Organization (WHO) in 1994;
- $TEQ\text{-}WHO_{98}$, adopted by WHO in 1998 as an update to the previous one; and
- $TEQ\text{-}WHO_{05}$, adopted by WHO in 2005 as an update to the previous one.

To be consistent with the dioxins and furans inventory prepared by the U.S. EPA (U.S. EPA, 2005), emissions of dioxins and furans are generally reported in g TEQ-WHO98/yr wherever possible. In some instances, however, only I-TEQ data are presented because the TEQ-WHO_{98} equivalents are not available. Yet in other cases (burn barrels and yard waste), dioxin and furan data are available in g TEQ-WHO_{05}/yr. See Appendix C for more details on units used. For example, an emission of 4 g dioxin in a year is reported using the different TEFs as follows:

- 4 g I-TEQ/yr
- 4 gTEQ-WHO_{94}/yr
- 4 gTEQ-WHO98/yr
- 4 gTEQ-WHO_{05}/yr

An examination of the TEF assigned to each congener within the I-TEQ, TEQ-WHO_{98} , and TEQ-WHO_{05} calculation methodologies show that the difference between the three methodologies is generally small and would likely result in similar estimates. It is unclear which would result in a higher estimate as it depends on the concentrations of the individual congeners, which vary by source.

3.4.1 Dioxin Reduction Goals

The reduction goals for dioxin, HCB, and OCS described in LaMP Stage 2 include the following (1990 baseline):

- No formal dioxin milestone for 2000
- 80% reduction by 2005
- No formal dioxin milestone for 2010
- 90% reduction by 2015
- 100% reduction by 2020

In order to meet the 90% reduction goal by 2015, an additional 1.03 TEQ/yr of dioxin must be reduced from 2010 loads. In Section 3.4.2 below, it is estimated that an 86% reduction of dioxin emissions and discharges has taken place since 1990. Due to the similar methods of formation of dioxin, HCB, and OCS, the lack of data concerning discharges and emissions of OCS, and the incompleteness of the HCB inventory, changes in the dioxin inventory will serve as a surrogate for both HCB and OCS inventories. Additional HCB discharge and emission information is presented in Section 3.7, but the percent reduction has not been estimated due to the incomplete inventory.

3.4.2 Sources of Dioxin

The dioxin inventory, listed in Table 3-6, includes releases to air and water for the baseline year, the year 2000, the milestone year of 2005 and the year 2010. It should be noted that similar to mercury, discharges to water are only a small portion of the releases inventoried in Table 3-6.

For example, in both the U.S. and Canada the amount of dioxin discharged to water and soil was <1% of the total releases to air, water, and soil in 2010 (see Appendix C).

Some important changes have been made since the first version of the inventory tables in LaMP 2000. The inventory has been adjusted downward mainly because the U.S. incineration numbers in LaMP 2000 reflected a different unit (g total polychlorinated dibenzo-p-dioxin [PCDD]/ polychlorinated dibenzofuran [PCDF]), although incineration is still the single largest category in the revised inventory for 2010. Also, coal-fired power plants were identified as the largest point sources of dioxin in 2005. However, based on revised data submitted under NPRI and discussions with representatives of the facility, it appears that previous dioxin emission estimates were incorrectly calculated by the Thunder Bay Generating Station (Todd, 2010). Current emissions from this source are documented as 0 g TEQ/yr in the NPRI database. Once these corrections are made, the pattern of incineration being the largest source and fuel combustion being the second largest source is the case for 2000, 2005, and 2010. The 1990 baseline year was dominated by an iron sintering plant, which was shut down before the 2000 milestone year.

While the inventories have improved, uncertainties and limitations still must be noted:

- In Canada, there is considerable uncertainty associated with the emissions estimate from landfill fires and burn barrels due to the lack of accurate data. The frequency and extent of landfill fires in the past is unknown and there is some uncertainty about the percentage of waste burned in rural areas. If more than 5% of annual rural waste is burned in landfill fires, emissions from landfill fires could be an important source of dioxins and furans. Additional information on the quantity of garbage burned is required along with appropriate emission factors.
- No releases were estimated from sediments on the Canadian side of the LSB. Low concentrations of dioxins and furans have been found to be widely dispersed within the watershed in those locations, with higher levels present in Thunder Bay, Peninsula Harbour and at the mouth of the Magpie River, as a result of industrial activity. Limited sediment sampling at Black Bird Creek and Lake "C" has shown contamination with dioxins and furans (Santiago, 2010). Elevated dioxin levels have also been found in sediments of Crawford Creek and floodplain soils within the St. Louis River AOC, below a former wood treatment facility in Superior, Wisconsin. Additional sediment sampling is underway to further characterize the extent of contamination. Insufficient information exists to estimate quantities.
- Due to reporting cycles, neither 2010 NPRI nor NEI data were available so values provided in the inventory were for year 2008. An update is desirable once the 2010 NPRI data are available.
- Information is needed on the extent to which land clearing and brush burning operations exist in the LSB. In the U.S., Lake Superior and national inventories, land clearing activities have a high uncertainty compared to controlled sources. Until the method and LSB-specific activity data improve, land clearing estimates will not be added to the inventory.
- There are potentially significant sources of dioxin emissions which have not been captured in the inventory. For example, the inventory does not include dioxin from

wildfires which may have human or natural origins. Additional information on the use of outdoor wood furnaces in the LSB is required.

- In some categories, no evidence was found of changes in practices between the one milestone year and the next so the estimate stayed the same.
- The U.S. and Canada used different methods in estimating dioxin emissions from open burning of trash. The Canadian method assumes that: (1) 11% of the total population in northwestern Ontario burn garbage, and (2) 60% of residential waste generated by households was burned in burn barrels. The U.S. inventory assumes that: (1) 19% of basin residents in Michigan and Wisconsin and 16.8% in Minnesota burn garbage, and (2) 43% of waste generated by rural households was burned.
- The difference between states' burning rate is due to a 2010 Minnesota statewide survey which documents a decrease in the rate of burning in northeastern Minnesota since 2006. Since a LaMP open burning abatement project had been carried out in northeast Minnesota in 2006 and 2007, but not elsewhere, it was felt that the new Minnesota rate should not be applied in Michigan and Wisconsin.
- The U.S. NEI (U.S. EPA, 2008) was used as a last resort for some categories since methodologies and reporting may not be nationally consistent.

Table 3-6 shows the current estimates of dioxin discharges and emissions from sources in the LSB. Table 3-7 shows the estimated percent reduction over time.

The largest single reduction was due to the closing of Algoma Steel's iron sintering plant in Wawa, Ontario in 1998, which alone was responsible for about 66% of the dioxin emission reduction between 1990 and 2010. In 2010, the largest source of dioxin is incineration, and most of the incineration emissions are from U.S. unpermitted burning of trash via backyard burning, landfill fires, and small incinerators at businesses. At roughly half the emissions from incineration, fuel combustion was the next largest source in 2010. Perhaps the most striking finding is that there has been little progress in these two largest sources between 2005 and 2010. The percent reductions for both sources are virtually unchanged and their relative size means that the overall percent reduction between 2005 and 2010 is also unchanged.

Figure 3-5 shows dioxin releases since 1990 compared to the reduction schedule identified in the Stage 2 LaMP. (Note that there were no Stage 2 LaMP goals for 1995, 2000, and 2010. The goal of 85% reduction for 2010, which was half-way between the 2005 and 2015 goals, was extrapolated). The trend shows a decrease in releases since 1990. The 2010 estimated releases are just below the milestone reduction target. To reach the 2015 target of 90% reduction, an additional 4% reduction from 1990 levels is needed. Regardless of the TEF schema used to calculate, this equates to an additional reduction of dioxin of about 1 g TEQ/yr. However, to reach the ultimate 2020 target of 100% reduction, an additional 14% reduction from 1990 levels is required. This equates to an additional reduction of dioxin of about 4 g TEQ/yr from 2010.

Figure 3-6 shows the distribution of sources from various sectors remaining in the LSB in 2010. Incineration accounts for 65% of the dioxin emissions for 2010. This includes burn barrel emissions, landfill fires, small incinerators, and Canadian (but not U.S.) open burning. Fuel combustion accounts for 32% of emissions. The remaining percentage is from commercial byproduct and industrial releases. Municipal/institutional sectors did not contribute to dioxin releases in 2010.

3.4.3 2020 Milestone Conclusions

There was not a LaMP Stage 2 dioxin goal for 2010. However, a goal of 85% was extrapolated, which was halfway between the 2005 (80%) and 2015 (90%) goals. Of the individual sources that currently contribute dioxin emissions, only two – industrial releases and municipal/institutional releases – were successful in achieving the theoretical 85% reduction target; however, these two sources contribute less than 2% of the current emissions total. Additional effort is required to reduce emissions from other sources.

Table 3-6. Dioxin Releases to Air and Water from Sources in the Lake Superior Basin, g TEQ/yr

Source	1990 (g TEQ/yr)			2000 (g TEQ/yr)			2005 (g TEQ/yr)			2010 (g TEQ/yr)		
	U.S. 1990	Canada 1990	Total 1990	U.S. 2000	Canada 2000	Total 2000	U.S. 2005	Canada 2005	Total 2005	U.S. 2010	Canada 2010	Total 2010
Iron Sintering	NA	19.40	19.40	NA	0.00	0.00	NA	0.00	0.00	NA	0.00	0.00
Incineration	6.43	0.80	7.23	3.47	0.66	4.13	2.35	0.29	2.65	2.29	0.30	2.59
Fuel Combustion	1.52	0.25	1.77	0.82	0.22	1.04	1.02	0.23	1.25	1.05	0.19	1.25
Industrial	0.01	0.72	0.73	0.01	0.03	0.04	0.01	0.04	0.05	0.00	0.03	0.03
Commercial By-Product	0.04	0.10	0.14	0.04	0.07	0.11	0.02	0.08	0.10	0.02	0.08	0.10
Municipal/ Institutional	0.0004	0.05	0.0504	0.0004	0.05	0.0504	0.0045	0.00	0.0045	0.0001	0.00	0.0001
Total*	**7.99**	**21.33**	**29.31**	**4.34**	**1.04**	**5.38**	**3.40**	**0.65**	**4.05**	**3.36**	**0.60**	**3.96**

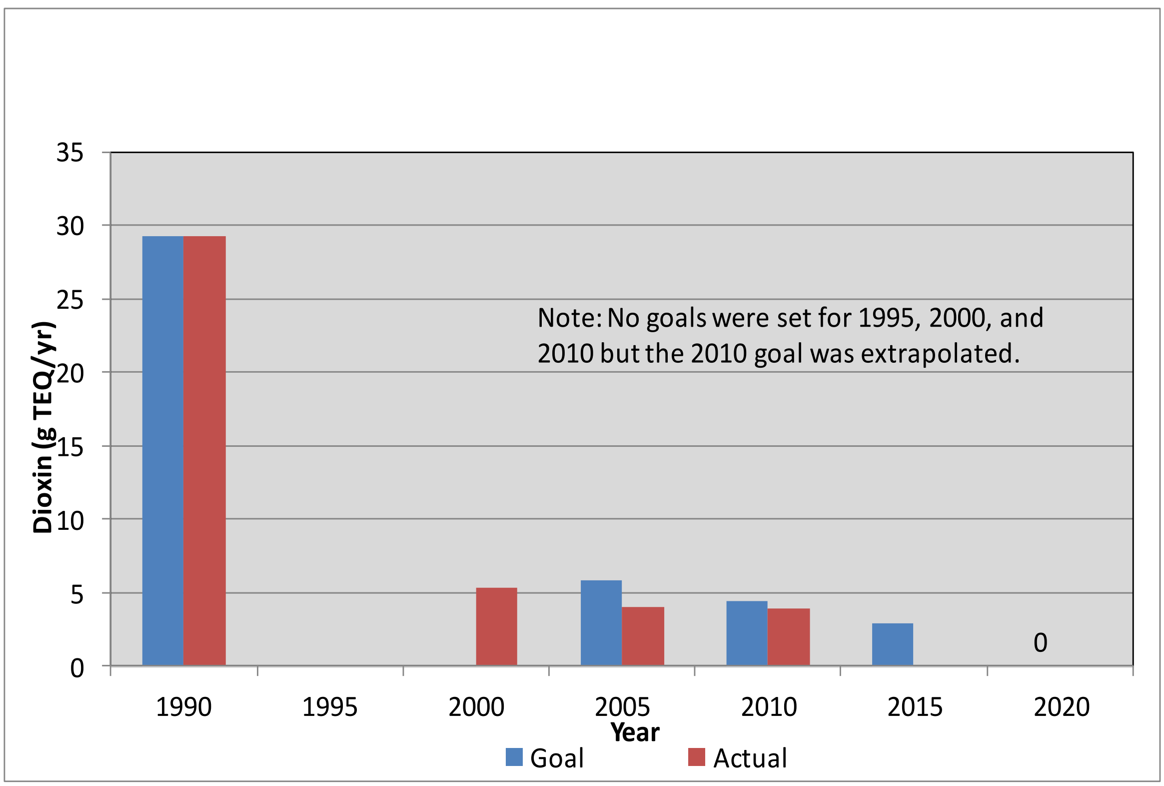
NA = Not Applicable; The Algoma Steel Plant was located in Wawa, Ontario (Canada).

* Totals are estimates only. It is recognized (and discussed in text) that U.S. and Canadian values were generated using different TEFs as indicated by the TEQ. Different dioxin units are used in different databases, sometimes even within categories. For the purposes of tracking percent reduction over time, these unit differences do not influence the analysis. For the U.S., generally facility emission estimates are $TEQ\text{-}WHO_{98}$ and mobile and area source emissions use $TEQ\text{-}WHO_{05}$. For Canada, generally coal and wood combustion, industrial emissions, incinerators, cremation, and emissions from products are reported as I-TEQ, petroleum combustion and landfill fires are reported as $TEQ\text{-}WHO_{98}$, and backyard trash and yard waste burning are reported as $TEQ\text{-}WHO_{05}$. More information on TEQs used is provided in Appendix C.

Table 3-7. Percent Reduction of Dioxin Releases from 1990 to 2010

Source	Reduction (%) 1990-2000	Reduction (%) 1990-2005	Reduction (%) 1990-2010
Iron Sintering	100	100	100
Incineration	43	63	64
Fuel Combustion	41	29	29
Industrial	94	93	96
Commercial By-Product	21	28	31
Municipal/ Institutional	0	91	100[1]
Total	**82**	**86**	**86**
Stage 2 Reduction Goal	**na**	**80**	**85[2]**

[1] A small amount of dioxin discharge was estimated for 2010, so technically, the reduction is 99.8%.
[2] Although the LaMP Stage 2 did not have a 2010 dioxin reduction goal, 85% is halfway between the 2005 and 2015 goals.



Total values are expressed g TEQ/yr. These values are estimates generated summing data with I-TEF and TEQ-WHO_x.

Figure 3-5. Estimated Reductions of Dioxin Releases to Air and Water from Lake Superior Sources between 1990 and 2010 Compared to Stage 2 Load Reduction Goals, g TEQ/yr.

Overall, the extrapolated 2010 goal of 85% was barely achieved, with an overall reduction of 86%. This goal was met due to the elimination of the iron sintering sector from the inventory (closure of the Algoma Steel plant). In order to meet the 90% reduction goal in 2015, the remaining in-basin sources must be reduced by an additional 1.03 g TEQ/yr of dioxin. U.S. and Canadian unpermitted burning contributed 2.59 g TEQ to 2010 dioxin releases, and as a preventable source of dioxin, elimination of unpermitted burning by 2015 should be targeted. After incineration, mobile sources were the largest emitters of dioxin in 2010 (i.e., 0.71 g TEQ for gasoline and diesel, on-road and non-road sources). Several national initiatives to improve emissions from cars, trucks, and ships have the potential to decrease dioxin emissions. The possible closure or conversion to natural gas of the largest coal-fired power plants in the basin as discussed in Section 3.3.3 would lead to decreases of dioxin emissions in the range of 0.01 to 0.03 g TEQ.

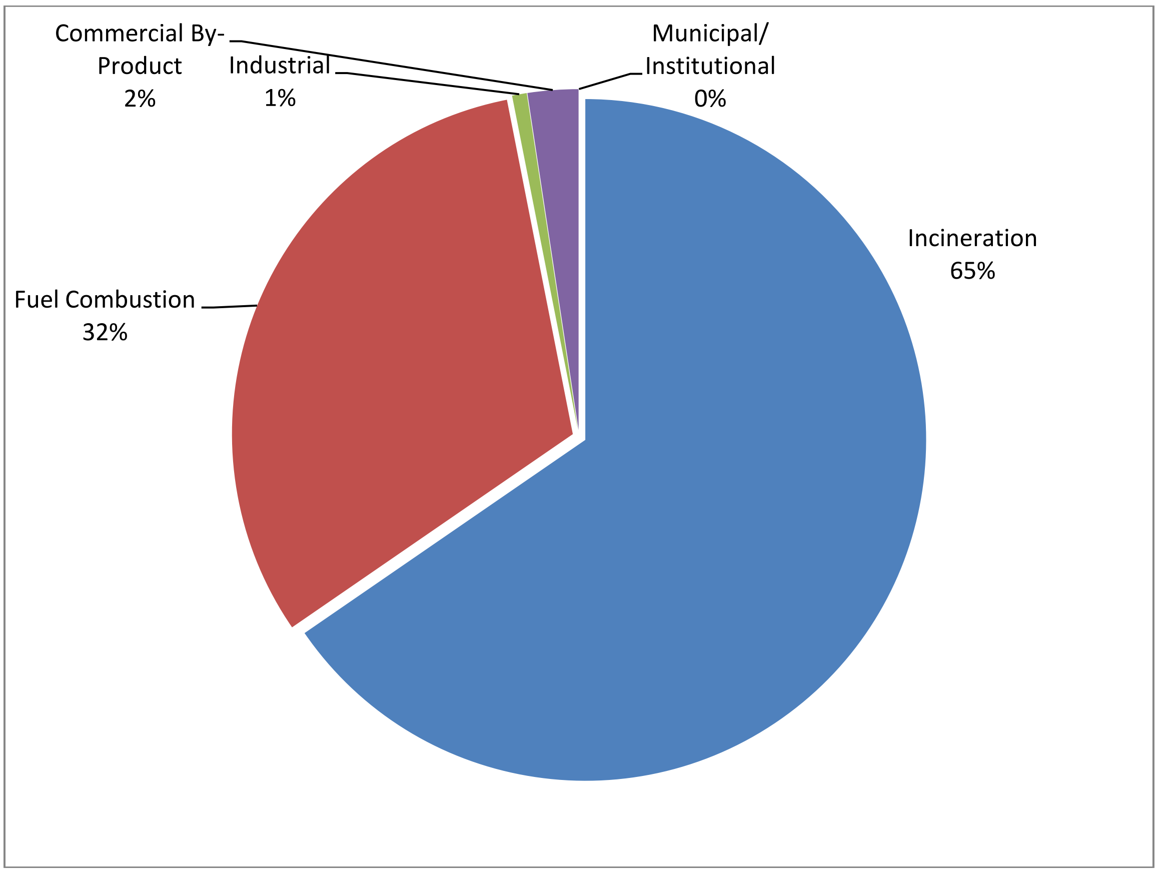


Figure 3-6. Percentage of Dioxin Releases from Different Sectors in the Lake Superior Basin, 2010.

3.5 PCBs

The original intent of the Stage 2 PCB reduction schedule was to inventory all the PCB equipment still in service or use, track its disposal and calculate the grams of PCBs reduced from the concentration of PCBs and the weight or volume of materials disposed. With this information, and respective calculations, it would be possible to estimate the percent reduction over time. As noted in Chapter 4 of LaMP 2006 (LSBP, 2006), "The PCB inventory has been a challenge as there is no comprehensive and up-to-date inventory." Furthermore, no current inventory - U.S. or Canadian - tracks various smaller pieces of equipment that were exempt from provisions in U.S. and Canadian statutes and regulations or had reduced regulatory requirements, yet still contain PCBs many times at high concentrations (e.g., capacitors, insulators, etc.). Finally, through data collected over the period since the initiation of the Toxic Substances Control Act (TSCA), it is not clear that the initial baselines utilized for current partial inventories account for the actual amount of PCB equipment or PCBs present in U.S. commerce. Without the availability of a complete PCB inventory, and the lack of complete information in the currently available inventories, it is extremely difficult to have a full picture of PCB reductions over time. However, certain pieces of knowledge on the existence of PCB equipment may be ascertained through the data currently available.

To date, proposals for improving the inventory of PCB equipment have not been approved by potential funders, who prefer a more proactive approach to PCB reduction directed toward known sources of PCBs. In the absence of a complete inventory, the LSB Work Group has considered what information is actually available for determining priorities and measuring progress. This information includes: the current PCB management approaches that are used by the LSB jurisdictions (<u>see</u> Appendix D), the information that is available on quantities of PCB materials in existence or removed for disposal or storage, and the need for consistency with the Great Lakes toxics strategies.

Table 3-8 shows the amount of PCB materials decommissioned and put into storage in Ontario from facilities in the basin for selected years. The Ontario inventory covers large equipment that is ≥50 ppm PCBs, but not small equipment or equipment <50 ppm PCBs. Therefore, small high concentration PCB equipment is not counted. Even with these caveats, it is clear that there have been substantial reductions in the quantities stored at provincial sites in the LSB over the last 20 years. Significant quantities of PCB-containing equipment and PCB material have been moved out of storage for disposal since some of the categories are down to zero PCBs in storage in 2006 and other categories have dwindled to low amounts.

Table 3-8. Summary of PCB Waste in Storage at Province of Ontario-Monitored Sites in LSB 1990-2010

Type	**Quantity of PCB Waste in Storage (assorted units)**				
Year	**1990**	**1995**	**1997**	**2006**	**2010**
HL Liquid (L)	85,112	163,217	128,001	16,389	25,814
LL Liquid (L)	61,268	41,528	20,336	11,144	6,066
HL Solid (tonne)	146	114	69	5.0	1.8
LL Solid (tonne)	136	144	128	1.4	9.8
Misc.[1]	2,576	1,158	977	975	No Data

HL = High Level (>500 ppm); LL = Low Level (<500 ppm)

[1] Miscellaneous includes PCB-contaminated pallets (kg), transformer carcasses (kg), empty drums (no. of units), and unidentified waste (kg).

Environment Canada provided an estimate of the quantity of High Level (HL) liquid in use, totaling 93,528 L, and that 3,695 kg of Low Level (LL) solids and 144 L of LL liquids were sent for destruction.

It is expected that PCB waste will continue to drop on the Canadian side of the LSB in response to the requirements of the 2008 PCB regulations (Environment Canada, 2008) which call for:

- The phase-out of all HL PCBs (over 500 ppm PCBs) and PCBs over 50 ppm in sensitive areas that are currently in use by December 31, 2009.
- The phase out of all equipment between 50 and 500 ppm PCBs that are not in sensitive areas and the phase out of pole top (contaminated mineral oil) transformers and PCB light ballasts by December 31, 2025.
- The destruction of all PCBs that were stored on September 5, 2008 no later than December 31, 2011.
- The phase out of all PCB storage sites at sensitive locations by September 5, 2009.

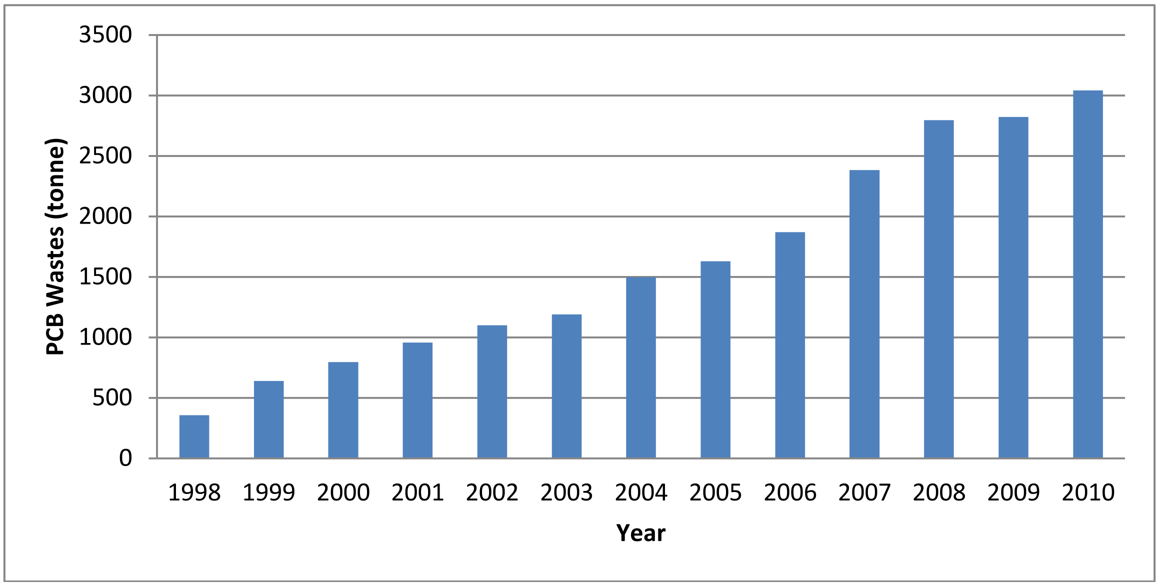
For the U.S. side of the basin, Table 3-9 shows the amount of PCB materials that have been disposed of from facilities in Minnesota. Unlike other states within the Basin, Minnesota performs TSCA compliance monitoring and enforcement actions for U.S. EPA; additionally, Minnesota considers PCBs in concentrations $\geq$50 ppm to be a hazardous waste under the Resource Conservation and Recovery Act (RCRA) Subtitle C program, making PCBs a jointly regulated waste under RCRA and TSCA. However, Michigan and Wisconsin do not administer TSCA use programs, nor do they have a state statute equivalent to TSCA. As a result, Minnesota is the only state where it is possible to track disposal of PCBs from facilities located within the LSB. Despite these major distinctions, for simplicity, it is assumed that the actual disposal amounts for large PCB equipment $\geq$50 ppm PCBs among the three states will be similar as TSCA applies to all of those cases; however, the same may not be true for PCB equipment from sources not tracked explicitly by or regulated under TSCA (<50 ppm PCB equipment, small equipment, etc.), where only Minnesota requires additional waste management actions. In addition, the pathway for disposing of PCB materials will differ since Michigan and Wisconsin generators can dispose of many $\geq$50 ppm PCB waste streams as solid waste while those same wastes are classified as hazardous waste in Minnesota.

Table 3-9. Summary of All PCB Waste Disposed from Minnesota Lake Superior Facilities, 1998 – 2010 (tonnes/yr)

Type	1998	1999	2000	2001	2002	2003	2004[1]	2005	2006	2007	2008	2009	2010
PCB Waste	356	283	157	160	143	89	2M	133	242	512	414	26	220

[1]The high quantity for 2004 was due to an incident at a recycling facility where a PCB containing item passed through a shredder and resulted in a large amount of contamination (though at low concentrations).

Despite the reporting limitations, over time, the total amount of PCB materials will decrease since the amount of PCB equipment is finite and most PCB equipment is nearing the end of its projected life. The cumulative total PCB waste has been graphed in Figure 3-7, which shows that the rate of total PCB wastes has not flattened over the years. It is likely that large amounts of equipment in both the U.S. and Canada are approaching the end of their useful service, which will result in further increases in disposal, followed by the anticipated flattening in the cumulative disposal curve.



[1] Due to an incident at a recycling facility in 2004 where a PCB containing item that passed through a shredder contaminated a large amount of low level contaminated material, the material associated with that incident has been removed for graphing purposes. The facility in question was assumed to have disposed of 500,000 pounds (~227,000 kg) of PCB waste instead. In other years, the facility reported 370,000 to 1,000,000 pounds (170,000 to 460,000) but in 2004, it was 4,412,976,000 pounds (~2,000,000,000 kg).

Figure 3-7. Cumulative Total of All PCB Wastes Disposed from Minnesota Facilities in the Lake Superior Basin, 1998 – 2010 (tonnes/yr)[1]

An attempt was made to further analyze trends in different types of PCB materials, including the three categories below. Except for ballasts as noted below, reports of "PCB wastes" were not added to either the low or high level wastes.

- LL waste, which was primarily PCB oil >50 ppm plus solvent rinsate;
- HL waste, which was primarily PCB oil and transformers >500 ppm;
- Ballasts, which include records that specify ballasts were disposed, as well as records of "PCB waste" from schools, medical facilities and appliance and electrical businesses (due to the high probability that the "PCB waste" was actually ballasts).

Table 3-10 shows the amounts of HL waste, LL waste, and ballasts collected since 1998. There may be a leveling off for the LL and HL wastes, although this may simply reflect recent changes in reporting practices (e.g., reporting as "PCB waste"). For ballasts, Figure 3-8 suggests there is no sign of leveling off.

Table 3-10. Low Level PCB, High Level PCB, and Ballasts Disposed from Minnesota LSB Facilities, 1998 – 2010 (tonnes/yr)

Waste	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
Low Level	128	113	87	58	76	0	1	0	13	13	0	0	0
High Level	27	13	0	64	2	2	3	8	0	0	0	0	0
Ballasts	4	4	3	4	3	3	3	4	7	7	7	4	7

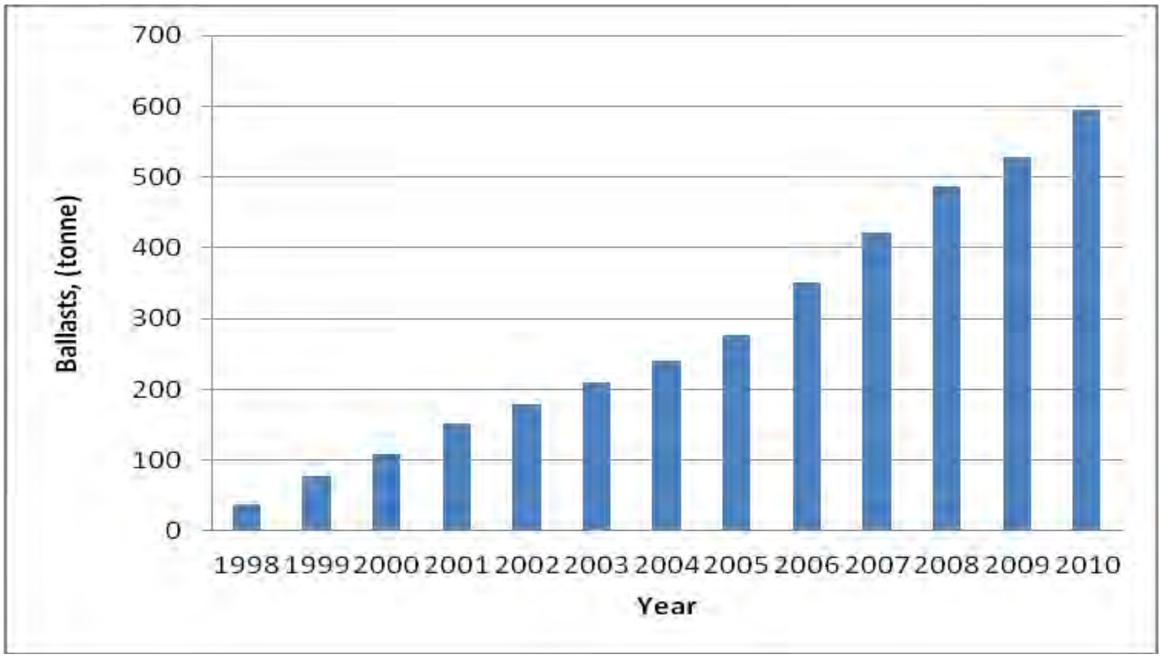


Figure 3-8. Ballasts Disposed from Facilities in the Minnesota Lake Superior Basin, 1998 – 2010 (tonnes/yr).

The ballast trend is of particular interest since it shows that ballasts are still being disposed of in large numbers. Ballasts are still in use in fluorescent lamps in schools, colleges, health care facilities, and workplaces. For example, the Minnesota records show that in 2010, two health care facilities disposed of over 400 kg of PCB wastes, two colleges disposed of 3,000 kg, and five school districts disposed of 27 kg of ballasts or "PCB waste" presumed to be ballasts. There are documented cases of leaking ballasts in U.S. classrooms; exposure to children and pregnant females is a concern.

The Chemical Committee proposes the following alternative method for tracking the Lake Superior PCB inventory and establishing a means of measuring progress:

1. Track disposal and storage via the Ontario database for PCB storage, the Environment Canada database for PCB disposal and the Minnesota hazardous waste database for PCB

disposal. In preparing this report, the Ontario and Environment Canada databases have been reviewed and found to be slightly different.

2. Examine the storage and disposal category trends every 5 years (e.g., the weight of HL capacitors stored in Ontario or the weight of PCB oil in Minnesota). Produce figures showing the cumulative total for various categories and the total weight of materials removed or stored. Figures 3-7 and 3-8 are examples of this.[3]
3. In Canada, show how much of the stored PCBs are destroyed.
4. Compare trends with province-wide or state-wide trends.
5. Measure progress by the cumulative total of PCB materials disposed. PCB reductions should be as great or greater in the LSB as the province or state.

In addition to PCBs reported in Minnesota's hazardous waste disposal database, Minnesota records indicate that a spill of oil from PCB transformers was discovered in the LSB in 2007. Three transformers that were improperly stored leaked oil in a storage area. The site was cleaned up and the mining company that was responsible for the transformers was fined. An estimated 75 gallons (284 L) of PCB oil was spilled. Also, two distribution transformers that leaked a total of 6 gallons (23 L) of PCBs were cleaned up in 2006. As these are Comprehensive Environmental Response, Compensation and Liability Act (CERCLA)-based incidents and not allowed events under TSCA, these spills may not be representative of conditions elsewhere in the LSB or nationally. However, PCB release incidents are currently being analyzed by the U.S. TSCA program to better evaluate the issue of "releases as PCB sources" within the Great Lakes Basin.

PCBs may also be discharged and emitted from sources in the basin as part of combustion processes, through inadvertent production in manufacturing processes (such as pigment and dye making), or leaks or illegal discharges to stormwater or wastewater treatment plants/publicly owned treatment works. Although manufacturing processes that are associated with inadvertent PCB production may not be present in the LSB, it is possible that products made from these processes may be used in the LSB. The Superior Work Group (SWG) attempted to calculate discharges and emissions from the U.S. portion of the basin for the first time in this Milestones report. The number was estimated to be 36 kg, mostly from backyard burning, although there is a low confidence in this number.

[3] Regulatory and/or reporting mechanisms are not currently available to separately track the Wisconsin and Michigan PCB disposal records from facilities within the LSB. See Appendix D for the PCB management approach in Michigan and Wisconsin.

3.6 Pesticides

3.6.1 Pesticide Reduction Goals

Use of dichlorodiphenyltrichloroethane (DDT), toxaphene, chlordane and aldrin/dieldrin peaked in the mid 1960s to mid 1970s. All of these pesticides' registrations were cancelled; production is legal, but sale and distribution within the U.S. and Canada is illegal. Cancellations occurred in the 1980s for domestic use in the U.S. and by the 1990s for domestic use in Canada. U.S. companies may still produce and export cancelled pesticides or "trans-ship" pesticides produced in one country and shipped to another. As recently as 2003, the Foundation for Advancement and Scientific Education found evidence of production or trans-shipment of these pesticides by U.S. companies in the Port Import Export Retrieval Service records (Smith et al., 2008).

The LSBP goal was to retrieve and destroy all remaining stockpiles of the cancelled pesticides in the basin by the year 2000. Cancelled pesticides targeted for collection include DDT, dichlorodiphenyldichloroethylene (DDE), aldrin/dieldrin, toxaphene, dicofol (also known as Kelthane), hexachlorobenzene, mercury pesticides, HCB pesticides, 2,4,5-T (Silvex) and other pesticides that have the potential to be contaminated by dioxin or HCB. Although significant quantities have been collected, it is not possible to assure that all stockpiles have been removed. In fact, significant quantities of some cancelled pesticides continued to be collected annually since 2000.

3.6.2 Pesticide Collections

Although U.S. and Canada domestic production has ceased and uses have been cancelled, these pesticides continue to have an environmental presence. Furthermore, it should be noted that toxaphene and other pesticides in Lake Superior mostly originate from regions outside the basin and significant amounts arrive through aerial transport and deposition. Collection programs in the LSB continue to gather these pesticides. Lake Superior strategies for pesticides include continued or expanded collection opportunities coupled with concerted public outreach. This approach has the advantage of collecting not only the pesticides targeted for zero discharge, but the other pesticides that are considered critical chemicals for Lake Superior (i.e., heptachlor and hexachlorocylcohexane [HCH]). The collections carried out in each Lake Superior jurisdiction are described below.

<u>Michigan</u>

In Michigan, the safe and proper disposal of outdated, unused or unwanted pesticides is accomplished primarily through the Michigan Clean Sweep Program, which is administered by the Michigan Department of Agriculture and Rural Development. Participating Clean Sweep sites will accept pesticide products and mercury free of charge from any Michigan resident. Over the past 14 years, nearly 1.7 million pounds (850 tons) of pesticides have been removed from circulation and properly disposed of via permanent collection sites. Currently, there are over a dozen Clean Sweep sites established around the state. The only long-standing collection site solely in the Lake Superior basin is located in Marquette and is operated by the Marquette County Solid Waste Management Authority.

Determining long-term trends in waste pesticide collections in Michigan's Upper Peninsula is difficult because of the limited availability of historical data. In general, pesticide usage has been considered low in this area of the Lake Superior basin, and few pesticides were collected between 1990 and 1996 (Knorek, 2005). Data from 1996 to 2006 are limited because they are not in electronic form and/or are not readily available. However, approximately 434 kg (955 pounds) of pesticides were collected in Upper Peninsula counties from September 2002 to October 2003, including 15 kg (33 pounds) of DDT (LSBP, 2004).

Beginning in 2003, the State of Michigan began to systematically track collected products by U.S. EPA registration number or active ingredient. Soon thereafter, the state developed a database to better track, monitor and report collection data. In recent years, Michigan has been able to document the collection of tens of thousands of pounds of pesticides from around the state. In 2010, the Marquette collection site alone brought in a total of 3,457 kg (7,622 pounds) of pesticides (formulated product, including unknowns).

Table 3-11a shows the recent collection data specifically for zero discharge pesticides at the Marquette collection site. Amounts are listed as weight of active ingredient, not whole product. Overall, the amounts have been low for these pesticides, possibly indicating their relative scarcity in the region or the fact that they were brought in during earlier collections. It is unclear why DDT-containing products were received in 2010 but not in other years. It is possible that such products were received but without clear labeling. In fact, the Marquette site collects a large quantity of "unknown" pesticides (material without clear formulation information) annually. For example, in 2010 the total unknown submissions (formulated product) weighed approximately 298 kg (658 pounds).

In addition to the Clean Sweep site at Marquette, several other Upper Peninsula counties have organized pesticide collections independent of the Clean Sweep Program. Specifically, both Chippewa County and Mackinac County provided collection opportunities in recent years (Chippewa County joined Clean Sweep in 2011). As shown in Table 3-11b, these sites collected a large quantity of unused pesticides. Product amounts were tracked by net weight. Although breakdowns by active ingredient are not available, it is likely that some of the ZDDP chemicals were received at these collection sites. Unlike Marquette, both Chippewa and Mackinac Counties straddle multiple Great Lakes basins and thus some of the collected stockpiles derive from outside the Lake Superior basin. Overall, the data suggest that demand continues to exist for pesticide collection programs in the Upper Peninsula.

Table 3-11a. Waste Pesticides Collected in Marquette County, Michigan, Fiscal Years 2006 to 2010 (kg)*

Pesticide	2006	2007	2008	2009	2010	Total
Chlordane	1.2	NA	0.3	0	0.8	2.3
DDT	0	NA	0	0	3.2	3.2
Dieldrin/ Aldrin	0	NA	0	0	0.9	0.9
Dioxin†	0.3	NA	0.2	0.2	0	0.7
Heptachlor	0	NA	0	0	0	0
Mirex	0	NA	0	0	0	0
Toxaphene	0	NA	0	0	0.1	0.1
Total	1.5	NA	0.5	0.2	5.0	7.2

NA = Not Available
* All amounts reported are weight of active ingredient, not formulated product. Data from 2007 were not reported to the state.
† The pounds listed are for two pesticides containing dioxin, Silvex and 2,4,5-T.
Source: Michigan Department of Agriculture and Rural Development, 2011

Table 3-11b. Waste Pesticides Collected in Chippewa and Mackinac Counties, Michigan, 2005 to 2010* (net weight, kg)

Pesticide	2005	2006	2007	2008	2009	2010	Total
Chippewa – liquids	272	149	101	144	63	43	772
Chippewa – solids	50	143	47	39	24	35	338
Chippewa – aerosols	0	0	0	0	0	0	0
Mackinac – liquids	0	0	92	225	33	36	386
Mackinac – solids	39	0	10	262	152	22	485
Mackinac – aerosols	0	0	6	11	1	4	22
Total	361	292	256	681	273	140	2,003

Source: Drug & Laboratory Disposal, Inc., 2012

Minnesota

The Minnesota Clean Sweep Program began to collect waste pesticides in 1990. Over 2 million kg (4.6 million pounds) of waste pesticides have been collected and documented since the program began. Many of these pesticides were collected during clean sweep collections organized and staffed by the Minnesota Department of Agriculture (MDA). Sources included farms, small businesses, golf courses, nurseries, greenhouses, city and county parks, and road maintenance departments. The first waste pesticide clean sweeps were held in the LSB in 1992.

Since 1996, the WLSSD has had a cooperative agreement with MDA to collect and inventory all pesticides collected from households in Carlton, Cook, Lake, and St. Louis Counties. As part of this agreement, MDA would pay for the disposal of household pesticides collected at the Duluth Regional HHW facility, run by WLSSD, and shipped for disposal.

In 2002, the MDA began clean sweep operations with reduced funding. To address the continuing need for pesticide disposal, MDA worked with county and regional HHW establishments to expand existing partnerships to provide continuous opportunities for businesses and farmers seeking disposal of unwanted pesticides. MDA would pay for disposal of collected pesticides as well as continue to dispose of a predetermined amount of HHW waste pesticides. WLSSD has continued its cooperative agreement with MDA and documents certain pesticides disposed of from the area. Since spring of 2004, WLSSD is a cooperator in a new partnership with MDA that allows WLSSD to collect, store, and ship agricultural/business pesticides for payment by MDA. WLSSD also ships HHW waste pesticides for payment by MDA. To date, MDA has been able to pay for and record all pesticides that WLSSD has collected; however, future collections and payments depend on funding.

The new waste pesticide collection program has dropped the requirement to inventory household pesticides due to the time demand it places on HHW staff. However, partners are still required to record all PBT household pesticides (including dioxin bearing pesticides) that are received for disposal by MDA. MDA also requires participating facilities to document agriculture and business waste pesticides in order to distinguish them from HHW waste pesticides.

Table 3-12 presents data for pesticides targeted for zero discharge in the LSB. The table includes pesticides collected from northeastern Minnesota counties, which are mostly non-agricultural. WLSSD may have collected and disposed of household waste pesticides and even some business waste pesticides prior to 1996; however, these are not included in the table due to the difficulty in retrieving and analyzing paper records prior to 1996. All agriculture special event clean sweep collections held in the Basin are included. Any pesticides shipped by WLSSD for payment by MDA are recorded in MDA's database and thus are included in the tables.

The greatest amount of pesticides was collected from St. Louis County, where WLSSD is located. This may be influenced by how collected pesticides were inventoried. If a pesticide was not identified to a county, then it was listed under St. Louis County. No pesticides were collected from Cook County. During the period 1992-2007, approximately 3,569 kg (7,700 pounds) of DDT was collected. No mirex was collected from these counties from 1992 to 2007.

Wisconsin

The Wisconsin Department of Agriculture, Trade and Consumer Protection supports clean sweeps in Wisconsin counties. The first year that clean sweep grants were awarded in LSB counties was 1992. However, the data for 1992 were not broken down by individual pesticide. No data were reported for LSB counties in the years 1993-1995. In 1995, the Northwest Cleansweep Program was established for the collection and disposal of hazardous wastes in the northwest Wisconsin region. The program, run by the Northwest Wisconsin Regional Planning Commission (NWRPC) with funding from the Wisconsin Department of Agriculture, Trade and Consumer Protection, began agricultural collections in LSB counties in 1996. Table 3-13 presents data on agricultural pesticides collected by the NWRPC (from farmers and agribusinesses) beginning in 1996.

In addition to agricultural clean sweeps, periodic HHW collections have been conducted in northwestern Wisconsin counties. The Environmental Resources Center at the University of Wisconsin-Madison compiles and maintains data on Wisconsin Hazardous Waste Collection Programs, featuring households and very small quantity generator programs, at www.uwex.edu/erc/hazwste.html. The type of pesticide collected is not reported for HHW collections. In 2004, in the four-county LSB area of Wisconsin, no pesticides were collected during HHW collections held in Douglas County (no collections were held in Ashland, Bayfield, or Iron Counties).

While the Minnesota and Wisconsin pesticide collection reporting requirements are similar, it is not possible to combine them due to different periods of record (i.e., 1992-2007 for Minnesota and 1996-2010 for Wisconsin) and areas of coverage. Counties that are outside the Lake Superior basin but are lumped with Lake Superior counties in Minnesota (i.e., Aitkin, Itasca and Koochiching are not in the Lake Superior basin) are much less agricultural than the Wisconsin counties that are lumped with the Lake Superior counties (i.e., Burnett, Eau Claire, Price, Rusk, Sawyer, St. Croix, and Taylor are not in the Lake Superior basin).

Assuming the Minnesota data are the closest fit for the mostly non-agricultural basin, Figure 3-9 shows the rate of collections over time using a cumulative analysis (e.g., the amount of pesticides shown for 2007 is the cumulative amount collected between 1992 and 2007). Figure 3-9 shows that the rate of collection of these particular banned pesticides began to slow starting in 2001. While it is difficult to extrapolate based on a rate change, this suggests that most stockpiles of these banned pesticides have been collected and the current low rate of disposal reflects previously unknown stockpiles that are discovered during property transfer. Because the "break point" between the old and new rates is 2001, it appears that the LaMP Stage 2 goal of collecting all the stockpiles by 2000 was not met, but only missed the goal by one year in Minnesota and quite possibly in the rest of the Lake Superior basin.

Table 3-12. Waste Pesticides Collected in Minnesota Lake Superior Counties[1], 1992-2007 (kg of Product[2])

Pesticide	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	Total
Chlordane	74	23	5	47	90	92	83	64	83	72	4	23	42	20	37	9	767
DDT	451	336	24	51	1403	135	253	267	306	134	32	34	59	40	40	5	3569
Dieldrin/ Aldrin	5	5	0	0	6	1	27	8	4	24	0	0	0	0	0	0	80
Dioxin[3]	0	0	0	0	0	0	0	0	0	0	0	0	0	60	25	33	117
Heptachlor	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0.454
Mirex	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Toxaphene	16	5	0	10	6	5	1	13	1	3	0	0	0	0	0	0	61.23
Total	**547**	**368**	**29**	**108**	**1505**	**234**	**364**	**352**	**394**	**234**	**36**	**58**	**100**	**120**	**104**	**47**	**4597**

[1] Includes data for pesticides collected in Carlton, Cook, Lake, and St. Louis Counties as well as Koochiching, Itasca and Aitkin Counties.
[2] Weight is for the pesticide product, not the active ingredient.
[3] The kilograms listed are for pesticides containing dioxin. These include Silvex, 2,4-D with 2,4,5-T, fenchlorphos, Ronnel, some Weedones, and a few others.
Source: (Kaminski, 2011)

Table 3-13. Waste Pesticides Collected in Wisconsin Northwest Cleansweeps[1], 1996-2010 (kg of Product[2])

Pesticide	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	Total
Chlordane	0	2	0	19	45	0	33	34	1	9	6	25	18	27	26	244
DDD/DDT	36	3	0	61	76	101	30	5	0	8	11	5	8	15	13	372
Dieldrin/ Aldrin	0	4	0	330	6	10	0	0	0	0	0	0	3	14	23	390
Dioxin [3]	375	73	268	588	516	476	422	364	221	345	295	220	179	188	269	4798
Heptachlor	0	0	0	0	0	0	0	4	0	0	0	5	0	0	0	10
Mirex	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Toxaphene	218	0	0	0	0	0	0	0	0	0	0	0	4	0	0	221
Total	**629**	**82**	**267**	**998**	**643**	**587**	**485**	**407**	**223**	**362**	**312**	**255**	**212**	**244**	**331**	**6036**

[1] Includes data for pesticides collected in counties served by the NWRPC as follows:

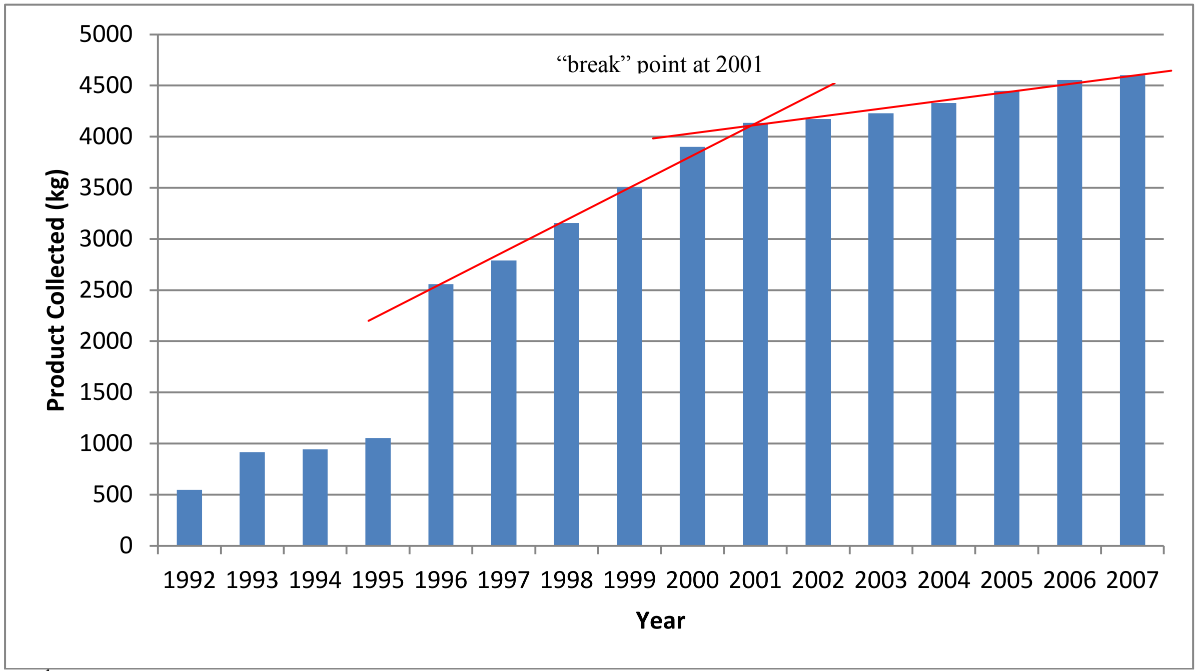
1996: Ashland, Bayfield, Douglas, Iron; 1997: Ashland, Price, Taylor, Washburn; 1998: Counties served were not specified; 1999: Ashland, Douglas, Eau Claire, Iron, Rusk, Sawyer, St. Croix, Taylor, Washburn

2000: Ashland, Bayfield, Douglas, Iron, Price, Rusk, Sawyer, St. Croix, Taylor, Washburn; 2001-2003: Ashland, Bayfield, Burnett, Douglas, Iron, Price, Rusk, Sawyer, St. Croix, Taylor, Washburn; 2004: Ashland, Bayfield, Burnett, Douglas, Iron, Price, Rusk, Sawyer, Taylor, Washburn; 2005: Ashland, Bayfield, Burnett, Douglas, Price, Rusk, Sawyer, Taylor, Washburn; 2006-2009: Ashland, Bayfield, Burnett, Douglas, Iron, Price, Rusk, Sawyer, Taylor, Washburn; 2010: Ashland, Bayfield, Burnett, Douglas, Price, Rusk, Sawyer, Taylor, Washburn

[2] Weight is for the pesticide product, not the active ingredient.

[3]The pounds listed are for pesticides containing dioxin. These include Silvex, 2,4-D, pentachlorophenol, and 2,4,5-T.

Source: (Johnson, 2012).



[1] Includes Carlton, Lake and St. Louis Counties (and possibly Aitkin, Itasca and Koochiching Counties)

Figure 3-9. Cumulative Amount of Pesticide Products Collected in Northeast Minnesota 1992-2007 (kg)[1].

Ontario

On April 22, 2009, Ontario's Cosmetic Pesticides Ban was implemented under the Pesticides Act and Ontario Regulation 63/09 to prohibit the use of certain pesticides for cosmetic purposes. There are exceptions to the ban for the use of prohibited pesticides, such as agriculture and forestry. There are also exceptions to the ban for golf courses, specialty turf and public works, that require integrated pest management certification of licensed exterminators and the preparation of annual reports on prohibited pesticide use.

Under the ban, the use of biopesticides and certain lower risk pesticides are allowed for cosmetic uses such as on lawns, gardens, parks and school yards, The Ontario government has also provided funding to the Agricultural Adaptation Council to administer a grant program for the research and development of new biopesticides and lower risk pesticides. Information about the ban is available on the ministry website at: www.ontario.ca/pesticideban.

Since 1990, ZDDP pesticides have been removed as waste from the LSB as one of the objectives. Representatives of the Ontario Ministry of the Environment (OMOE) and the Ontario Ministry of Agriculture Food, and Rural Affairs (OMAFRA) were contacted regarding quantities of waste pesticides removed from the LSB over the 1990 to 2010 period. The agencies do not have a central database or inventory that tracks removed waste pesticides and therefore are not able to provide the requested information.[4] OMAFRA, with the support of Croplife, conducted an Ontario Obsolete Pesticides Collection event in 2009 in the Thunder Bay area. A total of 1,027 kg was collected at the Thunder Bay Coop (Brooker, 2010).

The Thunder Bay landfill and hazardous waste carriers operating in the LSB were contacted by the Canadian engineering consultant regarding quantities of pesticides, including those targeted for zero discharge in the LSB, removed from the Basin. Waste pesticides are amalgamated into drums under the OMOE waste classification number and there is no record for quantities of specific pesticides disposed (i.e., wastes are tracked by waste class and not specifically by pesticides that are part of the ZDDP). As summarized in Table 3-14, a total of 1,435 L and 192 kg of pesticides were collected by the Thunder Bay landfill and 160 L by EcoSuperior in 2005 (Benazon Environmental Inc,, 2006). The landfill collected 2,255 L in 2009. Clean Harbors reported that over the period of 2006 to 2009, a total of 479 L plus 6.5, 205 L labpack drums of pesticides were removed. Potter Environmental documented a total of 320 kg collected in 2009. Thus, the total quantities collected over the period of 2005 to 2009 are 1,539 kg, 4,329 L and 6.5, 205 L labpacks.

The cumulative total collected by all agencies since 1990 is not known.

3.6.3 Conclusions

Although the LSB is mostly non-agricultural, some banned pesticides were used for residential, silvicultural, or property management purposes and a large amount of banned pesticides have been collected in or near the basin since 1992. While the LaMP Stage 2 reduction goal was to

[4] Hazardous wastes are tracked by waste classes.

collect all of the pesticides that contained any of the nine ZDDP chemicals by 2000, it is obvious that these pesticides are still present.

It appears that most known pesticide stockpiles have been depleted. Where long-term data sets exist (e.g., Minnesota), the collection rate for these pesticides began to slow by 2001 (Figure 3-9). At the very least, the message is getting out to the communities and these pesticides are being collected for proper disposal. Finding these pesticides and seeing a continuing disposal pattern is a clear indication of the need for waste pesticide collections to continue, even in non-agricultural area.

Table 3-14. Summary of Pesticides Collected from the Ontario Portion of the LSB

Organization	Quantities			
	2005	2006-2009	Total	Units
Thunder Bay Landfill	1435	2255	3690	L
	192	No Data	192	kg
EcoSuperior	160	No Data	160	L
OMAFRA	No Data	1027	1027	kg
Clean Harbors	No Data	479	479	L
	No Data	6.5	6.5	205 L Labpack
Potter Environmental	No Data	320	320	kg
Total kg	**192**	**1347**	**1539**	kg
Total L	**1595**	**2734**	**4329**	L
Total Labpacks,	No data	No data	**6.5**	205 L Labpacks

3.7 Hexachlorobenzene

The HCB inventory is incomplete and is subject to several caveats, as noted below:

- HCB data are missing from the former Algoma Steel sintering plant in Wawa. However, since 19.4 g of dioxin was produced at the sintering facility in 1990 and reports on other iron sintering plants show large quantities of both dioxin and HCB are emitted, HCB was also likely emitted from the Ontario facility.
- Representatives of the Thunder Bay Generating Station have indicated that previous HCB estimates were erroneously calculated by the facility and that the facility is not, and has never been, a source of HCB in the basin. NPRI data from 2003 and onwards have been revised to show zero emissions from this source.
- HCB emissions from coal combustion were not estimated for the U.S. side.
- Canadian emissions for landfill fires are estimated at zero for 2010.
- There are no current medical waste incinerators operating on the Canadian side of the LSB because of Ontario Regulation 323/02, which required that all hospital incinerators

be shut down by the end of 2003. Four operated in 1990: two were shut down in 1994 and the other two were shut down in 2003.

- On the Canadian side, total on-site emissions/releases have declined from 218 g/yr in 1990 to 122 g/yr in 2010—a reduction of 44%, which is well below the 85% interim target reduction. The decrease is mostly associated with reductions in HCB from pulp and paper, as a result of the conversion of the bleaching process to chlorine dioxide in place of elemental chlorine.
- In each of the milestone years, the main sources of HCB on the Canadian side of the LSB are atmospheric emissions from on-site residential waste combustion (burn barrels; 96 g/yr) and leaching from pentachlorophenol-treated poles (16 g/yr). There is considerable uncertainty associated with these estimates because emission factors are based on limited data and because of the uncertainty associated with the activity data (quantity of waste burned in on-site residential waste combustion). Emissions from these sources are not expected to drop significantly over the next five years.
- The largest source in the U.S. HCB inventory was open burning of trash, followed by mobile sources.

Table 3-15 provides a tabular summary of the HCB inventory. Due to the caveats listed above, percent reduction calculations were not made for HCB; trends with the dioxin inventory will continue to serve as a surrogate for HCB trends.

Table 3-15. Hexachlorobenzene Releases to Air and Water from Sources in the Lake Superior Basin, g/yr

Source	1990 (g/yr)			2000 (g/yr)			2005 (g/yr)			2010 (g/yr)		
	U.S. 1990	Canada 1990	Total 1990	U.S. 2000	Canada 2000	Total 2000	U.S. 2005	Canada 2005	Total 2005	U.S. 2010	Canada 2010	Total 2010
Industrial	0.7	68.0	68.7	0.7	2.2	2.9	0.7	0.8	1.5	0.7	1.8	2.5
Fuel Combustion	54.7	10.6	65.3	57.8	10.3	68.1	60.6	10.3	70.9	63.3	7.8	71.1
Incineration	776.3	117.0	893.3	509.8	147.0	656.8	361.2	94.0	455.2	347.3	96.0	443.3
Pentachlorophenol Use	35.9	22.0	57.9	33.5	18.0	51.5	32.5	17.0	49.5	29.2	16.0	45.2
Total	867.7	217.6	1085.3	601.8	177.5	779.3	455.1	122.1	577.2	440.6	121.6	562.2

Chapter 4. Re-evaluation of Critical Chemicals

The LaMP Stage 2 document sets out a process for categorizing and managing pollutants in Lake Superior. The management goals are to restore impaired uses and achieve environmental criteria and lake ecosystem objectives. Based on this process, 23 critical and 14 prevention pollutants were identified (LSBP, 1998a).

Initially, a list of "chemicals of concern" was developed by combining the U.S. Great Lakes Water Quality Guidance (GLI) Bioaccumulative Chemicals of Concern (BCCs) and the list of Tier I and Tier II substances under the Canada-Ontario Agreement (COA). The chemicals of concern were then systematically evaluated, along with other substances identified in the Stage 1 LaMP, following the Management Goal Flow Chart for Lake Superior Critical Chemicals and placed into either the "critical" or "prevention" pollutant categories.

The list of critical pollutants includes substances that require reductions at the source and/or removal from the ecosystem to restore beneficial uses, achieve ecosystem objectives, meet jurisdictional environmental criteria, or are one of the nine substances in the ZDDP. Prevention pollutants have properties that give them the potential to impair the lake ecosystem (e.g., fish consumption advisories, fish and wildlife health impairments, and human health impairments) but they either have been found below harmful levels or they have not been monitored in Lake Superior. The intention is to manage the prevention pollutants to avoid impairments in the future.

To guide the development of load reduction or remedial strategies, critical and prevention pollutants were grouped into management categories. The critical pollutants are subdivided into three management categories, while the prevention pollutants are grouped into one of two management categories. The substances are listed by management category in Table 1-1 and an explanation of the management approaches can be found in Table 4-1.

This chapter provides an overview of the environmental levels of critical pollutants, prevention pollutants, and substances of emerging concern in Lake Superior air, water, sediment, and wildlife. This is followed by a Three-Part Management Strategy for chemicals of emerging concern, and an overview of levels of chemicals of emerging concern in the Lake Superior ecosystem.

Table 4-1. Management Approaches for Lake Superior Critical and Prevention Pollutants

Management Category	Description
Critical Pollutants	**Levels of PBT chemicals should not impair beneficial uses of the natural resources of the LSB. Levels of critical pollutants which are persistent, bioaccumulative and toxic should ultimately be virtually eliminated in the air, water and sediment in the LSB.**[1]
Zero Discharge[2]	As a management approach, virtual elimination from the environment requires that zero discharge or emission is applied to the use, generation, and release of PBT substances originating from human activities. The effect of these chemicals is found both locally and lakewide. Sources may be local or outside of the basin.
Lakewide Remediation	These pollutants have less potential to bioaccumulate than those in the zero discharge. Some of the lakewide remediation pollutants are responsible for nearshore problems in multiple locations, and some exceed criteria in open lake waters. The management approach for these pollutants is to coordinate lakewide reductions in loadings.
Local Remediation	Local remediation pollutants consist of metals that impact AOCs or other nearshore areas. These are mainly metals that have both natural sources and sources due to human activity. The management approach is concurrent localized reduction in loads and remediation of hot spots.
Prevention Pollutants	**Prevention pollutants have properties that give them potential to impair the lake ecosystem, but they have been found below harmful levels or have not been monitored in Lake Superior. The intention is to manage the prevention pollutants to avoid impairments in the future.**
Monitor	Although these pollutants have not been found at harmful levels in the Lake Superior ecosystem, the ecosystem should be monitored to confirm the continued absence at levels of concern for these pollutants.
Investigate	Substances in this category have been identified as being of concern by Lake Superior programs such as GLI or COA. Because these pollutants were not sampled in previous surveys, they should be sampled for in the future.

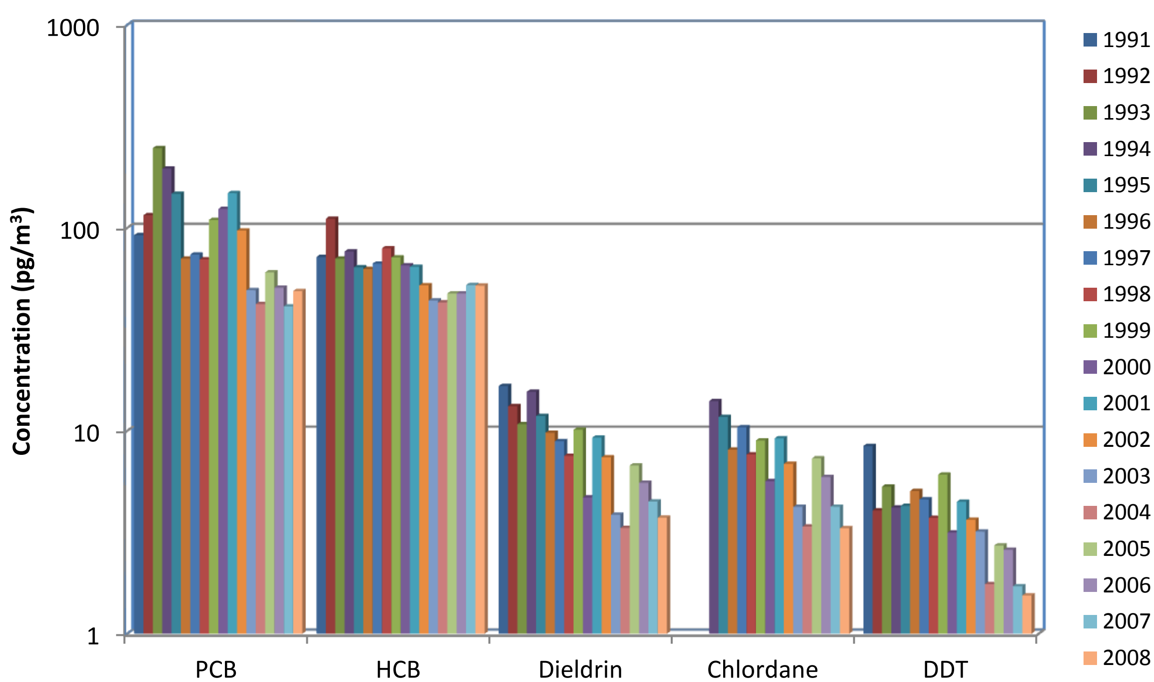
[1] Source: LSBP, 1998b
[2] This category was referred to as Virtual Elimination in the LaMP Stage 2 report.

4.1 Contaminant Levels and Trends Summary

This section provides general information on current levels as well as spatial and temporal trends of certain PBT chemical contaminants in the Lake Superior ecosystem. These contaminants are monitored in a variety of media including air, water, sediments, herring gull eggs, bald eagles and lake trout. Examining contaminant trends across multiple media provides insight into ecosystem-wide trends.

4.1.1 Atmosphere

The Integrated Atmospheric Deposition Network (IADN) has been monitoring levels of persistent organic pollutants (POPs) in the atmosphere at six sites throughout the Great Lakes since 1991. In general, the Lake Superior sampling site at Eagle Harbor has atmospheric POP concentrations among the lowest of the six stations. Most chemicals monitored (including PCBs, HCB, dieldrin, chlordane, and DDT) have decreased over time (IADN, 2008; Venier and Hites, 2010a, b). Figure 4-1 shows the time-trends for five ZDDP chemicals.

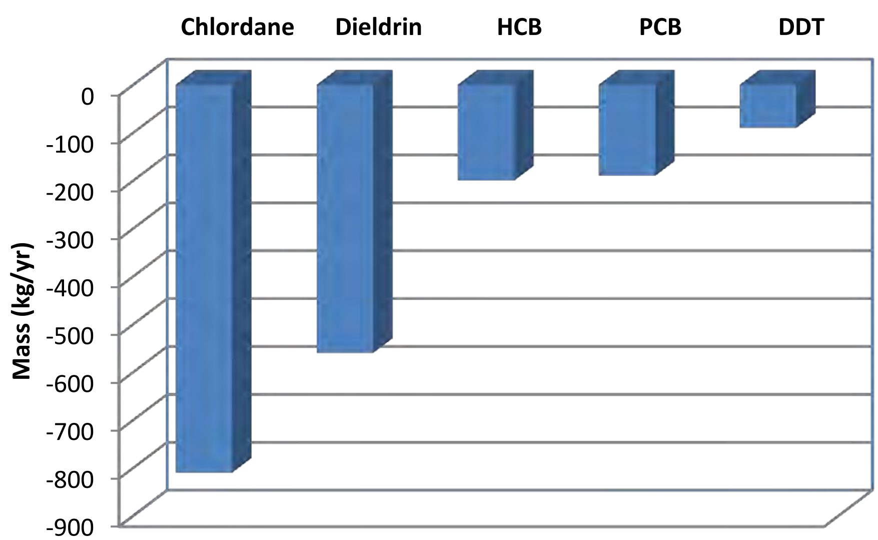


Source: Nettesheim and Craddock, 2011

Figure 4-1. Time Trends for Persistent Organic Pollutants Measured by IADN at Lake Superior's Eagle Harbor Station from 1991-2008.

The declines in atmospheric concentrations of critical pollutants are a result of reduced use or outright bans on these chemicals. Among this suite of chemicals, PCBs have shown the slowest rate of decline, particularly at the remote northern sites on Lakes Superior and Huron. The relatively small decline in atmospheric PCB concentrations over the last two decades is noteworthy since these chemicals were banned in the U.S. over 30 years ago. The observed trends in Lake Superior may be related to the fact that Lake Superior has colder water temperatures and a larger volume relative to the other Great Lakes (IADN, 2008). The slow decline overall across the Great Lakes Region may also indicate that PCBs in existing industrial/electrical equipment or in storage and disposal facilities may still be slowly leaking into the atmosphere (Venier and Hites, 2010a, b). Given that known quantities of PCB equipment nearing the end of life still exist within the LSB, this is highly likely.

Lakes respond more slowly to chemical use reductions than the atmosphere. Once banned, many pollutants quickly decline in the atmosphere. As a result, the atmosphere becomes a sink for these chemicals as they escape from the lake, rather than a source of contaminants to the lake. This is currently the case for PCBs, dieldrin, chlordane, DDT and HCB, as shown in Figure 4-2. Lake Superior is slowly moving towards a steady state where atmospheric inputs of these chemicals to the lake will equal outputs from the lake.



*Negative numbers indicate that the net flow of a chemical over the course of the year is from the lake into the atmosphere

Source: IADN, 2008

Figure 4-2. Atmospheric Flow (kg/yr) of Persistent Organic Pollutants at the IADN Eagle Harbor Site on Lake Superior in 2005*.

4.1.2 Water

A number of pollutants have decreased in Lake Superior waters over the last three decades. Nevertheless, some chemicals are still present at levels of concern. Of the nine ZDDP critical pollutants, the concentrations of three chemicals (PCBs, dieldrin and toxaphene) continue to exceed certain jurisdictional water quality standards. Table 4-2 shows recent open lake concentrations of certain critical pollutants relative to water quality yardsticks around the basin. As noted in Section 4.1.1, open lake water concentrations of these pollutants respond more slowly than atmospheric levels to reductions in use. Consequently, critical pollutants currently exceeding water quality guidelines could potentially remain above these thresholds for many years to come.

Table 4-2. Concentrations (ng/L) of Select Critical Pollutants in Lake Superior Open Lake Water Compared to Jurisdictional Water Quality Yardsticks[a,b]

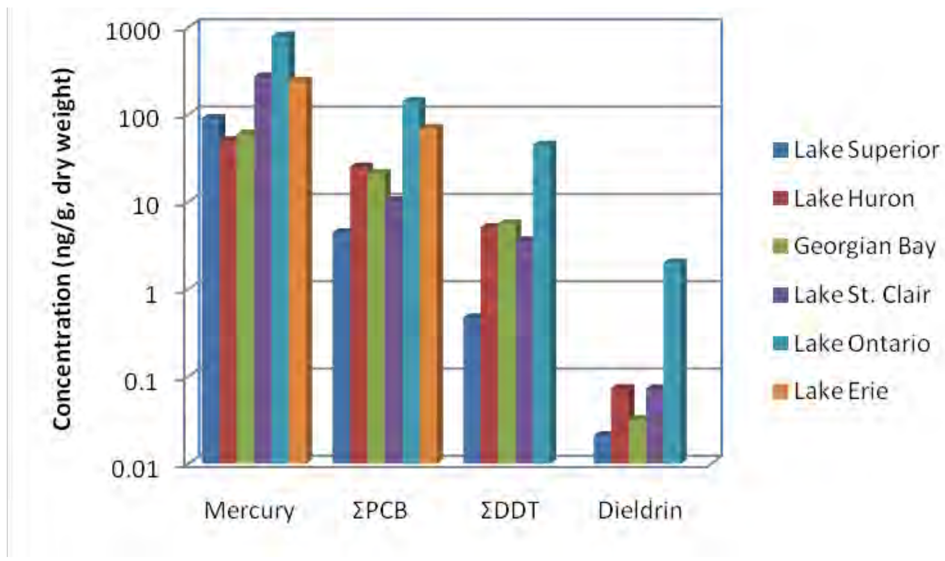
Critical Pollutant	Jurisdictional Water Quality Yardstick (ng/L)				Open Lake Concentration (ng/L)
	Minnesota[c]	Michigan[c]	Wisconsin[c]	Ontario	
PCB	0.0045	0.026	0.003	1.0	**0.043**[e]
HCB	0.074	0.45	0.22	6.5[d]	0.013[e]
Dieldrin	0.0012	0.0065	0.0027	1.0	**0.112**[e]
Chlordane	0.04	0.25	0.12	60	0.013[e]
DDT	0.011	0.011	0.011	3.0 [d]	0.005[e]
Mercury	1.3	1.3	1.3	200	0.39[f], 0.21[g]
Toxaphene	0.011	0.068	0.034	8.0	**1.0**[h]

[a] Red values exceed one or more established yardstick value.
[b] The purpose of listing yardsticks is not to compare numbers across jurisdictions, but to provide a reference for comparing water quality results to available yardsticks and determine if exceedances are occurring. Ontario's Provincial Water Quality Objectives (PWQOs) are intended to protect aquatic organisms based on no adverse effects on growth, survival or reproduction. U.S. water quality criteria are based on human health considerations or the protection of wildlife that consumes aquatic organisms and thus tend to be more stringent than PWQOs for substances that bioaccumulate. Thus, the various yardsticks are not directly comparable (Ontario Ministry of the Environment, 1994).
[c] Water quality standards for the Lake Superior states are based on GLI methodologies.
[d] The Ontario PWQO for dieldrin refers to the sum of dieldrin and aldrin and the PWQO for DDT refers to the sum of DDT and its metabolites.
[e] Waltho, 2010; Great Lakes Surveillance Program 2005 data, total PCBs are blank-corrected
[f] Dove *et al.*, 2012; 2008 data
[g] Jeremiason *et al.* 2009; 2006 data
[h] Jantunen, 2011; 2005 data. Note: Toxaphene concentrations in Lake Superior water was reported to be 0.7 ng/L in the 2005 Milestones report. However, these small differences seen between years can be attributed to analytical variability for this difficult to measure chemical mixture.

4.1.3 Sediments

Contaminant levels in the sediments of Lake Superior are generally among the lowest of the Great Lakes and below guidelines established to protect aquatic life. Environment Canada sampled mercury, PCBs, DDT, and dieldrin in Lake Superior and Lake Huron sediments at 87 stations in 2001-2002 and compared the results to existing sediment data throughout the region

(Gewurtz et al., 2008). Levels of these compounds in Lake Superior sediments were low, often approaching two orders of magnitude below levels previously measured in Lakes Erie and Ontario (Figure 4-3). None of these compounds exceeded the Canadian Sediment Quality Probable Effect Level (PEL; Canadian Council of Ministers of the Environment [CCME], 2002) at any of the 20 sample sites in Lake Superior. Similarly, Li et al. (2009) found that Lake Superior had the lowest sediment PCB concentrations of any of the Great Lakes.



Source: Gewurtz *et al.* 2008

Figure 4-3. Surficial Sediment 75th Percentile Concentrations (ng/g, dry weight) in the Great Lakes Region.

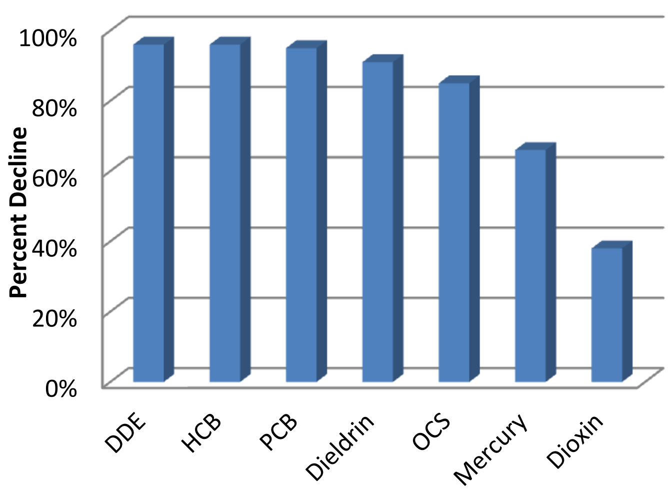
The primary source of contaminants to Lake Superior is atmospheric deposition (Gewurtz et al., 2008). Although dioxin, furan, and PCB congener profiles, measured as part of the Ontario Ministry of the Environment's Great Lakes Nearshore Monitoring and Assessment Program, show that local sources of contamination exist within Lake Superior (e.g., Thunder Bay, Peninsula Harbour, and the mouth of the Magpie and St. Marys Rivers), this is primarily a result of pulp and paper mills and wood preservation plants in the watershed (Shen et al., 2009).

Levels of legacy contaminants in Lake Superior sediments are generally stable or decreasing. Overall, it has been estimated that the total sediment PCB load in the Great Lakes has decreased by >30% from 420 to 300 tonnes (Li et al., 2009). But the reduction in contaminant loading is not uniform across the Great Lakes Basin; PCBs, DDT and mercury have shown little decline in Lake Superior sediments since they were first measured in the 1960s, which is not consistent with decreased releases of these chemicals over the last three decades (Gewurtz et al., 2008). This observation is believed to be due to the especially slow sedimentation rate in Lake Superior, with surficial sediments potentially integrating contaminant inputs over 30 or more years.

4.1.4 Herring Gull Eggs

Herring gulls are an especially useful avian species for monitoring contaminant trends because they are a top predator in the food web and they are permanent, year-round residents of the Great Lakes. Since they rely on both aquatic and terrestrial food sources, they are exposed to PBT chemicals through multiple pathways. Monitoring herring gull eggs provides information on both the level of contaminant exposure in this species and degree of maternal transfer of contaminants to the young.

The Canadian Wildlife Service has been monitoring a number of contaminants since 1974 through its Herring Gull Egg Monitoring Program. The program tests eggs from 15 colony sites including two on Lake Superior, Granite Island and Agawa Rocks. Typically, Lake Superior sites are among the least contaminated of the 15 sites, although mercury was intermediate relative to other Great Lakes colonies (Weseloh et al., 2011). Certain ZDDP chemicals, such as PCBs, DDE, HCB and dieldrin, have shown decreases of greater than 90% at the Lake Superior colony sites since monitoring began (Figure 4-4).



Chemical	Concentration[b]	
	1974/84[c]	2004/07/09[d]
DDE	16.7	0.70
HCB	0.25	0.009
PCB	62.8	3.29
Dieldrin	0.52	0.05
OCS	5.2	0.8
Mercury	0.36	0.11
Dioxin	16.0	9.87

[a] The concentrations reported are the average of the two Lake Superior sites: Agawa Rocks and Granite Island.
[b] All concentrations are reported in µg/g except OCS, which is in ng/g, and dioxin, which is in pg/g. All are reported on a wet weight basis.
[c] Dioxin monitoring began in 1984. All other contaminants have been monitored since 1974.
[d] The most recent data is 2009 for mercury; 2007 for DDE, HCB, PCB, and OCS; and 2004 for dieldrin and dioxin.

Sources: Weseloh and Havelka, 2005 & 2009; Weseloh et al. 2006; Weseloh et al. 2011

Figure 4-4. Percent Decline in Legacy PBT Chemicals in Herring Gull Eggs Collected at Two Lake Superior Sites between 1974/84 and 2004/2007/2009[a].

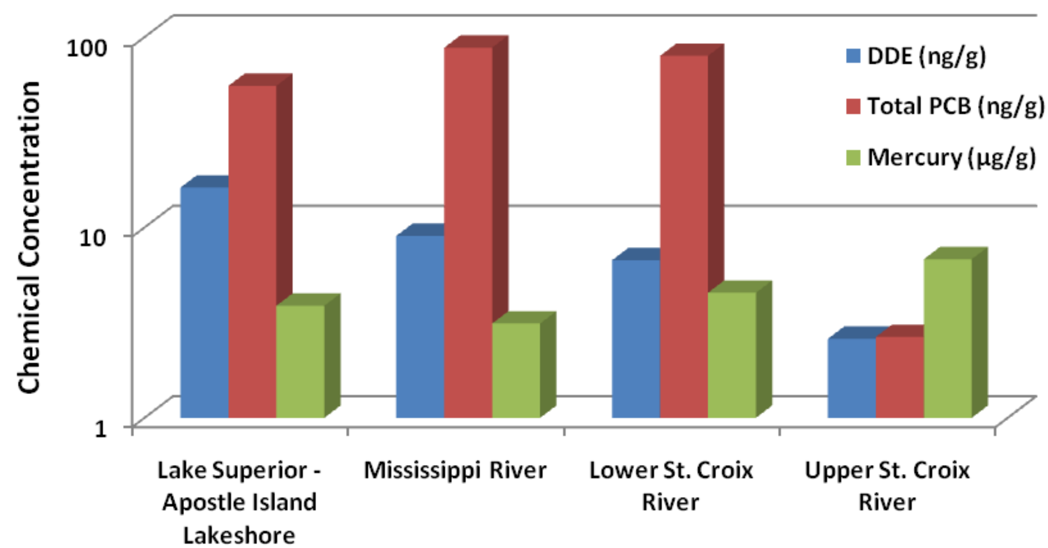
The measured contaminants have decreased 38% to 96% in Lake Superior herring gull eggs since monitoring began in 1974. But the most recent data show that in the last decade (1997-2007) there was no significant decline in most of these legacy contaminants (Weseloh and

Havelka, 2005 & 2009). Many chemical concentrations appear to be at or approaching a plateau in Lake Superior herring gull eggs. For example, Weseloh et al. (2011) found that in all 15 sites tested across the Great Lakes, including the two Lake Superior colonies, there was no significant decline in mercury concentrations in herring gull eggs from 1994-2009.

4.1.5 Bald Eagle Plasma and Feathers

Bald eagles are useful biosentinels of environmental contaminants because they reside at the top of the aquatic food web. In recent years, the U.S National Parks Service (NPS), the Wisconsin Department of Natural Resources (WDNR), and the MDEQ, as well as Clemson and Indiana Universities, have completed studies of bald eagles in the LSB investigating the levels and trends of a variety of contaminants including mercury, PCBs, and DDT.

Although some chemicals are lower in bald eagles nesting near Lake Superior relative to other locations, other contaminants may be higher in the Lake Superior coastal eagle population. For example, in northern Wisconsin, Dykstra et al. (2010) found DDE (a metabolite of DDT) was highest along the Lake Superior shore while mercury and PCBs were greater at certain inland locations south of the LSB (Figure 4-5). In addition, bald eagle nestlings from the Lake Superior shoreline in Wisconsin had feather mercury concentrations greater than in nestlings sampled at Voyageurs National Park (Minnesota), to the west of the LSB (Pittman et al., 2011). MDEQ found that mercury levels in nestlings tested between 2004 and 2008 were higher in the Lake Superior watershed than in the Lake Michigan or Huron watersheds (Weirda et al., 2009).

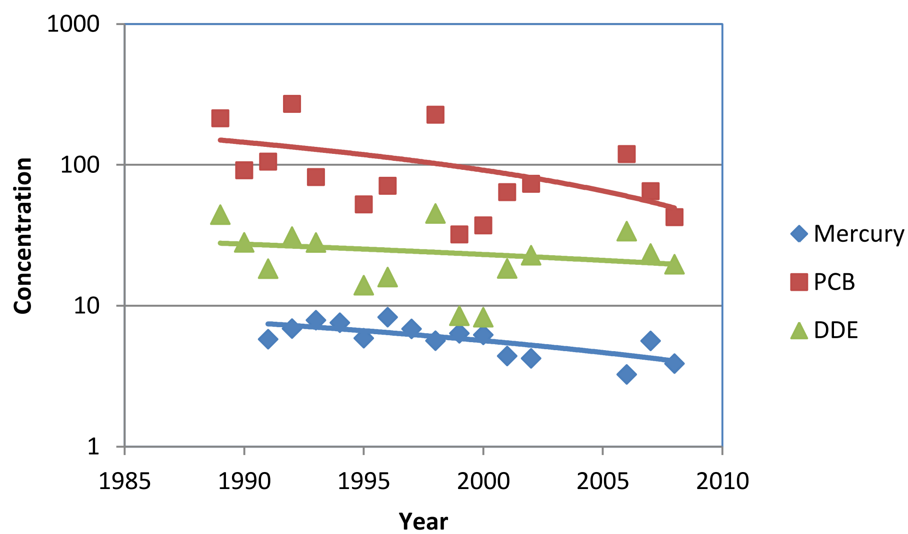


Source: Dykstra *et al.*, 2010

Figure 4-5. DDE and PCBs in Plasma and Mercury in Feathers of Bald Eagle Nestlings along Lake Superior and at Three Inland Sites (2006-2008).

Venier et al. (2010c) found evidence of contaminant "hot spots" along Lake Superior. Several flame retardants, PCBs and organochlorine pesticides were measured in nestling plasma at 15 sites in Michigan near the shores of Lakes Superior, Michigan and Huron. Bald eagles from one Lake Superior site (b-34) had the highest levels of DDTs, PCBs, dieldrin, chlordane and polybrominated diphenyl ethers (PBDEs) of any of the sites studied. Similar trends have also been observed in other piscivorous birds. For example, Evers et al. (2011) noted seven mercury hot spots in loons spread across the Great Lakes Region, including one in the LSB near Marquette in Michigan's Upper Peninsula. A biphasic mercury trend was also noted for this species in northern Wisconsin, with mercury levels decreasing from 1992-2000 but increasing from 2002-2010 (Meyer et al., 2011).

Although contaminants in bald eagles in the Great Lakes Region are below historic levels, there is evidence that certain chemicals may be reaching a plateau or once again increasing in this species. Dykstra et al. (2010) observed decreases in mercury (2.4% per year), DDE (3% per year) and PCBs (4.3% per year) from 1989-2008 in bald eagles nesting on the southern shore of Lake Superior (Figure 4-6). However, the rate of decline of PCBs and DDE over this entire time period was significantly lower than for the time period from 1989-2001 alone, suggesting organochlorine levels in eaglets may be nearing a plateau.



Source: Dykstra *et al.*, 2010

Figure 4-6. Time Trends (1989-2008) of Mercury (µg/g) in Feathers and PCBs (ng/g) and DDE(ng/g) in Plasma of Bald Eagle Nestlings at or Near the Apostle Islands National Lakeshore.

Similarly, MDEQ observed higher mercury levels in nestlings sampled during 2004-2008 than 1999-2003 within four out of 41 Michigan watersheds studied, while the opposite trend occurred in only one. Eagles in Voyageur National Park, just outside the LSB to the west, also displayed an increasing mercury trend between 2000 and 2010 (Pittman et al., 2011).

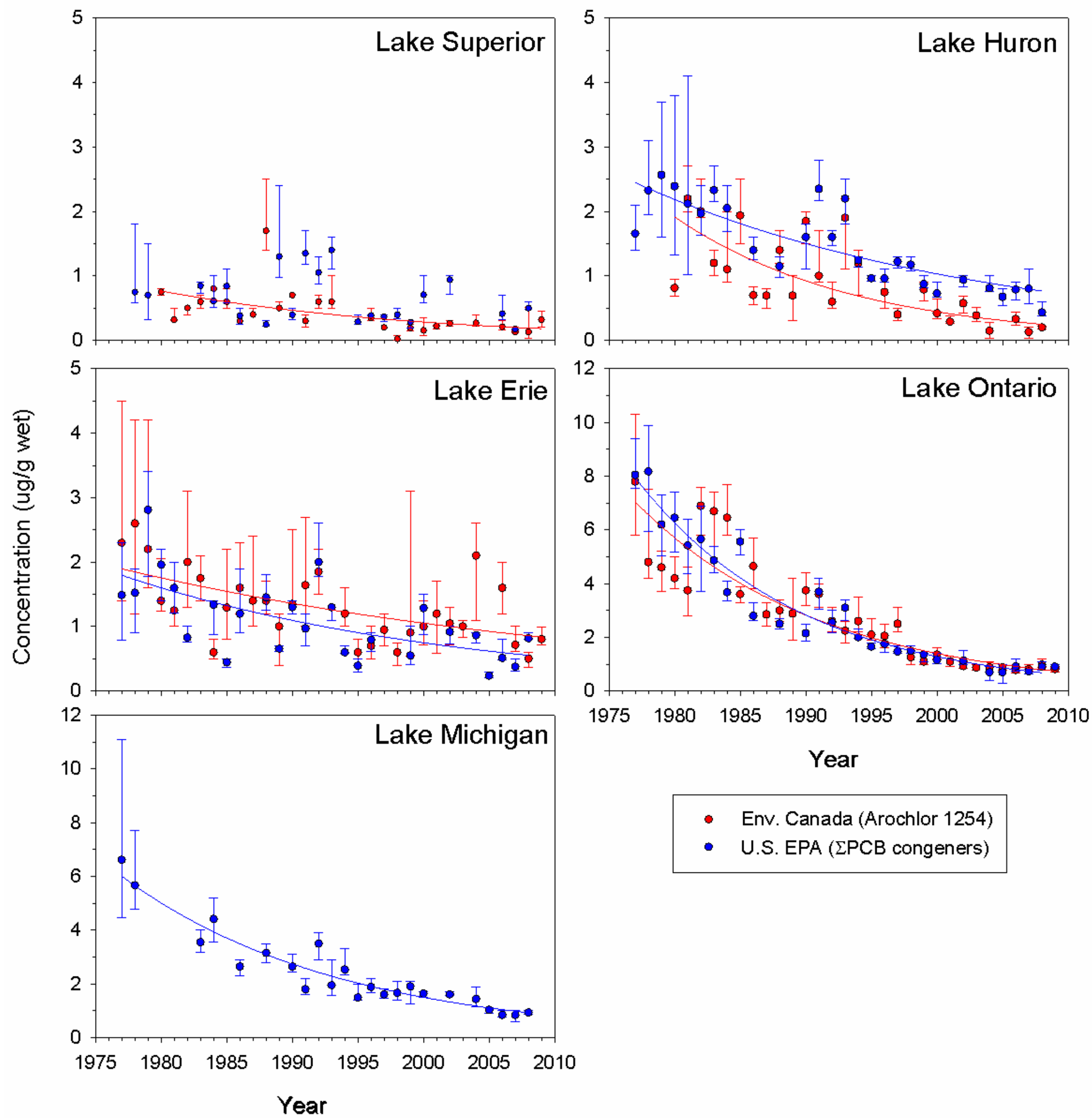
Taken as a whole, bald eagles demonstrate that Lake Superior is not, as is often assumed, always the least contaminated of the Great Lakes and may contain chemical "hot spots" of higher contaminant levels. Legacy PBT chemicals have declined significantly in this species over the last 50 years, but this decline may be reaching a plateau or even reversing in some instances. This is consistent with observations in other piscivorous bird species such as herring gulls (see Section 4.1.4) and common loons.

4.1.6 Whole Lake Trout

A number of programs have been in place since the 1970s to monitor spatial and temporal trends of chemical contaminants in Great Lakes lake trout. According to data collected by the U.S. EPA's Great Lakes Fish Monitoring and Surveillance Program (GLFMSP) and Environment Canada's Fish Contaminants Monitoring and Surveillance Program (FCMSP), whole lake trout from Lake Superior are typically less contaminated than those collected from the other Great Lakes (Carlson and Swackhamer, 2006; Bhavsar et al., 2007; 2008; Carlson et al., 2010). For example, PCBs have declined in top predator fish (lake trout in Lakes Superior, Michigan, Huron and Ontario and walleye in Lake Erie) across the Great Lakes since the two programs began monitoring in the 1970s. Throughout this time period Lake Superior lake trout were consistently lower in PCBs than the fish from the other Great Lakes (Figure 4-7).

The GLFMSP and the FCMSP have observed significant decreases in concentrations of PCBs, DDT, chlordane and dieldrin in whole lake trout over time, as illustrated for Lake Superior in Table 4-3. Similar long-term results have been found for POPs by MDEQ's Fish Contaminant Monitoring Program (FCMP), Environment Canada's Fish Contaminant Monitoring Program, Ontario Ministry of the Environment's Sport Fish Contaminant Monitoring program (SFCMP), and the Chippewa Ottawa Resource Authority (CORA).

In contrast to many POPs, mercury levels in lake trout from Lake Superior are consistently higher than in those from the other Great Lakes (Bhavsar et al., 2010). Similar observations were made for bald eagle feathers, as discussed above. This trend may be due in part to the presence of local industries such as mining, chlor-alkali production and pulp/paper production.



Source: McGoldrick, 2011; Murphy, 2011
Dashed lines show log-linear regression model if annual change is significantly different from zero ($\alpha = 0.05$).

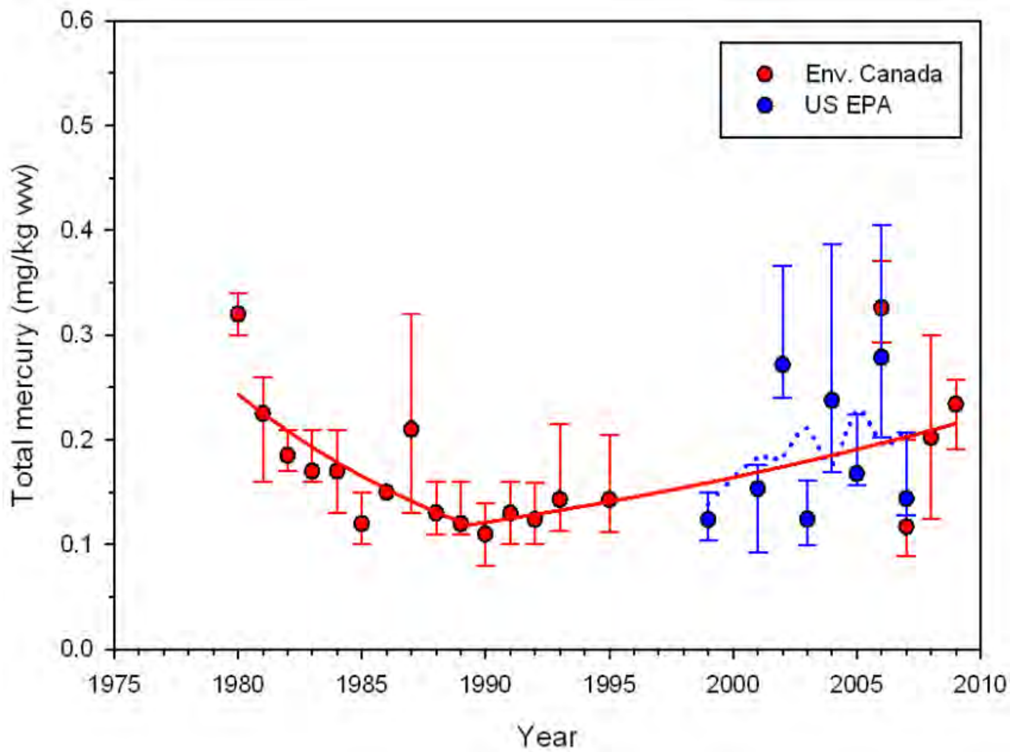
Figure 4-7. Total PCB Concentrations (median & IQR) for Individual (Environment Canada) and Composited (U.S. EPA) Whole Body Lake Trout or Walleye (Lake Erie) Collected from each of the Great Lakes.

Table 4-3. Long-term Rate of Decrease of Contaminants in Whole Lake Superior Lake Trout as Measured by the U.S. EPA and Environment Canada

Contaminant	Trend
Total PCBs	5% long-term annual decline[a]
Total DDT	6.8 % long-term annual decline[b]
Total Chlordane	Consistent decline since EPA ban in 1988. Steady state with no significant increase or decrease[c]
Dieldrin	Consistent decline since monitoring began. Long-term annual decline = 2 – 18%[d]

[a] PCB concentrations remain above the Water Quality Agreement criterion of 0.1 µg/g ww.
[b] DDT concentrations remain below the Water Quality Agreement criterion of 1.0 µg/g ww.
[c] Median chlordane concentration is 0.01 µg/g ww. There is no Water Quality Agreement criterion for this compound.
[d] There is no Water Quality Agreement criterion for this compound.
Sources: McGoldrick, 2011; Murphy, 2011

Certain studies have noted that despite the elevated levels relative to the other Great Lakes, mercury concentrations in Lake Superior lake trout have been declining since the 1970s (Bhavsar et al., 2010). More recent data have revealed that this trend may be reversing. A compilation of mercury data in Lake Superior lake trout collected by the U.S. EPA and Environment Canada (Figure 4-8) indicates that mercury levels began increasing around 1990 (McGoldrick, 2011; Murphy, 2011). This is consistent with another recent study showing a U-shaped mercury trend in Lake Superior lake trout collected near the Apostle Islands (Wisconsin), with a breaking point at 2005 when mercury began to once again increase (Zananski et al., 2011). This pattern has been seen on a broader scale in the Great Lakes Basin and adjacent regions, with mercury in walleye in Ontario lakes and walleye and northern pike in Minnesota lakes shifting to an upward trend in the 1990s (Monson et al. 2011, Monson, 2009).



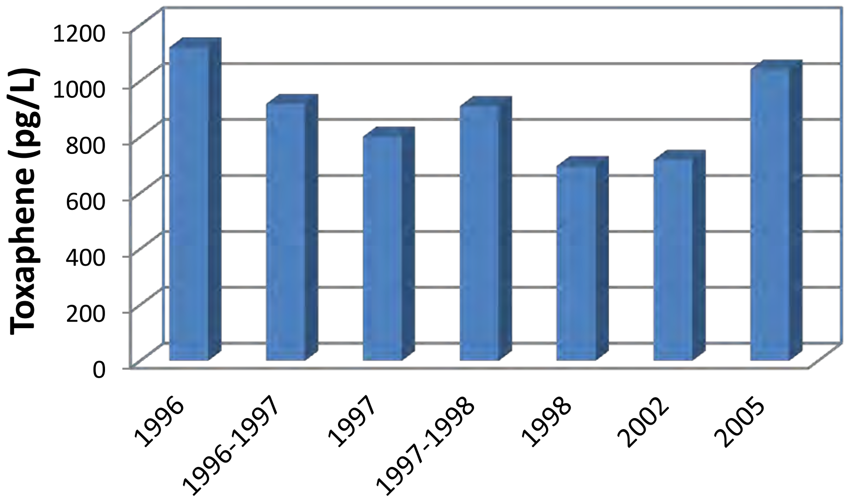
Source: McGoldrick, 2011; Murphy, 2011

Figure 4-8. Temporal Trends of Mercury in Lake Superior Lake Trout Collected by the U.S. EPA and Environment Canada.

4.1.7 Toxaphene

Toxaphene has emerged as a contaminant of great interest in Lake Superior. Although toxaphene is banned in many parts of the world, including the U.S., it can be carried to the Great Lakes region via long-range transport from the southern U.S., where it remains at substantial levels in the environment following years of heavy usage as an insecticide. While most of the ZDDP chemicals tend to be lower in Lake Superior than the other Great Lakes, toxaphene concentrations in the Lake Superior environment and biota often exceed levels found throughout the rest of the Great Lakes region. Decreases in toxaphene concentrations have been observed throughout the Great Lakes in all media following its ban in the mid-1980s.

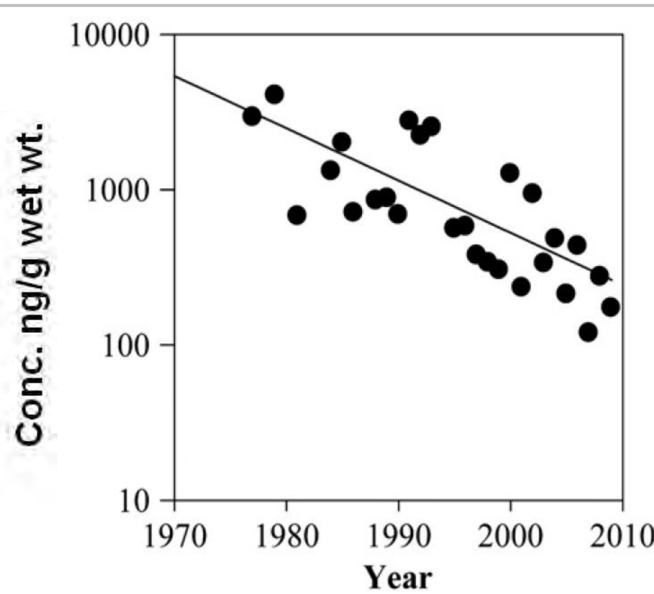
Toxaphene concentrations in the Great Lakes peaked in the late 1970s and early 1980s. Levels have decreased substantially since the mixture was banned in the early 1980s. Recently, the rate of decline of this chemical in Lake Superior waters has slowed substantially. Toxaphene concentrations in Lake Superior water did not change between 1996 and 2005, according to data collected by Environment Canada (Figure 4-9). The small differences seen between years can be attributed to analytical variability for this difficult to measure chemical mixture.



Sources: Jantunen, 2011; Jantunen and Bidleman, 2003; Swackhamer et al,. 1999; James et al., 2001

Figure 4-9. Toxaphene Concentrations in Lake Superior Water (pg/L).

A recent study on toxaphene trends in Great Lakes fish show that concentrations remain the highest in Lake Superior (up to ~480 ng/g) and lowest in Lake Erie (up to ~50 ng/g) (Xia et al., 2012). Concentrations of toxaphene in Lake Superior lake trout continue to exhibit exponential temporal declines (Figure 4-10); however, concentrations appear to level off starting in 2007 (Xia et al., 2012). Continued monitoring of toxaphene in top predator fish in the coming years should confirm whether toxaphene concentrations have reached a steady state in Great Lakes fish.



Source: Xia et al., 2012

Figure 4-10. Total Toxaphene in Lake Superior Lake Trout from 1977-2009.

The presence of higher toxaphene concentrations in Lake Superior relative to the lower Great Lakes has been attributed primarily to the physical properties of the lake (i.e., large volume, long residence time, and cold temperatures; Xia et al., 2011). These factors, in combination with the chemical properties of toxaphene (high vapor pressure, high solubility), cause toxaphene to be released more slowly from Lake Superior than the lower Great Lakes (Carlson and Swackhamer, 2006). Further, food web changes in Lake Superior over time may have had an effect on toxaphene concentrations in top predators such as lake trout by affecting bioaccumulation rates and altering trophic structure.

The toxaphene example demonstrates that despite its remote location and relative lack of industrial development, the Lake Superior ecosystem is susceptible to long-range transport of pollutants. As a result, Lake Superior is not always the “cleanest” of the Great Lakes.

4.1.8 Contaminants of Emerging Concern

Much of the current, basin wide, persistent toxic substance data that are reported focus on legacy chemicals that have been restricted through various forms of legislation but continue to be detected in fish (e.g., PCBs). However, programs in both the U.S. and Canada are making efforts to incorporate the monitoring and surveillance of emerging chemicals into their routine work. Chemicals of interest, also known as chemicals of emerging concern (CECs), are identified through scientific studies (e.g., Howard and Muir, 2010), and general screening of annual samples and also through risk assessments by regulatory bodies. As CECs are identified through this process, they will be reported out through the State of the Lakes Ecosystem Conference (SOLEC), particularly those chemicals with established criteria. Environmental Specimen Banks containing tissue samples are a key component of both the U.S. and Canadian monitoring programs, allowing for retrospective analyses of newly identified chemicals of concern to develop long-term trends in the short term.

Fostering collaboration between U.S. and Canadian monitoring programs for various media will be beneficial, especially in times of fiscal restraint. In 2009, an ad-hoc binational group was formed to bring together government representatives and researchers working on identifying new chemicals in the Great Lakes ecosystem with the objective to facilitate best management practices and sharing of information and resources. The group provides a forum for agencies and researchers to seek and provide information on emerging contaminant surveillance, monitoring, chemical methods development, and provides a place to collaborate on similar chemicals, or classes of chemicals, in different media. Collaboration among research in differing media also provides an excellent opportunity for cost sharing, an accelerated rate of discovery, and a validation of results among the Great Lakes research and monitoring community.

Section 4.2 presents a more detailed discussion of levels and trends of certain emerging contaminants of concern in Lake Superior media.

4.1.9 Fish Consumption Advisories

Despite decreasing critical pollutant concentrations in a variety of media, contaminant concentrations remain high enough to prompt fish consumption advisories both within Lake Superior and for inland lakes within the basin. A number of jurisdictions around Lake Superior, including states, provinces and tribal organizations, provide risk-based advice designed to limit human exposure to environmental contaminants through fish consumption. These advisories are especially critical for vulnerable consumers, such as children, women who anticipate bearing children, and frequent consumers, such as sport fishermen, Native Americans, and First Nations.

Jurisdictional differences in fish consumption advisory trigger levels and meal size definitions, in combination with regional variations in contaminant concentrations, result in variations among the advisories issued by each jurisdiction. Table 4-4 outlines some examples of 2011-2012 fish consumption advisories impacting the LSB. These are general guidelines. More restrictive guidelines for individual waterbodies with above average contaminant concentrations also exist. Table 4-4 summarizes only the U.S. consumption advisories for illustrative purposes. While sensitive populations (children and women of childbearing age) often have more restrictive guidelines, blanks in Table 4-4 indicate that there is no separate advice for the sensitive populations.

State fish consumption advice can be found at:
http://water.epa.gov/scitech/swguidance/fishshellfish/fishadvisories/general.cfm#tabs-4.
The Canadian guidelines can be found at:
www.ene.gov.on.ca/environment/en/resources/collection/guide_to_eating_ontario_sport_fish/index.htm

The majority of fish advisories are based on levels of mercury, dioxins, furans and PCBs. In general, other contaminants, such as mirex, toxaphene, and chlordane, are no longer consumption-limiting contaminants, although certain restrictions in Lake Superior fish are still based on toxaphene (OMOE, 2011). The pie charts in Figure 4-11 illustrate the percentage of consumption restrictions caused by each of the contaminants in the four Ontario Great Lakes and their connecting channels and inland locations for 2011-2012. In Lake Superior, consumption advisories issued for fatty species by OMOE (OMOE, 2011) were mainly due to levels of dioxins, furans, PCBs and, in some cases, toxaphene or mercury. Restrictions on inland waters, particularly for northern pike and walleye, were primarily due to mercury.

There is insufficient information to decipher whether and to what degree the nutritional benefits of Great Lakes fish may outweigh the risk of contaminant exposure to consumers (Turyk et al., 2011). Fish advisories continue to be based solely on risk, rather than from a risk-benefit standpoint that simultaneously considers the nutritional benefits of eating fish. However, as part of their advisory, all of the Lake Superior states make qualitative statements documenting the benefits of eating fish.

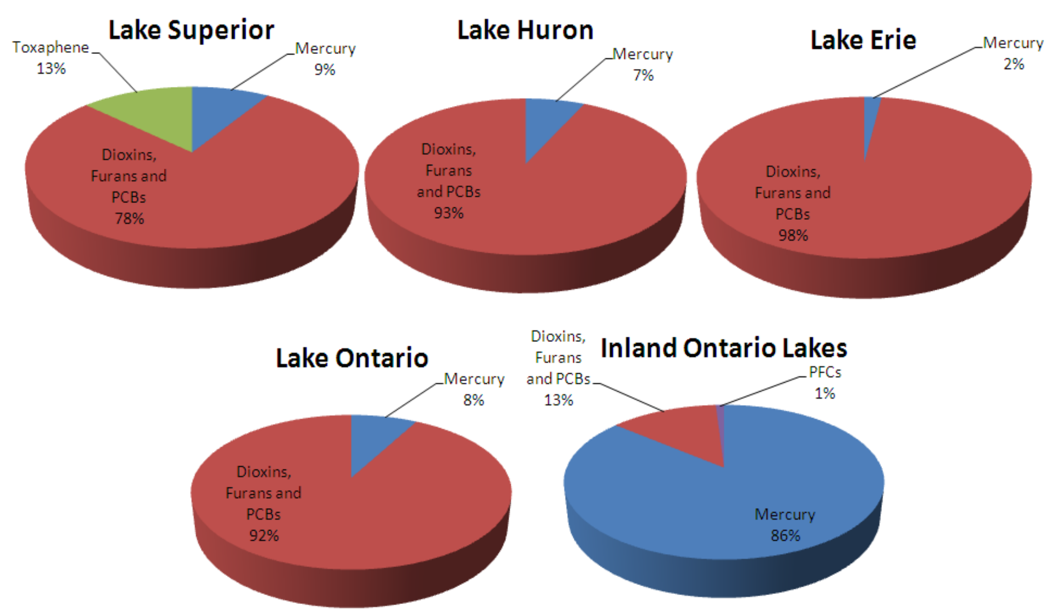
Table 4-4. Select U.S. Fish Consumption Advisories for Lake Superior and Inland Lakes

Agency Issuing Advisory	Waters	Contaminant of Concern	Species	Consumption Advice	
				General Population	Sensitive Population[b]
Wisconsin Department of Natural Resources	Inland Waters of Wisconsin (Exceptions for listed waters, e.g. St. Louis River, not presented here)	Mercury and PCBs	Bluegill, crappie, yellow perch, sunfish, bullhead, trout	Unrestricted	1 meal per week
			Walleye (inland), pike, bass, catfish, and all other species	1 meal per week	1 meal per month
			Muskellunge	1 meal per month	Do not eat
	Lake Superior (PCBs and Mercury are responsible for advice for most species)	Mercury and PCBs	Smelt	Unrestricted	Unrestricted
			Brown trout, Chinook salmon (<30"), chubs, coho, lake herring, lake trout (<22"), lake whitefish, rainbow trout	1 meal per week	1 meal per week
			Chinook salmon (>30"), lake sturgeon (>50"), lake trout (22"-37"), siscowet (<29")	1 meal per month	1 meal per month
			Walleye	1 meal per week	1 meal per month
			Lake Trout (>37")	1 meal per 2 months	1 meal per 2 months
			Siscowet (29"- 36")		
			Siscowet (>36")	Do not eat	Do not eat
Michigan Department of Community Health	Inland Lakes in Michigan (PCB advisories for listed waters, e.g. Torch Lake, not presented here)	Mercury	Crappie (<9"), rock bass (<9"), yellow perch (<9")	Unrestricted	Unrestricted
			Crappie (>9"), largemouth bass, smallmouth bass, muskellunge, northern pike, rock bass (>9"), walleye, yellow perch (>9")	1 meal per week	1 meal per month
	Lake Superior	Mercury	Burbot (<22"), walleye (<22")	Unrestricted	Unrestricted
			Burbot (>22"), walleye (>22")	1 meal per week	1 meal per month
		PCBs	Brown trout, coho salmon, cisco (6-30"), rainbow trout (>26"), suckers, whitefish	Unrestricted	1 meal per week
			Chinook salmon	Unrestricted	1 meal per month
		Mercury, chlordane, PCBs	Lake trout (<26")	Unrestricted	1 meal per week
			Lake trout (26-30")	Unrestricted	1 meal per month
			Lake trout (>30")	1 meal per week	Do not eat
		Chlordane, PCBs, dioxin	Siscowet (14-18")	Unrestricted	1 meal per month
			Siscowet (<18")	Do not eat	Do not eat
Minnesota Department of Health	Inland Lakes of Minnesota	Mercury and/or PCBs	Sunfish, crappie, yellow perch, bullhead	Unrestricted	1 meal per week
			Bass, catfish, walleye (<20"), northern pike (<30") and all other species	1 meal per week	1 meal per month

Table 4-4. Select U.S. Fish Consumption Advisories for Lake Superior and Inland Lakes, Con't.

Agency Issuing Advisory	Waters	Contaminant of Concern	Species	Consumption Advice	
				General Population	General Population
			Walleye (>20"), northern pike (>30"), muskellunge	1 meal per week	Do not eat
	Lake Superior	Mercury and PCBs	Smelt, pink salmon	Unrestricted	-
			Chinook salmon (<30"), coho salmon, lake trout (<23"), rainbow trout, brown trout, lake whitefish, lake herring (cisco)	1 meal per week	-
			Chinook salmon (>30"), lake trout (23-34"), siscowet (<25")	1 meal per month	-
			Lake trout (>34")	1 meal per 2 months	-
			Siscowet (>25")	Do not eat	-

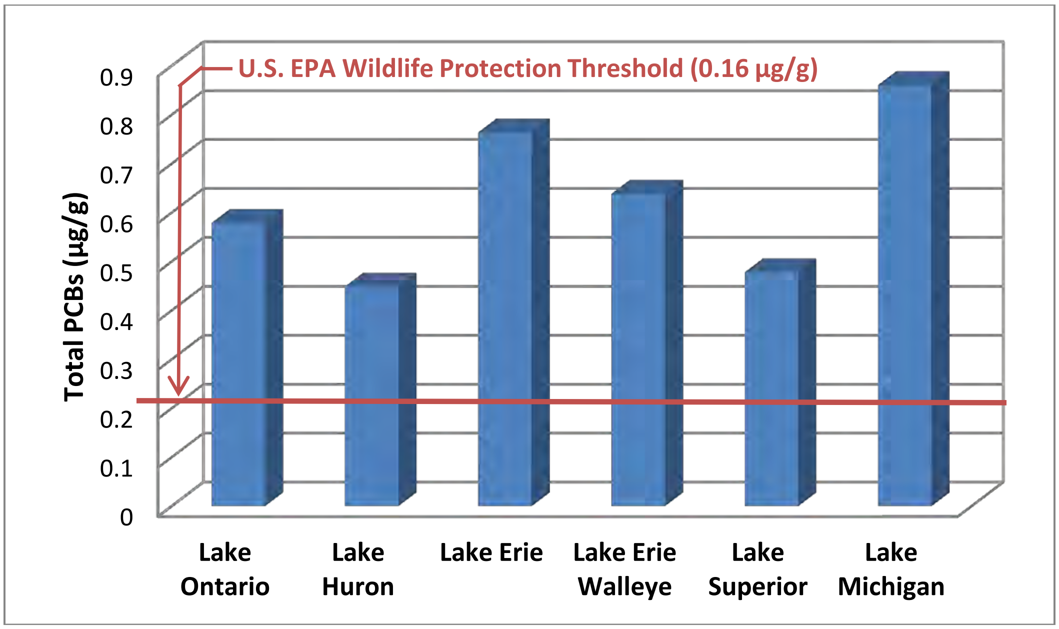
Sources: Wisconsin: WDNR, 2011; Michigan: MDCH, 2011; Minnesota: MDH, 2011a,b



Source: OMOE, 2011

Figure 4-11. Percentage of OMOE Fish Advisories for 2011-2012 Based on Specific Critical Contaminants in the Great Lakes and Inland Lakes of Ontario.

In addition to triggering human fish consumption advisories, chemical concentrations remain high enough to cause negative impacts on wildlife. For example, PCB concentrations measured by the GLFMSP in 2010 exceeded concentrations established by the U.S. EPA to protect the health of fish-dependant wildlife (0.16µg/g) in all five Great Lakes (Figure 4-12). The concentrations exceed the GLWQA PCB concentration objective for whole fish (0.1 µg/g) to an even greater degree.



Source: Murphy, 2011

Figure 4-12. 2010 PCBs in Whole Lake Trout (except Walleye in Lake Erie) Relative to the Established EPA Wildlife Protection Threshold.

Although concentrations of most contaminants are low (as compared to the other Great Lakes) and have decreased over time, they continue to impair the beneficial use goal of unrestricted fish consumption stated in Annex 2 of the GLWQA and may pose a threat to fish-consuming wildlife. Furthermore, because these critical contaminants are declining at slower rates over time, it is likely that their presence will continue to have impacts for future decades.

4.1.10 Human Biomonitoring

A number of large- and small-scale Great Lakes Region human biomonitoring programs have been carried out or are currently underway. Some of the key human biomonitoring studies specific to the LSB or the Great Lakes Basin as a whole that occurred in the last decade are outlined below.

The MDH's Mercury in Newborns in the Lake Superior Basin Study, funded by the U.S. EPA's Great Lakes National Program Office (GLNPO), assessed population-level mercury exposure for residents of the LSB, with a focus on newborn infants. Residual dried blood spots from 1,465 newborns from Minnesota (*n*=1126), Wisconsin (*n*=139), and Michigan (*n*=200) were collected and analyzed for total mercury by the MDH Public Health Laboratory. The amount of mercury found in the newborn bloodspots is indicative of the mothers' mercury exposure during pregnancy. The blood samples were anonymized; meaning, there was no personally identifying information attached to the blood sample. However, information was retained on the baby's sex, month and year of birth, state of residence, and whether the mother lived in an urban or non-urban area (Minnesota only). Most infants were found to have low or undetected total mercury levels. However, 8% of tested newborns had total mercury levels above the RfD for methylmercury (the highly toxic form of mercury found in fish) set by the U.S. EPA. Other significant findings from the study included:

- No relationships were seen between the level of mercury and the baby's sex or urban/non-urban residence.
- Babies born during the summer months were more likely to have an elevated mercury level. This seasonal effect suggests that increased consumption of locally-caught fish during the warm months is an important source of pregnant women's mercury exposure in this region.
- No Michigan samples were above the U.S. EPA RfD, 3% of the Wisconsin and 10% of the Minnesota samples were above this level. One possible explanation is that Minnesotans have reported eating more locally-caught fish than do people in Wisconsin or Michigan.

As a result of the study, MDH is strengthening outreach and communication efforts to health care providers and others to ensure that the public has information that promotes eating fish low in mercury.

Through the GLRI, the Agency for Toxic Substances and Disease Registry (ATSDR) is undertaking a large-scale human biomonitoring project in the Great Lakes Basin. ATSDR has competitively awarded funds to Minnesota, Michigan, and New York health departments to measure environmental toxicant levels in the blood and urine high risk fish consumers who live in the Great Lakes Basin. The purpose of the study is to determine if there are higher levels of contaminants in those people with greater exposure, such as people who eat Great Lakes fish. This information will guide actions that the state health departments take to protect citizens. Ontario, in collaboration with a federal agency, recently conducted a province-wide survey of fish consumption including for the Great Lakes region. This survey is expected to result in a better understanding of the sport fish consumption pattern on the Canadian side of the Great Lakes.

As part of the ATSDR study, the MDH received funding from the U.S. Centers for Disease Control and Prevention in 2010 for population-based contaminant biomonitoring of 500 American Indian adults within the LSB. MDH is collaborating with the Fond du Lac Band of Lake Superior Chippewa Human Services Division to conduct the study. The three-year project is currently in the planning and preparation stage, with recruitment and enrollment taking place in summer 2012. A broad range of contaminants arising from fish consumption as well as historical industrial activities in the region will be measured in blood and urine, including heavy metals (mercury, lead, cadmium), PCBs, mirex, HCB, DDT/DDE, toxaphene and 1-hydroxypyrene. Some contaminants of "emerging concern" are also included (bisphenol A, triclosan, perfluorinated compounds [PFCs]) as well as two nutrients associated with fish consumption (omega-3 fatty acids and selenium). American Indians are a population of concern because they may be more likely to eat fish than other subpopulations in the basin. Findings will be used to develop a data-driven public health action plan to reduce exposure to Great Lakes contaminants through targeted interventions.

During 1999-2000, the Effects on Aboriginals of the Great Lakes (EAGLE) Project was conducted by a partnership between the Assembly of First Nations, Health Canada and First Nations in the Great Lakes Basin to examine the effects of contaminants on the health of the Great Lakes Aboriginal population (Davies and Phil, 2001). The objectives of the Contaminants in Human Tissues Program, a major component of the EAGLE Project, were to determine levels of environmental contaminants in the tissues of First Nations people in the Great Lakes Basin, to correlate these levels with freshwater fish and wild game consumption, and to provide information and advice to First Nations people on the levels of environmental contaminants found in their tissues. Contaminants were tested in hair (n=393) and blood (n=528) from 26 First Nations in the Great Lakes Basin and included over 35 PCB congeners, 34 organochlorine compounds (such as toxaphene, DDE/DDT) and mercury. Some key findings included:

- PCBs, DDE and toxaphene (Congener 50) were the most commonly detected contaminants in blood.
- PCB and mercury levels, but not toxaphene or DDE, were higher in males than females. Although males also had higher consumption rates of freshwater fish and wild game, no significant statistical relationship was found between consumption rates and contaminant levels. Consumption of certain species was correlated with specific contaminant levels (e.g., walleye/pickerel and mercury, rainbow trout and PCBs).
- PCB, DDE and toxaphene levels, but not mercury, increased with increasing age-group.
- Levels of mercury in hair of First Nations people in the Canadian Great Lakes Basin suggest levels have decreased since 1970.
- Most participants had serum PCB and hair mercury levels that were below Health Canada's guidelines (where available) and were not associated with any adverse health effects.

The Wisconsin Department of Health Services (WDHS), in collaboration with a variety of researchers, has conducted human biomonitoring to look at contaminant exposure (e.g., PCBs, DDE, mercury) and associations with health outcomes among frequent consumers of Great Lakes fish. In 2004-2005, WDHS carried out a study of methylmercury exposure among 2000 Wisconsin residents (ages 18 to 92) (Knobeloch et al., 2007). Participants provided hair samples for mercury analysis and completed a survey about their fish consumption habits. The U.S. EPA exposure guideline, which equates to a hair mercury concentration of 1 µg/g, was exceeded in 29% of the hair samples provided by men and 13% of those provided by women. Hair mercury levels were positively correlated with both age and the number of monthly fish meals reported. Mercury was on average eight times higher in fish consumers than non-fish consumers. Despite reporting similar fish consumption rates, men had significantly higher hair mercury levels than women. On average, sportfish consumers had greater hair mercury levels than non-sportfish consumers, with 41% and 29% of men and women, respectively, from this cohort exceeding the 1 µg/g U.S. EPA guideline. It should be noted that this study was carried out statewide, not just within the LSB.

WDHS followed up the 2004-2005 study by advising Wisconsin residents whose hair mercury levels exceeded 1 µg/g to reduce their intake of large, predatory fish (Knobeloch et al., 2011). All study participants were re-contacted in 2008 with the opportunity to fill out a follow-up questionnaire and have their hair mercury levels retested. As a result of the 2005 intervention, residents whose hair mercury levels exceeded 1 µg/g significantly reduced fish intake, with 30% reporting eating smaller or different species of fish. The number of people with hair mercury levels exceeding 1 µg/g fell by over 30%.

The ATSDR Great Lakes Human Health Effects Research Program (GLHHERP), initiated in 1992, is designed to characterize exposure to contaminants via consumption of Great Lakes fish, and to investigate the potential for short- and long-term adverse health effects by providing grants to researchers. The funded research is occurring throughout the Great Lakes Basin, with a number of studies including research within the LSB. Descriptions of the numerous currently funded studies can be obtained through the ATSDR website (http://www.atsdr.cdc.gov/grtlakes/funded-institutions.html).

4.1.11 Summary and Potential Management Implications

- The main source of critical contaminants to Lake Superior is atmospheric deposition, although some local sources exist.
- Long-term monitoring of contaminant concentrations across several media is critical to assessing the health of the Lake Superior ecosystem.
- Concentrations of many legacy PBT contaminants have declined over time, indicating government interventions on the use of these chemicals have been effective.
- The rate of decline of PBT chemical concentrations in various media has slowed, suggesting the system is reaching steady-state in many cases. As a result, further decreases in contaminant concentrations in the region may take many years to become apparent.
- Lake Superior's physical, thermal and biological characteristics make it unique and especially sensitive to retaining PBT contaminants.
- Lake Superior is not always the "cleanest" of the Great Lakes, as is illustrated by examples such as mercury in lake trout, POPs in bald eagle plasma and toxaphene in several biotic and abiotic matrices.
- Many contaminants remain in Lake Superior and surrounding inland waters at concentrations sufficient to trigger fish consumption advisories.
- Because the Lake Superior ecosystem is sensitive to chemical inputs and efficient at retaining environmental contaminants, prevention is critical to its protection.

- The introduction of invasive species to the Lake Superior ecosystem must be stopped. Modifications to the existing food web affect contaminant transport as well as the biology of the lakes.
- To determine current concentrations and establish long-term trends, coordinated physical, chemical and biological monitoring efforts in the Great Lakes must continue under the various binational and domestic programs, such as the Coordinated Science and Monitoring Initiative (CSMI). 2011 was a Lake Superior CSMI year of intensive monitoring. The next CSMI year for Lake Superior is scheduled for 2016.
- Statistical design of monitoring programs and associated analytical methodologies may need to be altered to reflect lower environmental concentrations (i.e., to have greater power to detect small changes in concentrations).
- Analytical method development and chemical risk prioritization are necessary to support detection, monitoring, and regulation of the overwhelming number of emerging contaminants of concern.
- Action is needed beyond the LSB. The ZDDP is critical for the LSB but will have limited impact on PBT chemicals in the Lake Superior environment in the face of long-range transport from regional and global sources.
- There is a need to increase toxicity testing of chemicals of emerging concern in order to support the establishment of appropriate water quality standards, thresholds for the protection of aquatic and fish-consuming wildlife and human fish consumption advisories.
- Advocating for P2, conservation recycling, local and renewable energy sources, and reduced dependence on synthetic chemical substances are ways to ensure a sustainable society and a healthy Lake Superior.

4.2 Chemicals of Emerging Concern

4.2.1 Introduction

The term CEC has come to define the universe of newly detectable chemical substances being discovered in air, water, sediment and wildlife. The term has also come to define chemicals for which a growing body of research points to potential (or "emerging") risks or concerns.

Certain CECs are newly manufactured compounds, only very recently being released into the environment. Others have been in use for longer time periods but have only recently been the target of analysis due to pervious lack of analytical capabilities, restrictive analytical costs, or newly recognized health hazards. Regulations exist for some, but not all, CECs, and the regulations can be quite complex. For example, the manufacture of all PBDEs is banned in Canada and for new uses of octa- and penta-BDE—but not deca-BDE—mixtures in the U.S.; most U.S. work in this arena has been based on industry-led phase-outs. Canada and certain states also further restrict the import, use, and sale of certain PBDE mixtures. In Canada, all commercial, manufacturing, and processing uses of polybrominated biphenyls (PBBs) are banned.

While there are no federal restrictions on PBBs in the U.S., PBBs are no longer known to be produced and placed into the market. The PFC, perfluorooctane sulfonate (PFOS), is included on the list of toxic substances under the Canadian Environmental Protection Act, prohibiting its use, sale and the manufacture. Current uses of PFCs are largely unregulated by the U.S.; however, proposed new uses are prohibited without notice and risk management activities, and long-chain PFC phase-outs are currently underway through a U.S. EPA-industry stewardship and voluntary phase-out project (to be completed in 2015). To note, much like banned pesticides, chemicals with manufacture regulations in the U.S. can still be exported internationally. As a result, a U.S. ban, phase-out or production stoppage has no bearing necessarily on U.S. manufacture for international export(s). Also, manufacturing restrictions do not address chemicals already in commerce, such as articles or products. Such items will be in commerce until the end of the respective product's life.

Improvements in instrumentation and analytical methods, along with reduced sampling and analytical costs, have enabled scientists to detect more substances at lower concentrations than was possible a short time ago. This ability brings with it an emerging concern over the risk these substances may pose to human and ecosystem health and a formidable challenge for environmental scientists, managers, and policy makers.

The sheer number of potential substances for investigation, the difficult human health questions many emerging risks pose, and the challenges in chemical monitoring and surveillance combined with the resources required to investigate and manage a single substance, pose serious research and management challenges.

4.2.2 Three-Part Management Strategy

The Lake Superior LaMP has developed a management strategy for CECs, which can be found in the LaMP 2008 (LSBP, 2008). A summary of the strategy follows:
Three-Part Strategy for Chemicals of Emerging Concern

1. Focus on P2 projects in order to:
    - Look for co-benefits in current reduction programs. Substances of emerging concern may be produced through processes that generate some of the current critical or prevention pollutants.
    - Better utilize and publicize available Emergency Planning and Community Right-to-Know Act (EPCRA) and TSCA data on CECs to inform and improve the quality of P2 efforts.
    - Identify P2 opportunities collaboratively and binationally with stakeholders in the basin or in collaboration with chemistry or toxin reduction programs that focus on preventing or reducing release of a specific substance, a class of substances, specific uses, sectors, modes of action or endpoints.
    - Use P2 as the preferred management approach for all chemicals of concern including critical pollutants and substances of emerging concern. There will be no discrete list of substances for pollution prevention activities.

2. Use the Revised Management Goal Flow Chart (see Figure 4-2 in LaMP 2008) to:
   - Identify the five LSBP management categories and the process for assigning substances to each of them (Tables 4-5, 4-6 and 4-7 in LaMP 2008).
   - Identify a discrete list of substances for which monitoring or use data are lacking.
   - Recognize pollutants which are of special concern due to concentrations that exceed yardsticks (the current critical pollutants).
   - Identify, in conjunction with stakeholder input, additional critical pollutants.

3. Report on CECs
   - Adding a new section to the critical pollutants chapter of the LaMP to report on substances of emerging concern will:
     - highlight monitoring needs and the state of science in Lake Superior basin.
     - provide a record of relevant pollution prevention activities.
     - create awareness about outreach activities for these substances.
     - provide a forum for tracking reductions.
     - promote investigation of alternatives to these substances.
     - identify sources of substances of emerging concern in the Lake Superior watershed.

4.2.3 Levels of CECs in the Lake Superior Environment

CECs have been detected throughout the Lake Superior ecosystem. Most studies to date have focused on two classes of chemicals: brominated flame retardants, consisting of PBDEs and PBBs and PFCs, consisting of PFOS and perfluorooctanoic acid (PFOA), and a suite of 11 to 12 other PFCs. Pharmaceuticals and personal care products (PPCP) are another important group of chemicals of emerging concern, but there is currently insufficient information on their presence in the Lake Superior ecosystem.

The following is an overview of available information on the levels and trends of these contaminants in a variety of media in the LSB. Since the available information is limited, detailed interpretation of levels and trends data is difficult in many cases and determination of even short-term trends is often not possible.

Air

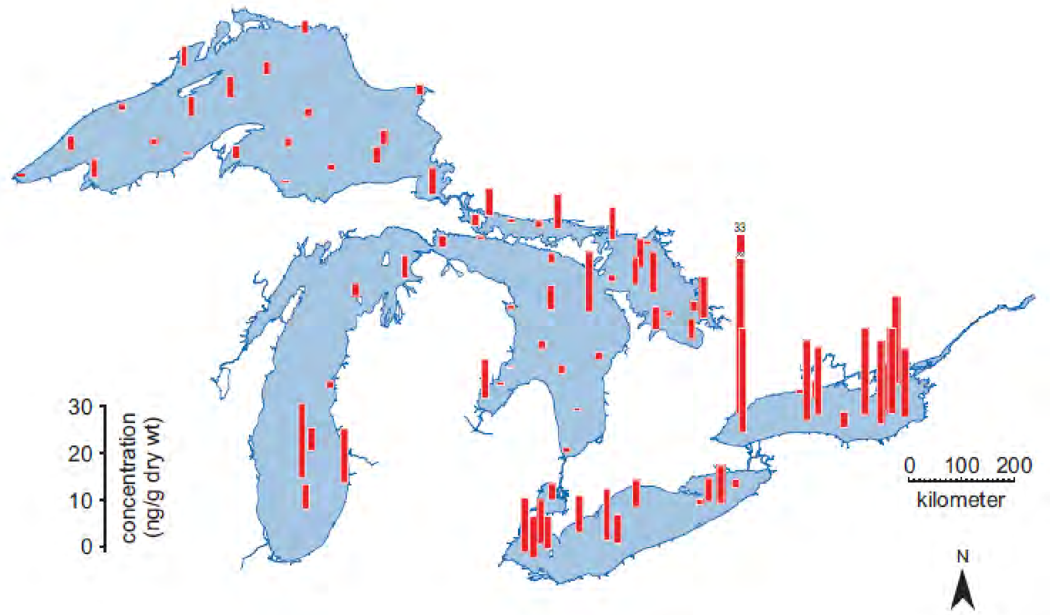
A major vector for PBDEs to enter the environment is through the atmosphere. PBDEs have been found in all air samples collected by the IADN, including those collected at the remote Eagle Harbor site on Lake Superior. The fact that these compounds were found at Eagle Harbor demonstrates that they are widespread and can be transported in the atmosphere to remote locations (Strandberg et al., 2001). Within the Great Lakes Basin, PBDEs were highest at the urban sampling sites of Chicago and Cleveland and lowest at Eagle Harbor. Spatial trends and levels of PBDEs in the Lake Superior atmosphere are similar to those observed for PCBs.

Temporal trends in air vary by PBDE congener. BDE 47 and 99, components of the commercial penta-BDE product, decreased rapidly between 2003 and 2006, reflecting decreased production of this chemical mixture in North America since 2004. In contrast, BDE 209, a component of the deca-BDE mixture, has not decreased at any IADN sampling site throughout the Great Lakes. This reflects the continued use of this commercial product, which is not yet regulated in the U.S. (Venier and Hites, 2008).

Water
PBDEs released into the atmosphere from manufacturing, landfills, and e-waste recycling facilities condense onto particulates and subsequently enter waterbodies. They may also enter the water directly from waste water treatment facilities (Hale et al., 2008) and as leachate from leaking landfills (Kim et al., 2006).

PFCs have been measured in Lake Superior surface waters. Mean PFOS and PFOA in Lake Superior water samples collected between 2002 and 2005 were lower than the concentrations measured in Lakes Ontario, Huron or Erie (Furdui et al., 2008). Between 2001 and 2005, PFOA concentrations ranged from 0.07 to 1.2 ng/L in Lake Superior surface waters and were generally 1.5- to 2-fold greater than PFOS concentrations (Scott et al., 2010).

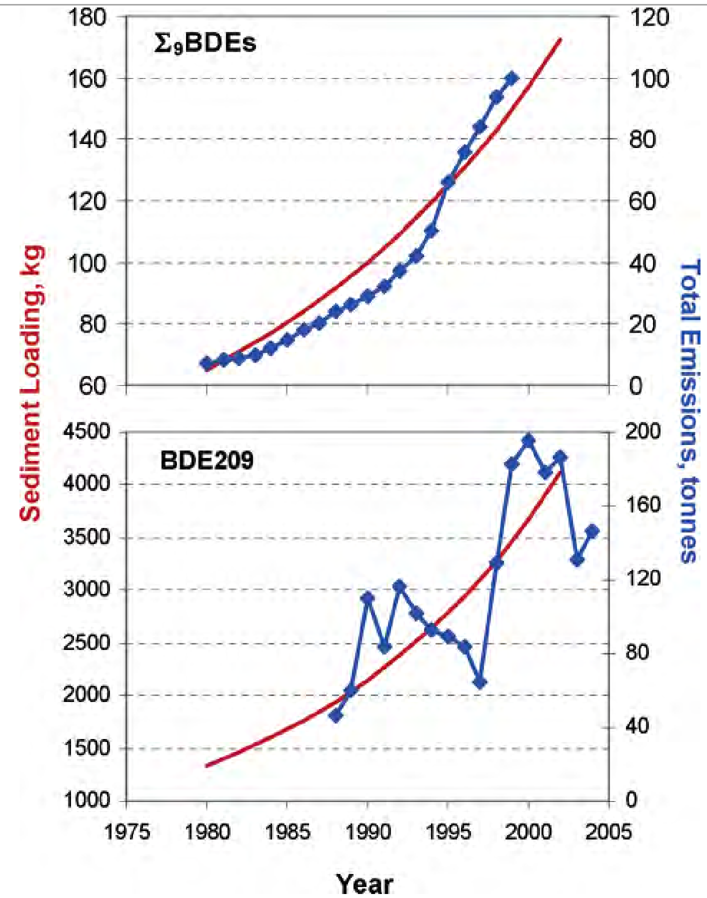
Sediment
PBDEs were assessed in sediment cores taken from Lake Superior at six locations during 2001 and 2002 (Song et al. 2004). Lake Superior's total PBDE load was estimated to be 2 to 6 metric tons, with a current loading rate of approximately 80 to 160 kg/yr. Despite its large surface area, Lake Superior had the lowest PBDE total and annual load of any of the Great Lakes. Studies by Environment Canada confirm that PBDEs levels in sediments are lowest in Lake Superior relative to the other Great Lakes, as shown in Figure 4-13 (Environment Canada and Ontario Ministry of the Environment, 2009a). This is likely due to patterns of urbanization and long-range airborne transport and the effects of lake characteristics on residence time.



Source: Environment Canada and Ontario Ministry of the Environment, 2009a

Figure 4-13. PBDE Concentrations in Surficial Sediment in Open Water Areas of the Great Lakes.

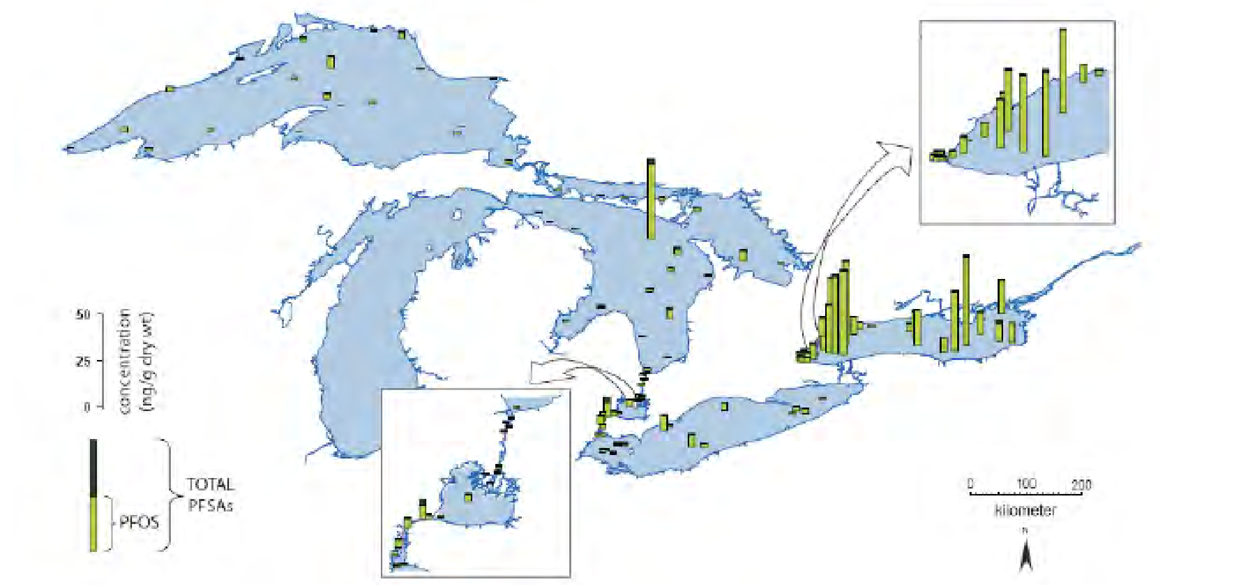
In contrast to PCBs, which showed declining trends or leveling trends, PBDEs increased in Lake Superior sediments in recent years. From the 1970s through 2002, PBDE fluxes into the sediments of all of the Great Lakes have increased exponentially (Li et al., 2006), correlating with PBDE emissions patterns in the Great Lakes Basin (Figure 4-14).



Source: Li et al., 2006

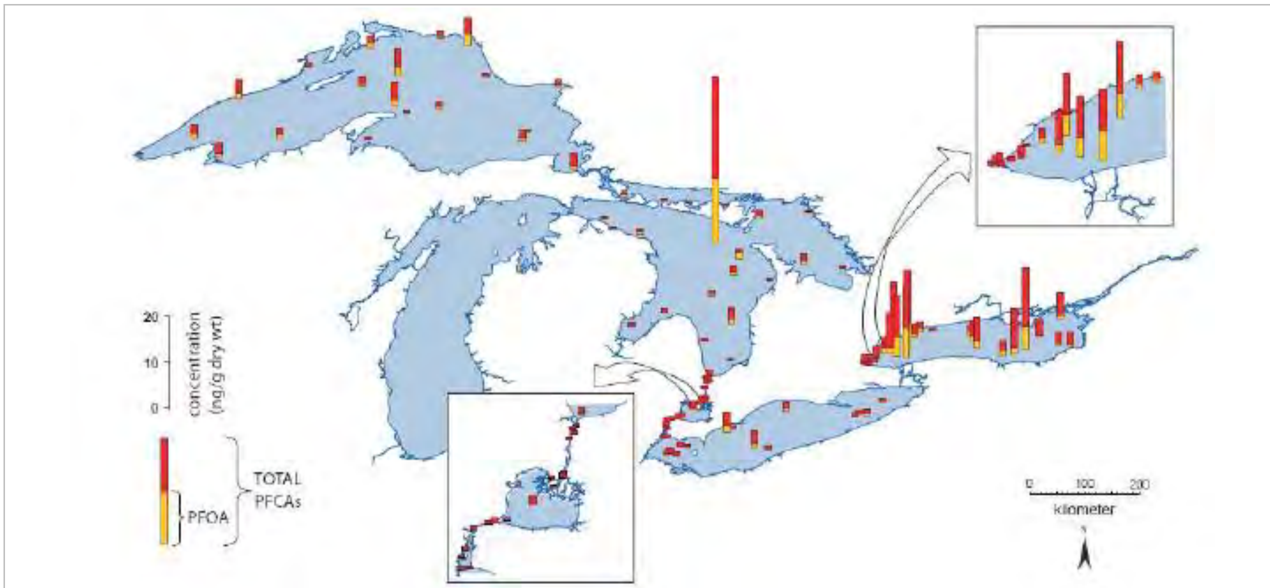
Figure 4-14. Sediment Loading and Emissions of Total PBDEs (excluding BDE 209) and BDE 209 for the Entire Great Lakes Basin.

Environment Canada and Ontario Ministry of the Environment (2009b) also measured levels of perfluorosulfonates (PFSAs), including PFOS, and perfluorocarboxylates (PFCAs), including PFOA, in surficial sediments in open waters of the Great Lakes (Figures 4-15 and 4-16, respectively). As was seen with PBDEs, both PFSAs and PFCAs were highest in the more urbanized and industrialized lower Great Lakes and lower in Lakes Superior and Huron.



Source: Environment Canada and Ontario Ministry of the Environment, 2009b

Figure 4-15. Total Perfluorosulfonates (PFSAs) and Perfluorooctane Sulfonate (PFOS) Concentrations in Surficial Sediments in Open Water Areas of the Great Lakes (excluding Lake Michigan).

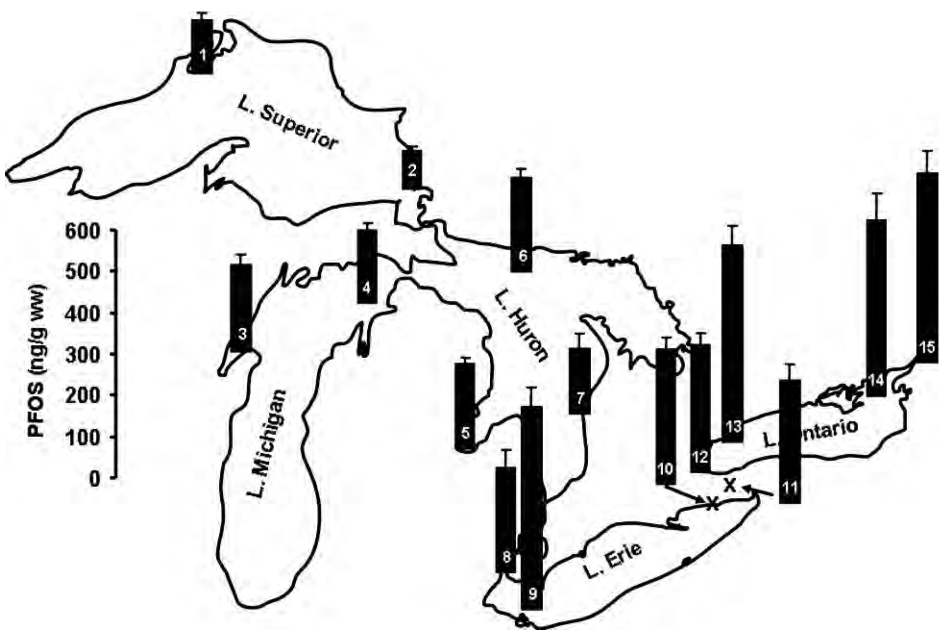


Source: Environment Canada and Ontario Ministry of the Environment, 2009b

Figure 4-16. Total Perfluorocarboxylates (PFCAs) and Perfluorooctanoic Acid (PFOA) Concentrations in Surficial Sediments in Open Water Areas of the Great Lakes (excluding Lake Michigan).

<u>*Herring Gull Eggs*</u>
Lower brominated PBDE congeners have stabilized or declined in herring gull eggs across the Great Lakes. These compounds increased from 1982 to 2000, but showed no significant increase between 2000 and 2006. In contrast, higher brominated congeners, especially BDE 209, appear to be increasing in recent years (Gauthier et al., 2008). This is similar to the patterns that have been observed in air samples collected by IADN (Vernier and Hites, 2008) and reflects the fact that lower molecular weight PBDE mixtures (e.g., penta- and octa-BDE) have been phased out or banned in most industrialized countries, while higher molecular weight mixtures (e.g., deca-BDE) are still in production. This is another example of environmental media responding to anthropogenic use patterns, as has been seen for a number of legacy POPs.

PFCs have also been quantified in Great Lakes herring gull eggs collected in 2007. Of the 15 colonies tested, PFOS concentrations were lowest at the two Lake Superior sites and generally increased from the northwest to the southeast across the Great Lakes (Figure 4-17; Gebbink et al., 2009).



Source: Gebbink et al., 2009

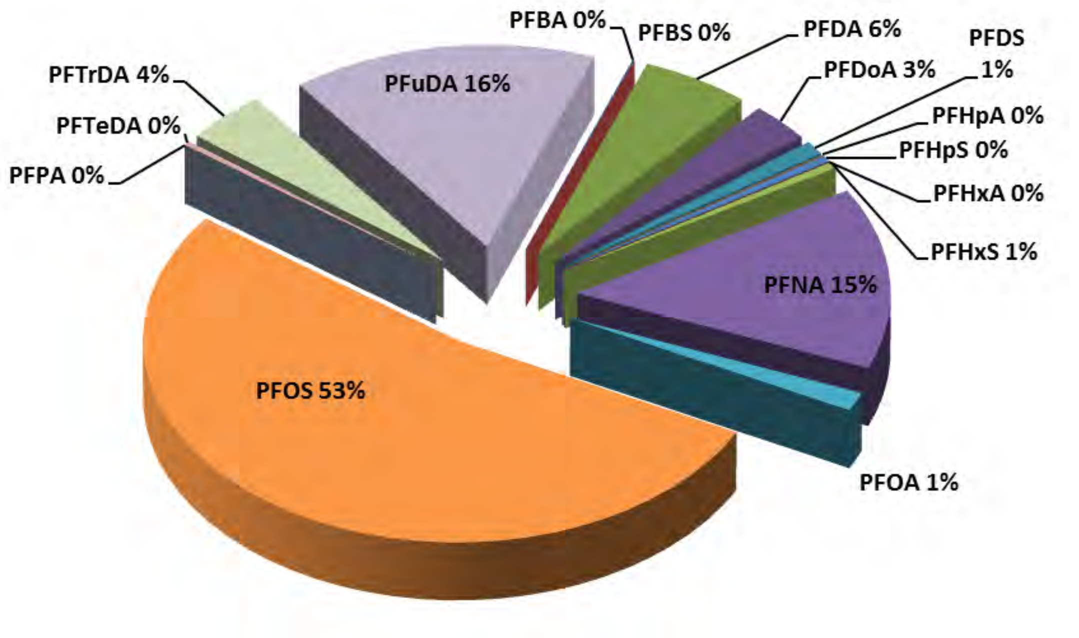
Figure 4-17. Mean PFOS Concentrations (ng/g) in Herring Gull Eggs Collected in 2007 from 15 Colonies in the Laurentian Great Lakes.

<u>*Bald Eagles*</u>
Total PBDEs were detected at a geometric mean concentration of 7.9 ng/g in five bald eagle nestling blood plasma samples collected from the Wisconsin shores of Lake Superior in 2000-2001 (Dykstra et al., 2005). A more recent study found mean PBDE levels in eaglet plasma

collected in 2005 ranging from 0.35 to 29.3 ng/g (arithmetic mean) at three Lake Superior sites (Venier et al., 2010c). These values represented both the highest and lowest values obtained among 15 sites throughout the basins of Lakes Superior, Huron and Michigan, although the mean of the three Lake Superior samples (11.3 ng/g) is similar to that found by Dykstra et al. (2005).

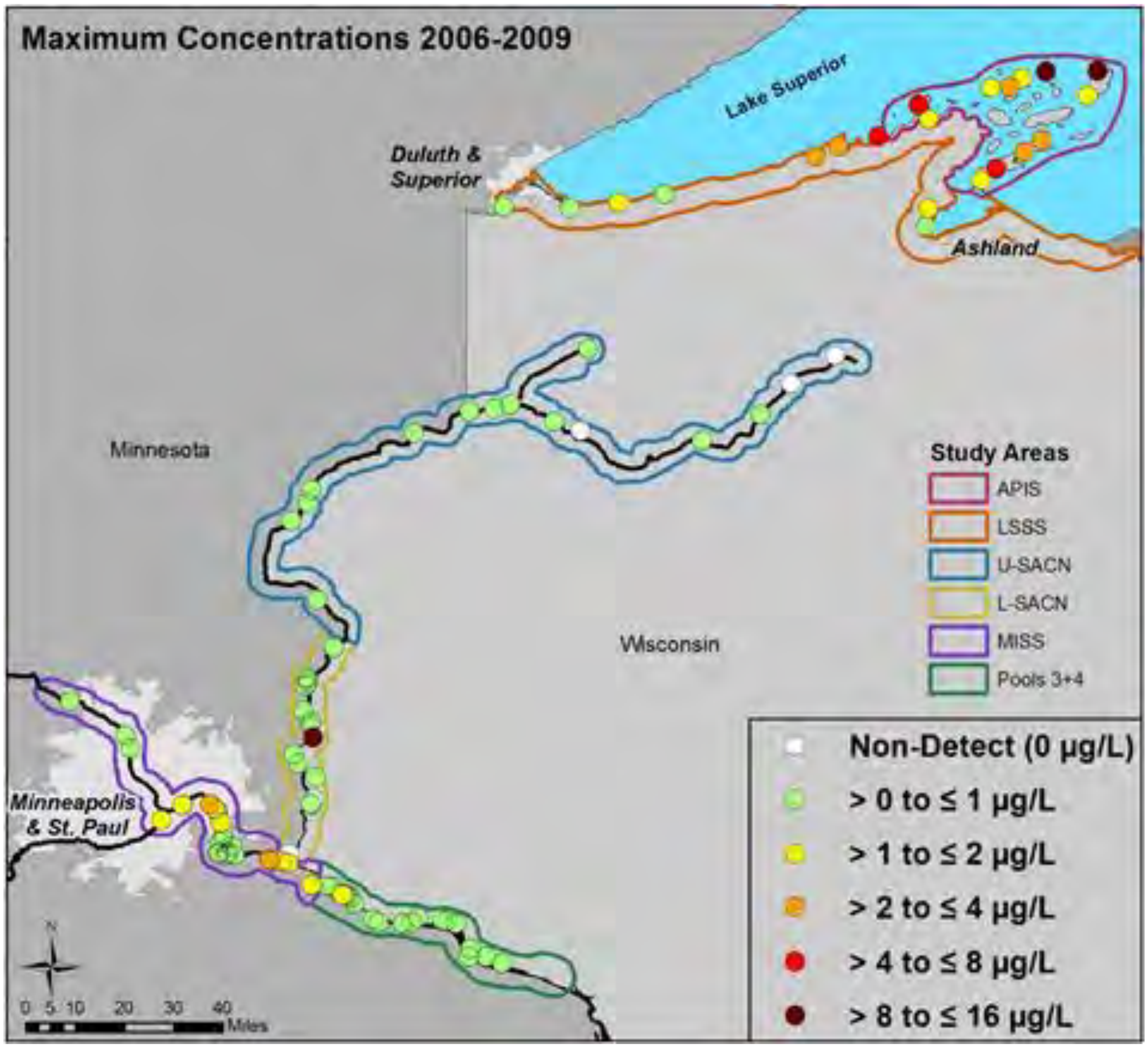
One of the largest studies of emerging contaminants in bald eagles is an ongoing monitoring program by the U.S. NPS Great Lakes Inventory and Monitoring Network. The most recent data published by NPS includes a four-year period from 2006 through 2009, when 154 nestlings were sampled for nine PBDE congeners and 16 PFCs, including PFOS and PFOA (Route et al., 2011). PBDEs and PFCs were found in all nestlings sampled. Geometric mean levels of PBDEs in Lake Superior nestlings ranged from 18 µg/L to 6.47 µg /L over the four years. Lake Superior eagles had significantly higher levels of PBDE congeners #99, #100, #153, and #154 than eagles sampled in some inland study areas. More recent unpublished data show penta- and octa-PBDEs increased through about 2006 in Lake Superior eagles and have steadily declined through 2011 (Route, 2011).

By volume, Route et al. (2011) found PFOS made up 53% of the PFC load in eaglets sampled on Lake Superior followed by PFuDA (16%), PFNA (15%), and PFDA (6%) (Figure 4-18). The remaining PFCs each made up <5% of the total. PFOA, a PFC of considerable concern globally, made up 1% of the PFC load in Lake Superior eagles. However, compared to inland study areas, PFOA was significantly higher in eaglets on Lake Superior, and were particularly high on outer islands of the Apostle Islands (Figure 4-19). The exact reason for this pattern of distribution needs more investigation. NPS is continuing to collect samples to further investigate these trends.



Source: Route et al., 2011

Figure 4-18. Percent by Volume of 16 Different PFC Analytes in Bald Eagle Nestling Plasma Sampled on the Wisconsin Shore of Lake Superior in 2008 and 2009.



Source: Route et al., 2011

Figure 4-19. Maximum Concentrations of PFOA Found in Plasma of Bald Eagle Nestlings in Six Study Areas in the Upper Midwest, 2006-2009.

Lake Trout

The production and use of three popular commercial formulations of PBDE have or are being voluntarily phased out by industry in North America. The phase out of the more toxic penta- and octa-BDE compounds started in 2004 and, by 2012, the use of deca-BDE formulations will also cease. In a national survey of PBDE concentrations in top predator fish from lakes across Canada, the highest concentrations were observed in fish from the Great Lakes and >95% of the PBDE compounds in the fish were tetra-, penta-, or hexa-BDEs (Gewurtz et al., 2011). Federal Environmental Quality Guidelines (FEQG) have been developed by Environment Canada for these three homologue groups which are meant to provide targets for acceptable environmental quality, assess the significance of observed concentrations, and to measure the success of risk management activities. The FEQGs to protect wildlife consumers of fish for tetra-, penta- and hexa-BDEs are 88, 1.0, and 420 ng/g ww, respectively.

Routine monitoring of PBDEs in whole top predator fish from the Great Lakes combined with retrospective analyses of archived samples by the U.S. EPA (Zhu and Hites, 2004) and Environment Canada have provided a complete picture of PBDE contamination in Great Lakes fish from 1977 to the present day. Concentrations of PBDEs in lake trout and walleye rose continuously through to the early 2000s then began to decline for penta-BDE. PBDE concentrations in Lakes Superior and Erie appear to be declining as the slopes of the regressions are all negative; however, the slopes are not significantly different from zero at $\alpha = 0.05$ with a power of 80%. The majority of tetra-BDE and all hexa-BDE concentrations reported for lake trout and walleye in 2009 from all the Great Lakes are below Environment Canada's FEQGs; however, all measured penta-BDE concentrations are well above the FEQG of 1.0 ng/g ww.

Two classes of brominated flame retardants, PBDEs and PBBs, were measured in composite samples of lake trout collected in 1997 from Lakes Superior, Erie, Huron, and Ontario (Luross et al., 2002). The study found that concentrations of PBBs were lowest in Lake Superior, but PBDEs were second highest in Lake Superior lake trout, lower only than Lake Ontario. A similar ranking of PBDE concentrations among lake trout from the Great Lakes was observed by the U.S. EPA Great Lakes Fish Monitoring and Surveillance Program (Carlson and Swackhamer, 2006).

PFCs were also measured in lake trout from the Great Lakes collected in 2001. Mean PFOS concentrations in fish were lowest in Lake Superior (5 ng/g) and were nearly 25-fold lower than lake trout from Lake Erie (121 ng/g), which had the highest fish tissue concentrations (Furdui et al., 2007).

<u>*Humans*</u>

Levels of major PBDE congeners in serum were assessed in a cohort of Great Lakes residents in 1994-1995 and again in the same individuals in 2001-2003 and 2004-2005 (Turyk et al., 2010). Total PBDEs increased 69% in serum over this time frame. Unlike many contaminants (e.g., mercury, PCBs, DDT, toxaphene), PBDE levels were not associated with the consumption of Great Lakes sport fish. Relative concentrations of individual PBDE congeners also shifted over time, potentially due to differences in persistence of the congeners or to changes in exposure associated with the phase out of the penta-BDE, but not deca-BDE, commercial product.

Chapter 5. Reduction Strategies

5.1 Previous Reduction Strategies

Critical chemical reduction strategies exist in several previous LaMP documents and can be summarized as follows:

- The **1991 Lake Superior Binational Agreement** (Appendix A.1) identified three approaches to zero discharge and zero emission, including special designations, P2, and controls and regulations. The agreement included some very specific strategies, many of which have been implemented.
- During development of the **LaMP Stage 2** load reduction schedules released in 1999, the Lake Superior Task Force developed the first set of guiding principles for targeting reductions of critical chemicals, which were revised and updated in the 2005 Milestones report (Appendix A.2)
- The **LaMP 2000** report was the first document to compare the 1990 baseline to a milestone year. The document also identified 22 reduction strategies and government agencies committed to activities within the strategies at either high or medium priority. This report is available for downloading at http://www.epa.gov/greatlakes/lakesuperior/index.html.
- The **2005 Milestones report** (available at the U.S. EPA website above) revised the strategies and, for the first time, actions to reduce CECs were discussed.
- **LaMP 2008** (also available at the U.S. EPA website above) identified 27 additional reduction and inventory activities in its Addendum 4C. In some cases, activities were specific to certain jurisdictions. The Appendix B reports in this 2010 milestones document use the same framework as Addendum 4C. LaMP 2008 also includes a *Three-Part Strategy for Chemicals of Emerging Concern* (see Section 4.2 of this report).

For this 2010 milestones document, chemical inventory and/or reduction strategies are provided below: in Section 5.2, recommendations are from the Canadian and U.S. contractors who updated the discharge and emission inventories; in Section 5.3, strategies were derived from the Appendix B reports for all jurisdictions and Chapter 3. The following strategies are not intended to replace the strategies identified in previous LaMP documents, but to augment them.

5.2 Inventory Improvement – Suggestions from Canadian and U.S. Consultant Reports

5.2.1 Canada

Mercury

- The recycling rate and fate of an estimated 44 kg Hg/yr from discarded mercury relays and 33 kg/yr from instrumentation and control equipment in industrial and commercial facilities is unknown. A systematic process for monitoring the waste stream for mercury-containing equipment used by industrial, commercial, and institutional facilities in the LSB is needed. Environment Canada should consider reinstituting the Mercury Recovery Program initiated in 2005 to assess whether mercury-containing equipment is being put into landfills and how to increase reuse and recycling. The information gathered in undertaking such a project will assist in providing a more accurate estimate of the fate of mercury in these products once they are discarded.

- Although the majority of mercury from consumer products has been accounted for in this inventory, Environment Canada may wish to estimate mercury releases from pharmaceuticals, reagents, and miscellaneous electronics such as pressure transducers, films, scanning electrodes and other products used in the LSB and include them in the inventory.
- Environment Canada should consider contacting waste service providers and agencies that collect mercury containing consumer products in the LSB on a yearly basis to get information on the total waste collected, and quantities of mercury-containing products, pesticides, and PCBs. This will ensure that a more accurate record of such wastes is documented. Alternatively, or in addition, Environment Canada could consider contacting Stewardship Ontario to obtain further information on hazardous and special waste collected and removed yearly from the Basin by the existing municipalities of the LSB.
- Environment Canada should consider contacting representatives responsible for the Federal Contaminated Sites Inventory and the Ontario Ministry of the Environment to obtain more specific information about contaminated sites in the LSB.

Dioxin

- Environment Canada should consider updating the industrial emission data obtained from NPRI once the 2010 NPRI data are available.
- Environment Canada should consider gathering information on the extent to which land clearing and brush burning operations exist in the LSB. If quantities of wood burned become available, existing forest fire emission factors could be used to estimate dioxin and furan emissions from this source.
- Outdoor wood furnaces is a recently identified potential source of dioxins and furans (and possibly HCB) emissions. A survey was conducted in the Province of Ontario to better understand the prevalence of such units in Ontario. Some of the municipalities in the LSB were included in the survey but the data have not yet been made public. Additional information on the extent to which this activity is practiced in the LSB and the content and quantity of the material burned is needed.
- Environment Canada should consider obtaining the results of additional sediment sampling that has been conducted recently in Black Bird Creek System when they become available over the next few months.

Pesticides

- Environment Canada may wish to contact the Thunder Bay Landfill, waste service providers operating in the LSB, and OMAFRA on a yearly basis to request information on quantities of amalgamated pesticides waste collected. This will ensure that a more accurate record of such wastes is documented in the future.

Small and Medium Facilities

- Emissions from larger industrial facilities meeting the NPRI reporting threshold requirements have been included in the emissions inventory. These are known as point sources. However, emissions from facilities that emit substances below reporting thresholds, such as small and medium enterprises (also known as area sources), are not reported or documented. This is a data gap. Environment Canada should develop methods for identifying and estimate ZDDP substances emitted from this source.

5.2.2 United States

- Move the year in which the inventory is prepared back at least one year, preferably two, so that actual data for that year are more readily available.
- Add "structural" (building) fires as a source of emissions; NEI has some data available but not for the chemicals of concern. This would seem a likely source of mercury and especially dioxins, PCBs and HCBs as products of incomplete combustion when chlorine-containing materials are burned, especially in poor combustion conditions.
- For significant sources that tend to vary from year to year (e.g., taconite processing), consider including average releases/year for past 5 years.
- Information needs to be acquired on HCBs from pesticides and past use as a fungicide in seed covering.
- Obtain more information regarding the PCB report program ("permit compliance system") and data added to the inventory in 2010 to ensure data are interpreted correctly.

5.3 Inventory and Reduction Strategies from Chapter 3 and Appendix B

The following inventory and reduction activities were gleaned from the evaluation of progress towards zero discharge and zero emission in Chapter 3 and the 2010 Appendix B tables. These activities are presented as recommendations to the agencies that implement the LSBP. In the following, "LaMP agencies" refers to the various federal, state, provincial, and tribal agencies that participate in the LaMP and which develop policy and implement programs to protect the environment and public health, both through regulations and by promoting voluntary actions. It is recommended that the Lake Superior Binational Task Force consider which of these activities will be implemented by their agencies. Additionally, it is recognized that some reduction strategies may be best carried out by LaMP agency partners, including local governments, non-governmental organizations, and industry.

5.3.1 Mercury

General

- LaMP agencies should increase the level of public education on mercury toxicity; pathways into fish, wildlife, and humans; and how they can help remove it from the basin.

Mining

- Chemical Committee members should participate in or track the various mining-related work groups, committees, partnerships and forums that take place around the Lake Superior basin, such as the Lake Superior LaMP's Mining Committee, to educate themselves about mining in the basin, to provide input to decision-makers on the basin-wide chemical programs and inventories (i.e., historical and current levels and trends), and to participate in discussions about best mining practices.
- The Chemical Committee, in coordination with other SWG committees and agencies, should track the opening of new mines, expansion of existing mines, reopening of closed mines, and closure of existing mines for inventory purposes so that in 2015, a list will be ready for inventory research.
- LaMP agencies should support activities that seek to reduce mercury emissions from mining through research activities, voluntary reductions, and/or enforcing controls and regulations.

Wastewater Treatment and Water Permitting

- LaMP agencies and partners, including municipalities, should investigate opportunities to remove mercury from the wastewater stream, including through both voluntary and regulatory means (e.g., local ordinances). They should build off existing success stories from around the basin, such as WLSSD, Thunder Bay, Superior, Bayfield, Marquette, and Ishpeming where wastewater treatment plant innovations and toxics reductions have been accomplished.

Utilities

- Canadian LaMP agencies should track the impact of Ontario Regulation 496/07 on mercury emissions from the Thunder Bay Generating Station.
- U.S. LaMP agencies should track the impact of the Mercury and Air Toxics Standards rule on mercury and dioxin emissions from coal-fired power plants in the basin, especially Presque Isle in Marquette, Bay Front in Wisconsin, and Taconite Harbor and Laskin in Minnesota.

Cremation

- Mercury emissions from cremation have increased due to increased cremation activity and quantity of amalgam in the teeth of deceased. Increases of mercury emissions from cremation are expected to continue over the next 15 years (MPCA, 2008) followed by a gradual decline as less amalgam will be present in future generations. LaMP agencies should track mercury emissions from crematoriums, and investigate opportunities to reduce emissions (e.g., removal of mercury fillings, crematoria emission controls, or non-cremation alternatives such as alkaline hydrolysis).

Products

- The Chemical Committee should work with agency hazardous waste disposal programs to characterize the different jurisdictions' policies or rules on fluorescent lamp drum-top crushers and identify points of agreement as well as differences. A consistent policy across the LSB is desirable to ensure that drum- top crushers used in lamp recycling do not release mercury to the atmosphere.

- The LaMP agencies should work with various jurisdictions and partners to promote wide-spread bans, restrictions, and voluntary phase-out of mercury-containing products with households, schools, municipalities, and businesses. Regulatory bans, restrictions and voluntary phase-outs of mercury in various products, combined with availability of mercury-free products, are resulting in many companies ceasing to manufacture or sell mercury-containing products, and corresponding declines in mercury emissions. However, because of varying service lives of products, varying quantities of the mercury-containing products will continue to be used (and/or appear in discarded products) even as these products are being replaced. As older mercury-containing products are discarded and replaced with non-mercury devices, it is expected that emissions from this source will continue to decline accordingly. Despite the restrictions, bans, voluntary phase-out and recycling/waste management activities, some mercury-containing products will still be found in use, storage, or being disposed past the 2020 ZDDP target.
- The Chemical Committee and the Canadian LaMP agencies should work with Stewardship Ontario to help promote the MHSW Program.The Canadian MHSW Program is designed to collect consumer hazardous and special materials so they can be recycled or disposed of safely. The first phase began in July 2008 and included nine materials. The second (consolidated) phase began July 1, 2010 and includes 22 materials (including the original nine).The program is expected to substantially increase the quantity of mercury-containing products recycled in the Canadian portion of the LSB.

5.3.2 Dioxins

- LaMP agencies and local governments should continue to support open burning abatement programs. For example, LaMP agencies should coordinate a basinwide survey of rural residents concerning the amount, frequency, and type of material burned.
- The Chemical Committee should consult with agency solid waste managers to determine the extent that accidental or unplanned landfill fires occur and how they contribute to regional chemical inputs. This includes gaining a better understanding of how they are reported in the different jurisdictions and what steps agencies can take to reduce the frequency and intensity of fires.
- The Chemical Committee should also consult with agency solid waste managers concerning leachate recirculation in landfills located in the Lake Superior watershed to improve understanding of the benefits of recirculation and the potential to convert existing landfills, if appropriate.
- The Chemical Committee should seek expert assistance in interpreting the U.S. land clearing estimate and work with Canadian partners to assure a consistent method.
- The Chemical Committee should seek expert assistance in tracking developments in mobile sources as regulations that affect 2010-2015 emissions from on-road and off-road sources.

- The Chemical Committee should consult with agency inspectors to develop a set of assumptions to apply to the small incinerator emissions since they have remained unchanged in the inventory since 1990 due to a lack of knowledge about trends in this mostly illegal source.
- LaMP agencies should increase the level of public education on dioxins, their toxicity; pathways into fish, wildlife, and humans; and how they can help remove them from the basin.

5.3.3 PCBs

- LaMP agencies and partners should continue to remediate locations of historical PCB contamination, including sediments at designated AOCs.
- The Chemical Committee should continue to track disposal and storage via the Ontario database for PCB storage, the Environment Canada database for PCB disposal and the Minnesota hazardous waste database, and U.S. EPA PCB records for PCB storage and disposal.
- The Chemical Committee should examine the storage and disposal categories trends every 5 years, and produce figures showing the various categories and the total weight of articles and materials removed or stored. As part of this examination, the Chemical Committee should pay particular attention to PCB article/equipment end-of-life considerations, the limits of PCB waste data, and how trends could affect PCB articles and equipment more broadly than current inventories allow.
- The Chemical Committee should identify and show the quantity of the stored PCBs that are destroyed in Canada.
- To the extent possible, progress should be measured by the cumulative total of PCB articles and equipment stored and disposed based on available PCB waste data.
- LaMP agencies and partners should increase the level of public education on PCBs, their toxicity; pathways into fish, wildlife, and humans; and how they can help remove them from the basin.

5.3.4 Pesticides

- LaMP agencies should continue to support existing pesticide collection programs, such as clean sweeps, and should consider expanding collections to additional geographic areas.
- Chemical Committee members should document which agency and local government entities collect and track the types and amounts of pesticides disposed. This includes identifying contacts and requesting their input on LaMP documents that discuss pesticides prior to the 2015 milestone data call.
- LaMP agencies and local governments should consider adopting policies or resolutions using the 2009 *Ontario Pesticides Act: Cosmetic Pesticide Ban Regulations.*

- LaMP agencies and partners should increase the level of public education on pesticides, including their safe and appropriate usage; their toxicity; pathways into fish, wildlife, and humans; and how they can help remove old or unused pesticides from the basin.

5.3.5 CECs

- The Chemical Committee should seek expert assistance in interpreting the 2011 Coordinated Science and Monitoring Initiative data on levels of chemicals of emerging concern in the Lake Superior environment.
- The Chemicals Committee should seek expert assistance in better linking CECs to products, processes and sectors so that voluntary pollution prevention and source reduction projects can be implemented by state, local, tribal and industry partners.
- The Chemicals Committee should work to make chemical substance and risk information on CECs more easily accessible to state, local, tribal, industry and non-governmental organization (NGO) partners, to enhance local CEC efforts that support the LaMP.
- The Chemical Committee should compile information on the type and status of different pharmaceutical collections in the basin, including the *Yellow Jugs Old Drugs* program in Michigan, *Medicine Cabinet Clean Out Day* in Minnesota, the *Take Your Medicine… Back to Your Pharmacy* program in Ontario, the U.S. Drug Enforcement Administration's *Prescription Drug Take-Back* initiative, and other efforts to locate and properly dispose of unwanted medication. Following the information gathering, the Committee should look for opportunities to "twin" successful projects across the basin.
- LaMP agencies should develop policies or programs that assist nursing homes and other health care facilities in proper disposal of unwanted medication.
- LaMP agencies should increase the level of public education on new and emerging chemicals; their potential toxicity; pathways into fish, wildlife, and humans; and how they can help remove them from the basin.

5.3.6 HCB

- Canadian LaMP agencies or their inventory consultant should seek a means to approximate the HCB emissions from the Algoma iron sintering plant in 1990 for baseline inventory purposes so that a percent reduction for HCB for basin sources can be estimated.

5.3.7 Other Inventory and Reduction Strategies

- LaMP agencies should conduct long-term monitoring of contaminant concentrations across several media since it is critical to assessing the health of the Lake Superior ecosystem. This should include the continuation of coordinated monitoring efforts. For example, the Coordinated Science and Monitoring Initiative is a binational, Great Lakes-wide effort between federal and state agencies and other partners designed to address the major priorities for each of the lakes. Comprehensive field monitoring is rotated between the each of the Great Lakes in successive years. For Lake Superior, the year of intensive monitoring just occurred in 2011. In the years approaching 2016, the next year of focused sampling for Lake Superior, LaMP agencies and partners should carefully consider contaminant monitoring needs (e.g., biota, sediments, etc.) as they plan for future monitoring activities. Statistical design of monitoring programs and associated analytical methodologies may need to be altered to have greater power to detect small changes in concentrations.

- LaMP agencies should recognize that because the Lake Superior ecosystem is sensitive to chemical inputs and efficient at retaining environmental contaminants, prevention is critical to its protection.
- LaMP agencies and partners should continue to identify and support various energy efficiency and energy conservation programs (e.g., Leadership in Leadership in Energy and Environmental Design) and provide resources to the public, private businesses, and municipal governments.
- LaMP agencies should promote activities that halt or slow down invasive species, pursuant to the Lake Superior Aquatics Invasives Species Complete Prevention Plan. The invasion of foreign species to the Lake Superior ecosystem must be stopped. Modifications to the existing food web affect contaminant transport as well as the biology of the lake.
- LaMP agencies should recognize that action is needed beyond the LSB. The ZDDP is critical for the LSB but will have limited impact on PBT chemicals in the Lake Superior environment in the face of long-range transport from regional and global sources. LaMP agencies should continue to track and, where possible, participate in out-of-basin chemical reduction activities.
- U.S. LaMP agencies should work with state TMDL programs to reduce ZDDP chemicals statewide and in specific waterbodies that require TMDLs for ZDDP chemicals, including mercury in the St. Louis River. Several states are implementing or developing state-wide TMDLs, including Minnesota (for mercury) and Michigan (for both PCBs and mercury).
- LaMP agencies should increase the level of public education on contaminants. There are many success stories that could be used as examples of how regulations, combined with changes in industry and public behaviors, have helped clean up the Great Lakes. Witness the success of the bald eagle from near extinction to a growing population due largely to taking DDT off the market. The many programs discussed in this document rely on voluntary participation from industry, small municipalities, and the public. A successful public education program is needed to meet the goal of zero discharge and zero emission.

Chapter 6. Conclusions

1990-2000 Retrospective

When the governments and stakeholders contemplated LaMP critical pollutant reduction schedules in the early 1990s, the goal of virtual zero discharge and zero emission was set for 2020, a quarter of a century into the future. With the completion of this 2010 Milestones report, less than one decade remains to meet the 2020 goal. The data provided in this report illustrates many successes – yet more needs to be done. Using the mercury and dioxin examples, reaching the reductions achieved so far (80% and 86%, respectively), may prove to be easier than reducing the remaining 15 to 20% needed to achieve zero discharge and zero emission.

Between 1990 and 2000 many industries in the LSB made changes to their production processes to reduce critical pollutants. The pulp and paper industry, for example, changed to chlorine dioxide and extended oxygen delignification, reducing dioxin to undetectable levels. Two mining operations that released large amounts of mercury and dioxin but were no longer profitable shut down. Permitted processes such as medical waste and sewage sludge incineration were stopped as standards became more difficult to meet and alternatives became more attractive. Manufacturers stopped adding mercury to products, due to a combination of voluntary and regulatory actions. States passed special designations such as the Outstanding International Resource Water. The Great Lakes Water Quality Initiative set tough and consistent water quality standards for BCCs.

In the decade following 2000, Canadian wastewater treatment plants made the transition from advanced primary treatment to secondary treatment and beyond. Provincial and state regulations caused coal-fired power plants operators to investigate and use lower mercury fuels and install mercury control equipment. Local open burning abatement programs increased the pressure to find alternatives to burning. Across the basin, as more residents had access to hazardous waste collections, stockpiles of mercury-containing products and old banned pesticides have dwindled to the point where the remaining stockpiles may be identified primarily during estate preparation. Local utilities took advantage of assistance programs to change-out transformers suspected of containing PCBs. Due to a U.S. EPA Maximum Achievable Control Technology requirement, the most polluting furnaces in the taconite industry were retrofitted with new technology that reduced particulates and had the co-benefit of reducing mercury. A scram mining operation emerged that made use of existing taconite tailings instead of expanded mining. States developed or started development of statewide TMDLs for mercury and PCBs.

Many critical pollutant reduction projects have occurred in all LSB sectors since reduction activities were identified in LaMP 2000. Many of these activities were a direct result of the LaMP, while others were closely aligned with LaMP goals. To this point, the most fruitful of the pollutant reduction methods identified in the Lake Superior binational agreement has been P2. Through P2, the easiest reductions have been achieved; those remaining are more difficult.

2000 to the Present

A majority of the loading of critical pollutants to Lake Superior is coming from out-of-basin sources. LaMP agencies have been effective at reducing critical pollutant loadings within the basin, but further action is needed beyond the basin if virtual elimination in the ecosystem is to become a reality.

Tracking critical pollutant emissions has been more straightforward for pollutants such as mercury and dioxins than for pollutants such as PCBs and pesticides. The amount of direct emission measurements and the quality of emission factors and emission estimates has improved since the publication of LaMP 2000. Despite the improvement, many gaps still exist to accurately and properly characterize emissions from diffuse sources such as landfills, mobile sources, and products containing these chemicals. For all critical chemicals, it is still difficult to estimate the impact of local reduction efforts, such as pesticide clean sweeps, on emissions within the basin. This is because in-service or in-storage equipment and products are not inventoried.

Despite estimates and knowledge gaps that exist within the LSB emissions inventory, reasonable and scientifically valid estimates about critical pollutant reductions within the basin have been made. For instance, it is estimated that mercury discharges and emissions declined 72% by the year 2005 and 80% by the year 2010 (compared to the 1990 baseline). In order to meet the Stage 2 LaMP goal of zero discharge and zero emission by 2020, an additional 417 kg/yr must be reduced. (In order to meet the extrapolated 90% reduction goal by 2015, an additional 204 kg/yr of mercury must be reduced from 2010 loads.) According to the inventory data, the largest remaining emission sources for mercury are mining and fuel combustion (mostly from coal), which together account for 91% of the current mercury emissions within the basin, although mercury from mining emissions is more than twice that from fuel combustion in the LSB. Data from 2010 indicate that mercury emissions from mining have dropped 21% from 2005 levels and fuel combustion has dropped by 43%.

For dioxin, it is estimated that dioxin discharges and emissions declined 82% by the year 2000 and 86% by 2005 compared to the 1990 ZDDP baseline. According to the inventory data, the dioxin decline remains at 86% in 2010. In order to meet the 90% reduction goal by 2015, roughly 1 g TEQ/yr of dioxin must be reduced from 2010 loads. Open burning is a completely preventable source of dioxin and elimination of open burning by 2015 would exceed the goal if all else remained equal. Fuel combustion (mostly from mobile sources) is the second largest source of in-basin dioxin and trends by 2015 are difficult to predict due to changes in pollution control for mobile sources such as diesel engines.

The HCB inventory is problematic since it is incomplete. For the first time since the inventory effort began, the Canadian and U.S. inventories were combined. The largest sources in 2010 were incineration and vehicles. It is not possible to estimate the overall reduction of HCB because an estimate is not available for the largest source identified in 1990 (i.e., the iron sintering plant), although it is likely to be similar to the dioxin reduction of 85% by 2010.

Tracking PCB reductions over time has not been possible without a complete inventory. As an alternative, the Chemical Committee has proposed to track disposal and storage via the Ontario database for PCB storage, the Environment Canada database for PCB disposal and the Minnesota hazardous waste database and U.S. EPA PCB databases for PCB disposal. Storage, disposal, and/or destruction of PCB capacitors and oil will be analyzed every 5 years for trends and cumulative progress. Reductions within the basin should be greater than or equal to state or province-wide trends.

Although the LSB is mostly non-agricultural, a significant amount of banned pesticides have been collected in or near the basin since 1992. Although the LaMP Stage 2 reduction goal was to collect all of the pesticides that contained any of the nine ZDDP chemicals by 2000, it is obvious that these pesticides are still present and that collections need to continue. The positive news is that the rate of disposal of these pesticides has stabilized or declined since 2001. Anecdotal evidence from collections indicate that the banned pesticides coming into the programs are from sources that were recently discovered, usually as part of preparing for an estate sale or other property transfer. The largest known stockpiles appear to have been disposed already.

In each of the milestone years, the LaMP agencies and consultants inventoried a variety of sources, searching for the best data each time to continually update and improve the inventories. However, the program has reached the point that some remaining conservative assumptions about certain discharges and emissions in the inventory are unusable as we continue to track progress towards zero. For example, the inventory estimate for U.S. small incinerators was assumed to be 0.6 g/yr TEQ dioxin in 1990, 2000, 2005 and 2010 since we had no direct evidence of change. This conservative assumption is not critical when the amount is low compared to the rest of the inventory, but in 1990, small incinerators were just 2% of the inventory compared to 15% of the inventory in 2010. LaMP agencies will need to invest in some additional inventory research in order to make more accurate emission estimates in 2015 and 2020.

In general, concentrations of critical pollutants have declined in various compartments of the Lake Superior ecosystem including air, water, sediment, herring gull eggs, and fish. These declines have occurred following government intervention in both the U.S. and Canada to restrict and/or ban the manufacture and use of PCBs and certain pesticides in the 1970s and 1980s; however, declines of most of these banned pollutants have occurred at a much slower rate in recent years due to continued atmospheric inputs. Critical pollutants continue to impair beneficial uses set forth in the GLWQA both locally and lakewide. Concentrations of PCBs, mercury, and other critical pollutants remain above levels that limit consumption of fish from Lake Superior. PCBs, toxaphene, and dieldrin in Lake Superior water remain above the most sensitive water quality yardsticks used by Lake Superior jurisdictions to evaluate water quality. A relatively recent finding is that levels of mercury in Lake Superior fish are slightly higher than in the other Great Lakes. In addition, some Great Lakes fish data indicate an upward trend in fish mercury levels. The elevated level of mercury provides an example of how, despite its remote location and relative lack of industrial development, Lake Superior's unique properties make it particularly susceptible to pollutant inputs.

Recent discoveries of many CECs in the Lake Superior ecosystem have led to an important challenge for lake managers. Chemicals that are used in our everyday lives, such as personal care products and pharmaceuticals, along with specific-use chemicals, such as PBDEs and PFCs, are being detected in various compartments of the Lake Superior ecosystem. Some of these, such as PBDEs, are increasing in concentration in fish and sediments. Because little is known on the potential toxicity and environmental fate and transport of many of these CECs, the management challenge lies in deciding which of them should be defined as chemicals of concern and subsequently monitored and/or remediated. The Chemical Committee advocates using the precautionary approach through pollution prevention measures to limit their release to Lake Superior, along with the understanding that prevention is a more cost effective approach than degradation followed by remediation.

The Path Forward

The ZDDP has documented reductions in emissions of critical chemicals. While it is tempting to point out that the sources within the LSB are small compared to atmospheric deposition from out-of-basin sources, as a demonstration, this program has succeeded and can actually document its success. By proving that people living around Lake Superior have succeeded in reducing sources of toxic chemicals, the ZDDP shows that this can happen elsewhere.

To put the accomplishments of Lake Superior's people into perspective, it is estimated that 2.1 tonnes (2.3 tons) of mercury was released from sources in the basin in 1990 but is down to 0.4 tonnes (0.4 tons) in 2010. Eliminating more than a tonne and a half of mercury is a resounding success -- but the work is not finished. The remaining 0.4 tonnes is still a lot of mercury. Dioxin is following a similar trend as mercury but preventable sources still dominate the inventory. 4.6 tonnes (5 tons) of banned pesticides were collected from just one state since 1992. Ongoing pesticide collections are continuing in Ontario and the other states as well. Stockpiles of PCBs are diminishing but by no means eliminated since the trend has not even flattened.

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List of Acronyms

AMRC	Association of Municipal Recyclers
AOC	Area of Concern (from Great Lakes Water Quality Agreement Annex 2)
ATSDR	Agency for Toxic Substances and Disease Registry
BCC	bioaccumulative chemical of concern
BHC	hexachlorocyclohexane (aka, HCH or benzene hexachloride)
CCME	Canadian Council of Ministers of the Environment
CEC	chemical of emerging concern
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
COA	Canada-Ontario Agreement [Respecting the Great Lakes Ecosystem]
CORA	Chippewa Ottawa Resource Authority
CSMI	Coordinated Science and Monitoring Initiative
DDE	dichlorodiphenyldichloroethylene (metabolite of DDT)
DDT	dichloro diphenyl trichloroethane
EAGLE	Effects on Aboriginals of the Great Lakes
EIA	Energy Information Administration
EPCRA	Emergency Planning and Community Right-to-Know Act, U.S.
FCMP	Fish Contaminant Monitoring Program
FCMSP	Fish Contaminants Monitoring and Surveillance Program
FEQG	Federal Environmental Quality Guideline
GIS	Geographic Information Systems
GLFMSP	Great Lakes Fish Monitoring and Surveillance Program
GLHHERP	Great Lakes Human Health Effects research Program
GLI	Great Lakes [Water Quality] Initiative
GLIFWC	Great Lakes Indian Fish and Wildlife Commission
GLNPO	Great Lakes National Program Office
GLRC	Great Lakes Regional Collaboration, U.S.
GLRI	Great Lakes Restoration Initiative
GLWQA	Great Lakes Water Quality Agreement
HCB	hexachlorobenzene
HCH	hexachlorocyclohexane (aka, BHC or benzene hexachloride)
HHW	household hazardous waste
HL	High level

IADN	Integrated Atmospheric Deposition Network
IJC	International Joint Commission
IMERC	Interstate Mercury Education and Reduction Clearinghouse
LaMP	Lakewide Management Plan (from Great Lakes Water Quality Agreement Annex 2)
LEED	Leadership in Energy and Environmental Design
LL	Low level
LSB	Lake Superior Basin
LSBP	Lake Superior Binational Program
MATS	Mercury and Air Toxics Standards
MDA	Minnesota Department of Agriculture
MDCH	Michigan Department of Community Health
MDEQ	Michigan Department of Environmental Quality
MDH	Minnesota Department of Health
MHSW	Municipal Hazardous and Special Waste
MPCA	Minnesota Pollution Control Agency
MPUC	Minnesota Public Utilities Commission
MW	megawatt
NEI	National Emissions Inventory, U.S.
NPRI	National Pollutant Release Inventory, Canada
NPS	National Parks Service
NWRPC	Northwest Wisconsin Regional Planning Commission
OCS	Octachlorostyrene
OIRW	Outstanding International Resource Water, U.S.
OMAFRA	Ontario Ministry of Agriculture Food, and Rural Affairs
OMOE	Ontario Ministry of the Environment
ORW	Outstanding Resource Waters, Wisconsin
P2	pollution prevention
PAH	polycyclic aromatic hydrocarbons
PBB	polybrominated biphenyl
PBDE	polybrominated diphenyl ether
PBT	persistent, bioaccumulative and toxic chemical
PCB	polychlorinated biphenyl
PCDD	polychlorinated dibenzo-p-dioxin
PCDF	polychlorinated dibenzofuran

PEL	probable effect level
PFC	perfluorinated compound
PFCA	perfluorocarboxylate
PFOA	perfluorooctanoic acid
PFOS	perfluoroalkyl sulfonates
PFSA	perfluorosulfonate
POP	persistent organic pollutant
PPCP	pharmaceuticals and personal care products
PWQO	Provincial Water Quality Objective
RCRA	Resource Conservation and Recovery Act
RfD	Reference Dose
SFCMP	Sport Fish Contaminant Monitoring Program
SOLEC	State of the Lakes Ecosystem Conference
SWG	Superior Work Group
2,4,5-T	2,4,5-trichlorophenoxy-acetic acid
TCDD	total polychlorinated dioxin
TEF	toxic equivalency factor
TEQ	Toxic Equivalence Quotient
TMDL	Total Maximum Daily Load, U.S.
TSCA	Toxic Substances Control Act, U.S.
UNEP	United Nations Environment Programme
U.S. EPA	United States Environmental Protection Agency
WDHS	Wisconsin Department of Health Services
WDNR	Wisconsin Department of Natural Resources
WHO	World Health Organization
WLSSD	Western Lake Superior Sanitary District, Minnesota
ww	wet weight
ZDDP	Zero Discharge Demonstration Program